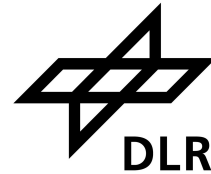




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Master Thesis

Phosphor-Based Temperature-Sensitive Paints for Thermometry: Material Development & Characterization

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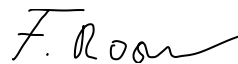
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Steinfurt, 6 May 2026

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Contents

List of Abbreviations	III
1 Introduction	1
2 Theory	2
2.1 Temperature-Sensitive Paints	2
2.2 Luminescence Thermometry	3
2.3 Temperature Readout Methods	5
2.3.1 Intensity-Based Methods	7
2.3.2 Lifetime-Based Methods	8
2.4 Thermal Quenching	9
2.4.1 Thermal Crossover	10
2.4.2 Thermal Ionization	12
2.4.3 Thermally Activated Concentration Quenching	13
2.5 Eu^{2+} -Activated Phosphors	14
3 Experimental Section	17
3.1 Sample Preparation	17
3.1.1 $\text{K}_3\text{LuSi}_2\text{O}_7:\text{Eu}^{2+}$ Phosphor Powder	17
3.1.2 Coated Samples	17
3.2 Measurements	20
3.2.1 X-ray Powder Diffraction	20
3.2.2 Diffuse Reflectance Spectroscopy	20
3.2.3 Photoluminescence Spectroscopy	20
3.2.4 Decay Curve Measurements	21
3.2.5 Temperature-Dependent Spectroscopy	21
3.2.6 X-ray Excited Spectroscopy	22
3.2.7 Particle Size Distribution	23
3.2.8 Further Photometric and Colorimetric Properties	23
3.2.9 Layer Thickness	24
3.2.10 Roughness	24
4 Results and Discussion	25
4.1 $(\text{Sr,Ca})\text{AlSiN}_3:\text{Eu}^{2+}$ Phosphor Powder	25
4.1.1 Luminescence Properties	25
4.1.2 Assessment of Possible Oxygen Quenching	26
4.1.3 Thermal Degradation	27
4.2 $(\text{Sr,Ca})\text{AlSiN}_3:\text{Eu}^{2+}$ Coating	28
4.2.1 Temperature-Dependent Emission Spectra	28

4.2.2	Temperature-Dependent Decay Curves	46
4.2.3	Layer Thickness, Roughness and Coating Quality	65
4.3	K ₃ LuSi ₂ O ₇ :Eu ²⁺ Phosphor Powder	67
4.3.1	X-ray Powder Diffraction	67
4.3.2	Diffuse Reflectance Spectra	69
4.3.3	Excitation Spectra	70
4.3.4	Emission Spectra	71
4.3.5	Temperature-Dependent Emission Spectra	72
4.3.6	Temperature-Dependent Decay Curves	73
4.3.7	Thermal Degradation	75
4.3.8	X-ray Excited Emission Spectra	78
4.3.9	Simplified Energy-Level Scheme of K ₃ LuSi ₂ O ₇ :Eu ^{2+/3+}	79
4.3.10	Further Photometric and Colorimetric Properties	81
4.4	K ₃ LuSi ₂ O ₇ :Eu ²⁺ Coating	83
4.4.1	Possible Improvement of Coating	85
5	Conclusions	88
6	Outlook	89
7	References	93
	Appendix	102
A.1	Supplementary Material	102
A.1.1	Supplementary Figures	102
A.1.2	Photographs of Coated Samples	106
A.2	Ceramabind® 880 Datasheet and Application Guidelines	111

List of Abbreviations

CB	Conduction band
CCD	Charge-coupled device
CMOS	Complementary metal-oxide-semiconductor
CN	Coordination number
CP	Crossing point
FIR	Fluorescence intensity ratio
FWHM	Full width at half-maximum
KLPE	$\text{K}_3\text{LuSi}_2\text{O}_7:\text{Eu}^{2+}$
LE	Luminous efficacy
LIR	Luminescence intensity ratio
NIR	Near-infrared
PAA	Poly(acrylic acid)
PMMA	Poly(methyl methacrylate)
PSD	Particle size distribution
PSP	Pressure-sensitive paint
PTFE	Polytetrafluoroethylene
SCASN	$(\text{Sr,Ca})\text{AlSiN}_3:\text{Eu}^{2+}$
TSP	Temperature-sensitive paint
UV	Ultraviolet
VB	Valence band

1 Introduction

Temperature-sensitive paints (TSPs) enable non-contact, spatially resolved temperature measurements and have therefore become highly attractive for applications in which conventional temperature sensors provide only limited information or are difficult to implement on complex surfaces [1–3]. Classical TSPs are mostly organic coating systems that rely on thermal quenching processes of luminescent molecules or complexes. Although such systems are well established, their applicability is inherently limited at elevated temperatures, since both the organic luminophores and the organic binder tend to lose stability or degrade under harsh thermal conditions [1]. If temperature measurements are to be extended into higher-temperature regimes, inorganic phosphors and inorganic binders therefore become necessary.

This challenge is directly connected to the broader field of luminescence thermometry, which addresses temperature determination by means of temperature-dependent luminescent properties such as emission intensity, spectral characteristics, or decay time [4–9]. In recent years, this field has attracted considerable attention because inorganic luminescent materials often combine good chemical stability, high thermal resistance, and strong optical response. At the same time, the transfer of such concepts into coating-based systems remains highly relevant for practical applications, especially when phosphor-based TSPs are intended for demanding thermal environments.

At the German Aerospace Center (DLR), this topic has already been addressed in previous work, including the studies of *Marlies Maroka* [10], *Linda Bauck* [11], and *Mark Tanai* [12]. In particular, the work of *Marlies Maroka* and *Mark Tanai* dealt explicitly with the inorganic phosphor $(\text{Sr,Ca})\text{AlSiN}_3\text{:Eu}^{2+}$ (SCASN) and its use in inorganic TSP coatings. Beyond these internal studies, SCASN has also been reported in the literature as a promising material for temperature-sensing applications by *Hertle et al.* [13] and *Bollmann et al.* [14].

Against this background, one central aim of the present work is to deepen the understanding of the SCASN system, particularly with regard to temperature-dependent lifetime-based measurements and possible temperature hysteresis during heating and cooling cycles, in order to establish a sound data basis for the implementation of a camera-based measurement setup. A second aim is the exploration of alternative phosphors for phosphor-based TSP applications. For this reason, $\text{K}_3\text{LuSi}_2\text{O}_7\text{:Eu}^{2+}$ (KLPE) was investigated as a further candidate. Like SCASN, KLPE is a Eu^{2+} -activated phosphor, but it exhibits an even larger Stokes shift and a broad deep-red to near-infrared (NIR) emission band [15], which makes it particularly interesting from the perspective of optical signal separation. Accordingly, this work focuses on the preparation, characterization, and temperature-dependent evaluation of both phosphor types in powder form and, where applicable, as inorganic coating systems, with particular emphasis on their suitability for intensity-based and lifetime-based phosphor thermometry.

2 Theory

2.1 Temperature-Sensitive Paints

Temperature-sensitive paints (TSPs) are luminescent coating systems that enable non-contact, spatially resolved surface temperature measurements, based on thermal quenching processes. They are particularly attractive in situations where conventional contact sensors, such as thermocouples or resistance thermometers, provide only local information or are difficult to apply to geometrically complex surfaces. In contrast, TSPs allow full-field temperature measurements with high spatial resolution and are comparatively low cost, which has made them especially relevant for aerodynamic testing in wind tunnels to investigate complex flow phenomena [1–3, 16].

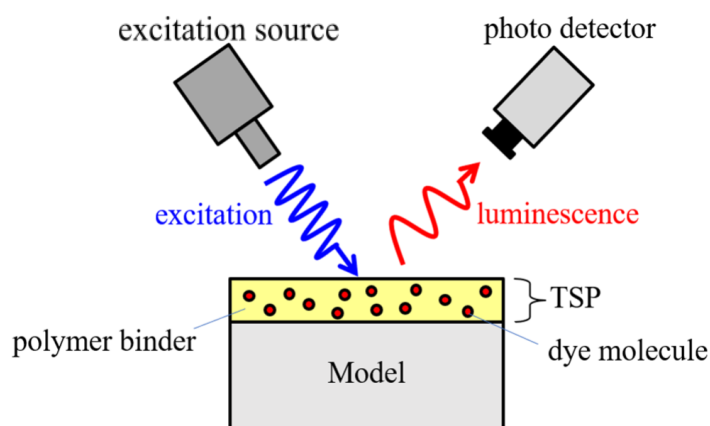


Figure 1: Schematic illustration of the structure and working principle of a TSP measurement. Adapted from [3].

The basic working principle of a TSP measurement is illustrated in **figure 1**. In a typical TSP system, luminescent sensor molecules, also called luminophores are immobilized in a suitable binder and applied as a thin coating to the surface of interest. Upon optical excitation, the coating emits luminescence, and the temperature-dependent change in emission intensity, decay time, or both can be evaluated to determine the local surface temperature [1–3]. In many cases, the luminophore/dye and the binder are first dissolved or dispersed in a solvent, after which the resulting formulation is deposited by spraying, airbrushing, or brushing. Following solvent evaporation, a solid coating remains in which the luminescent centers are immobilized within the matrix [1, 2].

A wide range of luminescent molecules has been investigated for TSP applications. Among the most common examples are Eu^{3+} -complexes and Ru^{2+} -complexes, two representative structures of which are shown in **figure 2** [1, 3, 16–18]. For practical use, these complexes should exhibit

strong and reproducible temperature-dependent luminescence as well as a sufficiently large Stokes shift, as also indicated in **figure 1** by the blue excitation light and the red luminescence, so that the excitation radiation can be separated efficiently from the emitted light by optical filters [1].

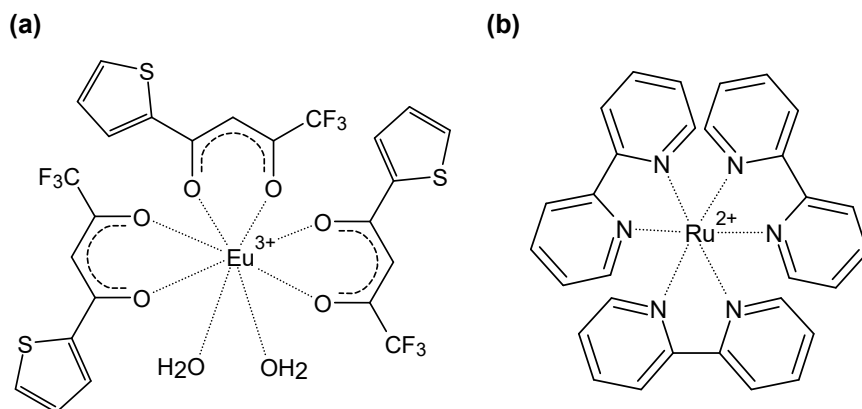


Figure 2: Chemical structures of two exemplary complexes used in TSPs: di(aqua)tris(thenoyltrifluoroacetato)europium(III) $[\text{Eu}(\text{ttfa})_3(\text{H}_2\text{O})_2]^0$ (a) and tris(2,2'-bipyridine)ruthenium(II) $[\text{Ru}(\text{bpy})_3]^{2+}$ (b). Adapted from [1].

Besides the luminophore itself, the binder plays a central role because it determines the mechanical integrity of the coating and strongly influences its thermal stability. Common organic binders reported for classical TSP formulations include poly(methyl methacrylate) (PMMA) and poly(acrylic acid) (PAA) [1, 19]. Polymer binders can offer good processability and optical performance, but their usable temperature range is limited. At elevated temperatures, not only can the luminescent complexes degrade, but the thermal stability and glass transition behavior of the polymer matrix may also become critical [1].

For this reason, conventional TSP systems based on luminescent molecules or complexes in polymer binders are generally not well suited for temperatures much above approximately 100 to 150 °C [1]. Consequently, the development of TSP systems for higher-temperature applications remains an active field of research. A central challenge is to replace the typically used organic luminophores with thermally more stable inorganic phosphors and, at the same time, to substitute the polymer binder with an inorganic matrix, while maintaining sufficient luminescence efficiency.

2.2 Luminescence Thermometry

The search for thermally more stable alternatives to conventional organic TSP systems is closely connected to the broader field of luminescence thermometry. This field has attracted considerable

attention, particularly during the last two decades, and encompasses a wide range of concepts, materials, and readout strategies for temperature determination based on luminescent processes [4–9]. Although luminescence thermometry is not restricted to inorganic materials, much recent work has focused on inorganic compounds because of their often superior thermal and chemical stability.

At the same time, the basic idea itself is far older. As early as 1949, *Urbach et al.* discussed the use of luminescent processes for temperature measurement and recording, including implementation in the form of a coating based on $\text{Zn}_{1-x}\text{Cd}_x\text{S}$ [5, 20]. Thus, the fundamental concept of luminescence-based surface thermometry has a long history, whereas modern research has greatly expanded both the accessible temperature range and the range of available materials.

Recent work illustrates this progress particularly well. For example, *Kulesza et al.* reported promising thermometric behavior for $\text{Ca}_3\text{Sc}_2\text{Ge}_3\text{O}_{12}:\text{Pr}^{3+}$ and $\text{Ca}_3\text{Sc}_2\text{Si}_3\text{O}_{12}:\text{Pr}^{3+}$ [21]. For $\text{Ca}_3\text{Sc}_2\text{Si}_3\text{O}_{12}:\text{Pr}^{3+}$, a relative temperature sensitivity of at least $0.5\% \text{ K}^{-1}$ was achieved over the wide temperature range from 250 to 1225 K by means of the so-called luminescence intensity ratio (LIR) method, which will be discussed later in more detail [21]. Such examples clearly demonstrate the considerable potential of inorganic luminescent materials for high-temperature thermometry.

One particularly important parameter in both luminescence thermometry and temperature-sensitive paints is the (negative) relative temperature sensitivity S_r . In the present work, the two principal temperature-dependent parameters investigated are the emission intensity and the luminescence decay time. Accordingly, the corresponding relative temperature sensitivities are denoted as $S_{r,I}$ and $S_{r,\tau}$, respectively. They are defined as follows [5, 7, 17]:

$$S_{r,I} = \frac{\Delta I \cdot 100}{\Delta T \cdot I_m} = \frac{-(I_2 - I_1) \cdot 100}{(T_2 - T_1) \cdot \left(\frac{I_1 + I_2}{2}\right)} \quad (1a) \quad S_{r,\tau} = \frac{\Delta \tau \cdot 100}{\Delta T \cdot \tau_m} = \frac{-(\tau_2 - \tau_1) \cdot 100}{(T_2 - T_1) \cdot \left(\frac{\tau_1 + \tau_2}{2}\right)} \quad (1b)$$

with:

- $S_{r,I}$: Relative temperature sensitivity for intensity-based thermometry in $\% \text{ K}^{-1}$
- I_2 : Emission intensity at the higher temperature
- I_1 : Emission intensity at the lower temperature
- T_2 : Higher temperature
- T_1 : Lower temperature
- $S_{r,\tau}$: Relative temperature sensitivity for lifetime-based thermometry in $\% \text{ K}^{-1}$
- τ_2 : Luminescence decay time at the higher temperature
- τ_1 : Luminescence decay time at the lower temperature

In this context, these quantities provide a convenient basis for comparing the performance of

different phosphors and different readout strategies within the same material system.

2.3 Temperature Readout Methods

As illustrated in **figure 3**, a broad variety of temperature readout methods is available in luminescence thermometry. These methods differ in the luminescence parameter from which the temperature information is extracted. Common approaches are based on temperature-dependent changes in emission bandwidth, spectral shifts of emission lines or bands, polarization-dependent intensity changes, luminescence intensity ratio (LIR) methods, temperature-dependent intensity changes of a single emission line or band, and temperature-dependent changes in luminescence lifetime. Since the present work focuses exclusively on intensity- and lifetime-based readout, the remaining approaches are only introduced briefly in the following to outline their general principles.

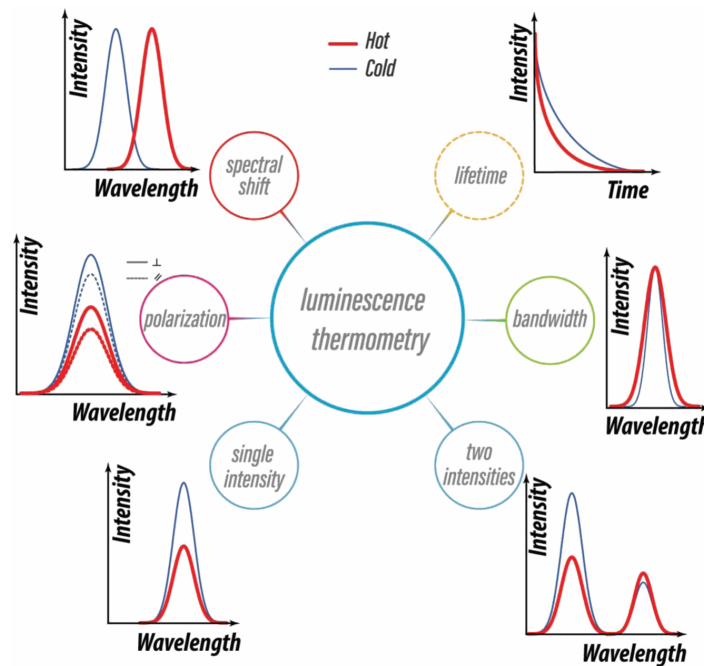


Figure 3: Overview of various temperature readout methods. Reproduced from [4].

Bandwidth

One possible readout strategy is based on the temperature-dependent broadening of emission lines or bands. The underlying origin of this behavior will be discussed later in more detail in the context of **equation (6)**. Although examples such as $\text{TiO}_2:\text{Tm}^{3+}$ [22] and $\text{YAlO}_3:\text{Nd}^{3+}$ [23] have been reported, temperature determination based on emission bandwidth is generally

considered impractical. The corresponding changes are usually rather small, and their accurate determination is comparatively time-consuming. For these reasons, this readout concept has so far not found broader practical application [7].

Spectral Shift

Another well-known approach is based on the temperature-dependent shift of emission lines or bands. A prominent example is ruby, $\text{Al}_2\text{O}_3:\text{Cr}^{3+}$, whose spectral shift is well-known [24]. In many luminescent materials, however, the absolute spectral shift is very small, often only on the order of a few picometers per kelvin. For instance, $\text{Y}_2\text{O}_2\text{S}:\text{Eu}^{3+}$ shows only a small spectral shift of roughly -3 pm K^{-1} [25], while for $\text{TiO}_2:\text{Tm}^{3+}$ a similarly small value of about -2.2 pm K^{-1} has been reported [22]. Accordingly, this method is often limited by the need for high spectral resolution and careful peak-position analysis. In practice, such comparatively small shifts can restrict its suitability for routine thermometric applications.

Polarization

Another possible temperature readout strategy is based on polarization-dependent luminescence. However, this concept is not considered further in the present work, since only very few representative examples have been reported in the literature and the underlying principle is generally not applicable to solid-state phosphors. It is included here merely for completeness, because it is mentioned in **figure 3**.

The basic idea is that, in anisotropic media, the luminescence intensity can depend strongly on the polarization state of the emitted light. The degree of anisotropy is temperature dependent because temperature influences the Brownian rotational motion of the emitting species [7, 26]. As the temperature increases, rotational motion becomes faster, so that a larger fraction of the emitted photons loses the polarization information of the incident excitation light during the fluorescence lifetime. Consequently, the degree of fluorescence anisotropy decreases with increasing temperature [7, 26].

Representative examples include the so-called green fluorescent protein [26] and the well-known fluorescein molecule, for which a decrease in the degree of anisotropy of about $3 \% \text{ K}^{-1}$ has been reported [7]. Since these examples are based on organic molecules in liquid solution, this approach is not suitable for the high-temperature applications considered in the present work.

Two Intensities / Luminescence Intensity Ratio

One of the most widely used temperature readout strategies is based on the luminescence intensity ratio (LIR) of two emission lines or bands [27], also referred to as the fluorescence intensity ratio (FIR) method [28]. According to *Suta* and *Meijerink*, it is by far the most commonly employed readout concept in luminescence thermometry [27].

In its classical form, the method relies on the ratio of two emission bands originating from thermally coupled excited states. Because the populations of these states are temperature dependent, the ratio of the corresponding emission intensities can be used to determine the local temperature [27]. If both emission bands arise from thermally coupled excited states of the same luminescent ion, the LIR is, in the classical case, expected to follow Boltzmann-type behavior at elevated temperatures, since the emission intensity is proportional to the population of the emitting state [27, 28].

Lanthanide ions are particularly well suited for this readout concept because their $4f^n$ electronic configurations provide numerous closely spaced excited states that can give rise to thermally coupled emission. In addition, their characteristic $4f^n \rightarrow 4f^n$ transitions usually produce narrow emission lines at well-defined wavelengths, which facilitates reliable intensity-ratio measurements with only minor spectral overlap [27]. A broad variety of lanthanide activators have therefore been found to be suitable for LIR-based thermometry. Representative examples include $\text{YF}_3\text{:Pr}^{3+}$ [29], Y_2O_3 doped with Er^{3+} , Dy^{3+} , or Eu^{3+} [30], NaYF_4 doped with Er^{3+} or Yb^{3+} [29], and $\text{YVO}_4\text{:Dy}^{3+}$ [31].

2.3.1 Intensity-Based Methods

Intensity-based methods based on a single emission line or band are conceptually closely related to LIR approaches. In this case, however, the temperature information is obtained from the absolute or relative change in the intensity of a single emission feature rather than from the ratio of two spectral components. Since the temperature dependence of the emission intensity is governed by thermal quenching, a suitable model for describing this behavior will be introduced later in the corresponding chapter (see **equation (4a)**). In the context of temperature-sensitive paints, intensity-based readout is particularly attractive because it can easily be combined with imaging detection. For this purpose, charge-coupled device (CCD) cameras and complementary metal-oxide-semiconductor (CMOS) cameras are commonly used to obtain spatially resolved temperature information [1].

2.3.2 Lifetime-Based Methods

Lifetime-based methods are based on the fact that the decrease of the emission intensity over time can be described according to the following formula [32, 33]:

$$I_t = A_0 + \sum_{i=1}^n B_i \cdot \exp\left(\frac{-t}{\tau_i}\right) \quad (2)$$

with:

- I_t : Emission intensity at time t
- A_0 : Intensity of the background signal
- B_i : Pre-exponential factor of the i th decay process at time $t = 0$ s
- t : Time
- τ_i : Decay time of the i th decay process

Accordingly, the decay time corresponds to the time after which the emission intensity has decreased to $1/e$, that is, to roughly 36.79 % of its initial value [32]. In the simplest case, the luminescence decay follows a mono-exponential behavior, meaning that $i = 1$. In many real systems, however, the decay is better described by two or more partial decay processes. For a bi-exponential decay, this corresponds to $i = 2$. In such cases, the so-called average decay time is often used to represent the overall decay behavior. For a bi-exponential decay, it can be calculated according to the following expression [32, 33]:

$$\tau_x = \frac{B_1 \cdot \tau_1^2 + B_2 \cdot \tau_2^2}{B_1 \cdot \tau_1 + B_2 \cdot \tau_2} \quad (3)$$

with:

- τ_x : Average decay time
- B_1 : Pre-exponential factor of the first decay process at time $t = 0$ s
- τ_1 : Decay time of the first decay process
- B_2 : Pre-exponential factor of the second decay process at time $t = 0$ s
- τ_2 : Decay time of the second decay process

Since the temperature dependence of the decay time is also closely connected to thermal quenching, an appropriate model for describing this behavior will be discussed later in the corresponding chapter (see **equation (4b)**). In temperature-sensitive paint applications, lifetime-based readout is commonly performed either by pulse methods, in which the temporal intensity decay is recorded after a short excitation pulse, or by phase methods, in which the phase shift between modulated excitation and modulated emission is evaluated to determine the decay time [1].

2.4 Thermal Quenching

Since thermal quenching is the driving force behind most of the temperature-dependent luminescence changes discussed so far, the underlying mechanisms are considered in more detail in this section. Particular emphasis is placed on the processes relevant to Eu^{2+} -activated phosphors, since Eu^{2+} is the activator employed throughout the present work.

The term thermal quenching refers to the general decrease in the efficiency of luminescent processes with increasing temperature [32]. This behavior can be attributed to different thermal quenching mechanisms, which depend strongly on the type of transition, for example $5d \leftrightarrow 4f$ or $4f \leftrightarrow 4f$ transitions, as well as on the specific host lattice into which the activator is incorporated.

Before discussing the different thermal quenching mechanisms, mathematical approaches used to describe the temperature dependence of luminescent properties, namely the emission intensity and the decay time, are introduced. The temperature-dependent decrease in emission intensity (see **equation (4a)**) as well as the temperature-dependent decrease in decay time (see **equation (4b)**) can be described using the *Struck-Fonger* model, which represents a modified Arrhenius-type equation [32, 34–36]:

$$I_T = \frac{I_0}{1 + D \cdot \exp\left(-\frac{E_A}{kT}\right)} \quad (4a) \qquad \tau_T = \frac{\tau_0}{1 + D \cdot \exp\left(-\frac{E_A}{kT}\right)} \quad (4b)$$

with:

- I_T : Emission integral at temperature T
- I_0 : Emission integral at temperature $T = 0$ K
- τ_T : Decay time at temperature T
- τ_0 : Decay time at temperature $T = 0$ K
- D : Frequency factor
- E_A : Activation energy for thermal quenching / eV
- k : Boltzmann constant (8.6173×10^{-5} eV/K)
- T : Temperature

From this model the quenching temperature $T_{1/2}$ can be determined, which corresponds to the temperature at which the emission integral decreases to 50 % of the emission integral at $T = 0$ K. The formula introduced above is only suitable when a single quenching process is present. If more than one quenching process contributes, the model has to be extended further, resulting in a two-weighted model that can again be used to describe the temperature-dependent decrease in emission intensity (see **equation (5a)**) as well as the temperature-dependent decrease in decay time (see **equation (5b)**) [36, 37]:

$$I_T = A \frac{I_{0,a}}{1 + B_a \exp\left(-\frac{E_{A,a}}{kT}\right)} + (1 - A) \frac{I_{0,b}}{1 + B_b \exp\left(-\frac{E_{A,b}}{kT}\right)} \quad (5a)$$

$$\tau_T = A \frac{\tau_{0,a}}{1 + B_a \exp\left(-\frac{E_{A,a}}{kT}\right)} + (1 - A) \frac{\tau_{0,b}}{1 + B_b \exp\left(-\frac{E_{A,b}}{kT}\right)} \quad (5b)$$

with:

- I_T : Emission integral at temperature T
- $I_{0,a}$: Emission integral of process a at temperature $T = 0$ K
- $I_{0,b}$: Emission integral of process b at temperature $T = 0$ K
- τ_T : Decay time at temperature T
- $\tau_{0,a}$: Decay time of process a at temperature $T = 0$ K
- $\tau_{0,b}$: Decay time of process b at temperature $T = 0$ K
- A : Fraction of process a
- $1 - A$: Fraction of process b
- B_a : Frequency factor of process a
- B_b : Frequency factor of process b
- $E_{A,a}$: Activation energy of process a / eV
- $E_{A,b}$: Activation energy of process b / eV
- k : Boltzmann constant (8.6173×10^{-5} eV/K)
- T : Temperature

After introducing these mathematical models, three general thermal quenching mechanisms relevant to Eu^{2+} luminescence are discussed in the following section. The categorisation and structure of this section are strongly influenced by *Pier* [38].

2.4.1 Thermal Crossover

One prominent concept of thermal quenching processes, especially for $5d \leftrightarrow 4f$ transitions, is the so-called thermal crossover, which is also referred to as thermally activated crossover [5, 39, 40]. The basic idea of this concept is illustrated in **figure 4**.

Upon excitation of the $4f$ ground state by incident radiation, represented by the blue arrow, the system reaches the excited $5d$ state. From there, the absorbed energy can either be released radiatively during relaxation to the ground state, represented by the red arrow, or the excited state can undergo further thermal activation until the so-called crossing point (CP) is reached, where the two parabolas intersect. At this point, vibrational levels of the excited and ground states become energetically resonant [41]. This enables non-radiative relaxation to the ground

state, which contributes to the temperature-dependent decrease in luminescence intensity [6, 42].

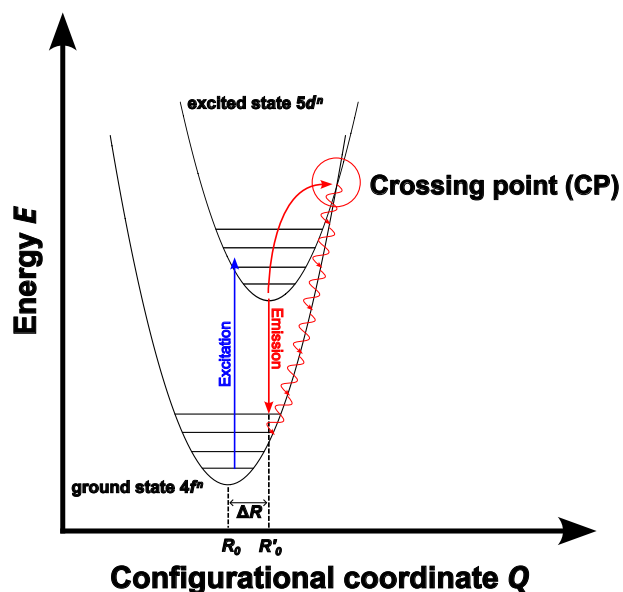


Figure 4: Schematic illustration of thermally activated crossover of a $5d \leftrightarrow 4f$ transition in a configurational coordinate diagram. Adapted from [7, 39, 41, 43]. The blue arrow indicates excitation from the ground to the excited state. From the excited state, either radiative relaxation to the ground state occurs, resulting in emission, or a thermally assisted non-radiative transition via the crossing point (intersection of the parabolas) takes place.

One particularly important parameter is ΔR , which is also indicated in **figure 4**. It describes the horizontal offset between the two parabolas and can be interpreted as the difference between the equilibrium positions of the ground and excited states [44, 45]. In structural terms, it corresponds to the average change in bond length between the luminescent center and the surrounding atoms of the coordination polyhedron in the ground and excited state [46].

If ΔR denotes the displacement between the equilibrium configurations of the ground and excited state, the absorption and emission process can be visualized as follows. Absorption initially occurs without a change in nuclear configuration. After excitation, the system relaxes to the equilibrium geometry of the excited state, which is shifted relative to that of the ground state by ΔR . Emission then takes place from this relaxed excited state, again without an instantaneous change in configuration, followed by relaxation back to the equilibrium geometry of the ground state [41]. This representation corresponds to the classical configurational coordinate model, although real systems may deviate from this simplified picture, for example when the excited state is distorted relative to the ground state [41].

A larger value of ΔR generally leads to a lower quenching temperature [44], because the crossing

point is then located at lower energy and can therefore be reached more easily by thermal activation.

Whether ΔR is positive or negative depends on the type of luminescent center. In the case of cation excitation, the excited electron density is distributed farther away from the nucleus [47, 48]. This increases the effective positive character of the cation and strengthens its attraction to the surrounding anions, which results in a negative value of ΔR [47, 48]. For anion excitation, the inverse trend is expected, and ΔR is therefore predicted to be positive [47, 48].

2.4.2 Thermal Ionization

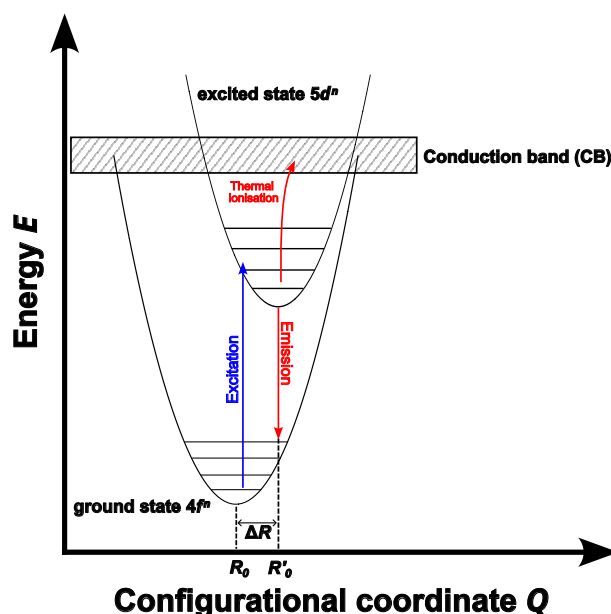


Figure 5: Schematic illustration of thermally induced ionization of a $5d \leftrightarrow 4f$ transition in a configurational coordinate diagram. Adapted from [39, 43]. The blue arrow indicates excitation from the ground to the excited state. From the excited state, either radiative relaxation back to the ground state occurs, resulting in emission, or a thermally assisted transition into the conduction band takes place.

Another thermal quenching mechanism that has been proposed is the so-called thermal ionization. This mechanism has been discussed particularly extensively by *Dorenbos* in the context of Eu^{2+} luminescence, whose work suggests that thermal ionization is the main contributor to thermal quenching in Eu^{2+} phosphors [49]. Thermal ionization refers to the idea that an excited state can be thermally promoted further into the conduction band (CB) states, if the energy barrier between the relaxed excited state and the conduction band is small enough [32, 49].

A basic illustration of this mechanism is given in **figure 5**. As before, the $4f$ ground state is

excited by incident radiation, represented by the blue arrow, so that the system reaches the excited $5d$ state. From there, the absorbed energy can either be released radiatively during relaxation to the ground state, represented by the red arrow, or the excited electron can be thermally promoted further into the conduction band, from where non-radiative relaxation to the ground state occurs.

This mechanism is particularly important for Eu^{2+} luminescence, since *Dorenbos* argued that if thermal crossover were the actual quenching mechanism in Eu^{2+} phosphors, the Stokes shift, meaning the difference between excitation and emission wavelength [32], should correlate with the quenching temperature and scale with the size of the site occupied by Eu^{2+} in the lattice [49]. However, the expected relationships were not found for all compounds in a study by *Dorenbos* covering more than 300 different Eu^{2+} compounds [50]. Further studies have shown that quenching of Eu^{2+} emission often proceeds via thermal ionization of the $5d$ electron into the conduction band [43].

Interestingly, although this example concerns Pr^{3+} luminescence, work by *Ueda et al.* demonstrated that thermal crossover and thermal ionization are not mutually exclusive quenching processes within a lattice [39]. Their results indicate that the $5d \leftrightarrow 4f$ luminescence of Pr^{3+} in $\text{Y}_3\text{Al}_{5-x}\text{Ga}_x\text{O}_{12}$ depends on the concentration of Ga^{3+} . For $x = 0, 1, 2$ thermal crossover is dominant, whereas for $x = 3, 4, 5$ thermally activated ionization becomes dominant due to the reduced energy gap between the lowest excited state ($5d_1$) and the conduction band at higher Ga^{3+} concentrations. As a result, less thermal activation energy is required, which promotes thermal ionization [39].

In another publication, *Ueda et al.* investigated the thermal quenching mechanism of $\text{CaAlSiN}_3:\text{Eu}^{2+}$, which is of particular interest in the context of the present work, since it is closely related to the phosphor system studied here, differing only in the partial substitution of Ca^{2+} by Sr^{2+} . Their results showed that the quenching process in $\text{CaAlSiN}_3:\text{Eu}^{2+}$ is governed by thermal ionization [51]. The same conclusion was later reached by *Tian et al.*, who likewise identified thermal ionization as the main thermal quenching mechanism in $\text{CaAlSiN}_3:\text{Eu}^{2+}$ [52].

2.4.3 Thermally Activated Concentration Quenching

Concentration quenching generally refers to the decrease in luminescent efficiency observed in many phosphors with increasing activator concentration [41, 53]. The commonly accepted mechanism is based on non-radiative energy migration from one excited luminescent center to a sequence of non-excited luminescent centers due to their resonance, ultimately ending at so-called killer centers, which are typically defects within the crystal lattice [53, 54]. Whether energy migration occurs between luminescent centers is strongly influenced by the spectral overlap between their emission and absorption spectra [41].

This is particularly relevant because, at elevated temperatures, the emission bands of activators such as Eu^{2+} tend to broaden. This behavior is attributed to the increased phonon population at higher temperatures, which enhances electron-phonon coupling and can be described by the following equation for the full width at half-maximum (FWHM) of the emission band [55, 56]:

$$\text{FWHM}(T) = W_0 \sqrt{\coth \frac{h\nu}{2kT}} \quad (6)$$

with: T : Temperature
 W_0 : FWHM at 0 K
 $h\nu$: Vibrational phonon energy
 k : Boltzmann constant (8.6173×10^{-5} eV/K)

As a result of this thermal broadening, the spectral overlap between the excitation and emission bands increases with rising temperature [54, 57]. This, in turn, can lead to faster energy migration and therefore to stronger concentration quenching [57].

This phenomenon has mainly been described for Ce^{3+} -activated phosphors, which also exhibit $5d \leftrightarrow 4f$ transitions [54, 58], but it has also been reported for Eu^{2+} -activated nitride-based phosphors such as $(\text{Sr,Ca,Ba})\text{Si}_2\text{O}_2\text{N}_2:\text{Eu}^{2+}$ [57], making it an important aspect to consider in the present work.

2.5 Eu^{2+} -Activated Phosphors

Eu^{2+} -activated phosphors constitute a highly versatile class of luminescent materials, exhibiting emission across an exceptionally broad spectral range. Their emission extends from the ultraviolet-A (UV-A) region through violet, blue, green, yellow, and red, and can even reach the near-infrared (NIR). A small selection of Eu^{2+} -activated phosphors covering this spectral range is summarized in **table 1**.

Eu^{2+} has the electron configuration $[\text{Xe}]4f^7$, which has a $^8\text{S}_{7/2}$ ground state. Eu^{2+} -activated phosphors generally exhibit broad excitation bands arising from transitions from this ground state to the $4f^65d^1$ excited states, as well as broad-band emission originating from the corresponding interconfigurational $5d \rightarrow 4f$ transition [59–61]. In Eu^{2+} -activated phosphors, both the $4f \rightarrow 5d$ excitation and the corresponding $5d \rightarrow 4f$ emission involve a change in parity. According to the so-called *Laporte* rule, electric-dipole transitions must involve such a change in parity [32]. These transitions are therefore electric-dipole allowed and typically intense [32, 60].

Table 1: Examples for Eu^{2+} -activated phosphors covering the spectral range from ultraviolet-A (UV-A) to near-infrared (NIR) emission, along with their peak emission wavelengths and corresponding references.

Phosphor	Spectral region	λ_{max} / nm	References
$\text{SrB}_4\text{O}_7:\text{Eu}^{2+}$	UV-A	368	[62]
$\text{BaLiZn}_3(\text{PO}_4)_3:\text{Eu}^{2+}$	Violet	387	[63]
$\text{BaMgAl}_{10}\text{O}_{17}:\text{Eu}^{2+}$	Blue	453	[64]
$\text{Ba}_3\text{La}_6(\text{SiO}_4)_6:\text{Eu}^{2+}$	Green	507	[65]
$\text{Sr}_3\text{SiO}_5:\text{Eu}^{2+}$	Yellow	570	[66]
$\text{Rb}_3\text{YSi}_2\text{O}_7:\text{Eu}^{2+}$	Red	622	[67]
$\text{K}_3\text{LuSi}_2\text{O}_7:\text{Eu}^{2+}$	NIR	740	[15]
$\text{Ca}_3\text{Sc}_2\text{Si}_3\text{O}_{12}:\text{Eu}^{2+}$	NIR	840	[68]

Although the $\text{Eu}^{2+} 4f^6 5d^1 \rightarrow 4f^7$ transitions are parity allowed, its decay time is still comparatively long, typically on the order of 0.1 to 1 μs depending on the host lattice, and thus noticeably longer than that of many Ce^{3+} -activated phosphors. This apparent discrepancy is commonly attributed to the multiplet structure of the excited $4f^6 5d^1$ configuration. While transitions from octet components to the $^8\text{S}_{7/2}$ ground state are spin allowed, the excited state also contains sextet character. Owing to spin orbit coupling, octet and sextet states are mixed, which imparts partial spin-forbidden character to the transition, since one relevant selection rule is $\Delta S = 0$ [32], thereby reducing the radiative transition probability. As a result, the luminescence lifetime of Eu^{2+} remains relatively long despite the formally parity-allowed nature of the $5d \rightarrow 4f$ transition [41, 59, 61].

The exceptionally broad spectral range over which Eu^{2+} -activated phosphors emit can be explained by the strength of the activator-ligand interaction, as illustrated in **figure 6**.

In contrast to the relatively well-shielded $4f^7$ states, the $4f^6 5d^1$ excited states are much more sensitive to the local coordination environment because of the larger spatial extent of the $5d$ orbitals. As a result, the energy of the $\text{Eu}^{2+} 5d$ states is strongly influenced by the host lattice, in particular by the coordination number, the bond lengths between Eu^{2+} and the surrounding anions, the site symmetry, and the covalent character of the chemical bonds [60, 70].

A key factor is the nephelauxetic effect, which is commonly described in terms of a centroid shift, that is, a lowering of the center of gravity (barycenter) of the $5d$ levels, and thus of the average energy of the $4f^6 5d^1$ excited-state levels, due to covalent interactions between Eu^{2+} and the

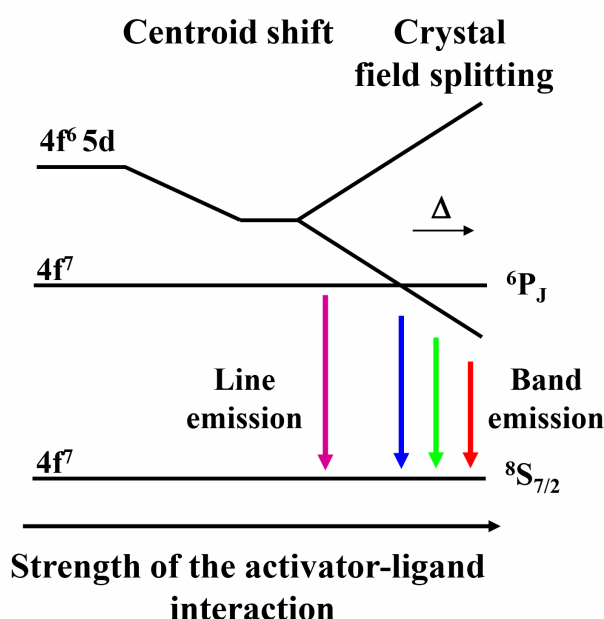


Figure 6: Effect of chemical environment on the emission of Eu^{2+} -activated phosphors [69].

surrounding ligands [32, 41, 71, 72]. In addition, the crystal field splits the $5d$ orbitals into several components, with the magnitude of this splitting depending on the symmetry and strength of the local ligand environment [70]. If the crystal-field splitting is relatively small, the energy distance between the lowest $5d$ level and the $8S_{7/2}$ ground state remains comparatively large, which leads to emission in the violet or blue spectral region. With increasing crystal-field splitting, however, the lowest $5d$ level is shifted to lower energy, thereby reducing the energy gap to the $8S_{7/2}$ ground state and causing the emission to shift progressively toward longer wavelengths. The combined effect of centroid shift and crystal-field splitting substantially modifies the position of the $4f^6 5d^1$ levels relative to the $4f^7$ configuration. Consequently, the transition energy can vary over a very wide range, which explains why Eu^{2+} -activated phosphors can exhibit emission from the UV to the red and even into the NIR spectral region [41, 60, 71, 72].

Besides their use as conventional luminescent materials, Eu^{2+} -activated phosphors are also of considerable importance in persistent luminescence materials, for which afterglow durations can range from a few seconds to more than 30 h [71, 73]. The basic principle of such afterglow phosphors is the delayed release of previously trapped charge carriers, either by thermal activation or by photostimulation, followed by radiative recombination and luminescence emission. In this context, trap states, lattice defects and, in many cases, additional co-dopants play a crucial role in determining the persistent luminescence behavior. Although a variety of mechanisms and models have been proposed to describe the detailed processes underlying persistent luminescence [73], these will not be discussed further here, since persistent luminescence is not the focus of the present work.

3 Experimental Section

3.1 Sample Preparation

3.1.1 $\text{K}_3\text{LuSi}_2\text{O}_7\text{:Eu}^{2+}$ Phosphor Powder

Samples with nominal compositions $\text{K}_{3-x}\text{Eu}_x\text{LuSi}_2\text{O}_7$, where $x = 0 - 0.05$, were prepared by solid-state synthesis. Stoichiometric amounts of Lu_2O_3 (99.99 %, Leuchtstoffwerk Breitenungen) and SiO_2 (99.9 %, ThermoFischer) were transferred into an agate mortar with isopropanol and thoroughly ground. To achieve very low doping concentrations, 4.4004 g of Eu_2O_3 (99.99 %, Tailorlux) were dissolved in a small amount of HNO_3 (≥ 65 %, Honeywell) and water, and the solution was made up to 100 mL in a volumetric flask, resulting in a Eu_2O_3 solution with a concentration of $0.044004 \text{ g mL}^{-1}$. This enabled the addition of volumes ranging from 20 μL for $x = 0.001$ up to 1000 μL for $x = 0.05$ to the mixture of Lu_2O_3 , SiO_2 , and isopropanol using a micropipette. After complete evaporation of the isopropanol, K_2CO_3 (≥ 99 %, Sigma-Aldrich) and 1.5 wt-% KF (≥ 99 %, VWR Chemicals), relative to the expected product mass, were additionally added as fluxing agents. The resulting white powder was then thoroughly ground again in the agate mortar under dry conditions because of the hygroscopic nature of K_2CO_3 and KF .

An excess of 30 wt-% K_2CO_3 and 10 wt-% SiO_2 was used to compensate for the evaporation of volatile compounds during calcination.

The powder was then transferred to a corundum crucible, and calcined at 1200°C for 3 h in a tubular furnace under a flowing reducing atmosphere of 90 % N_2 and 10 % H_2 to ensure the reduction of Eu^{3+} to Eu^{2+} . Finally, the obtained powder was again thoroughly ground in an agate mortar to ensure homogeneity. At lower Eu^{2+} concentrations, the powder exhibited a pale orange color, whereas at higher Eu^{2+} concentrations, it appeared orange-red.

Since Eu^{2+} and Eu^{3+} are known to occupy multiple sites within the host lattice, as will be discussed later, the samples are here referred to not by their nominal composition $\text{K}_{3-x}\text{Eu}_x\text{LuSi}_2\text{O}_7$, but rather as $\text{K}_3\text{LuSi}_2\text{O}_7\text{:xEu}^{2+}$ or more general $\text{K}_3\text{LuSi}_2\text{O}_7\text{:Eu}^{2+}$, in order to reflect the complexity of the actual $\text{Eu}^{2+}/\text{Eu}^{3+}$ site occupation.

3.1.2 Coated Samples

For the preparation of the coated samples, Ceramabind® 880, a commercially available inorganic, water-based binder based on a silicone resin, was used. A corresponding data sheet, including

the application guidelines, is provided in the appendix, see **section A.2**.

All coated samples were subjected to the same curing procedure. They were first kept at room temperature for 1 h. During this time, the oven was heated to 230 °C. Afterwards, the samples were placed in the preheated oven and kept at 230 °C for 1 h. The oven was then switched off, the oven door was briefly opened, during which a small amount of vapor escaped, and was subsequently left slightly ajar. The samples were then cooled to room temperature overnight.

(Sr,Ca)AlSiN₃:Eu²⁺ Coating

20250812-15: A suspension was prepared in a 30 mL test tube by suspending 2.5 g of (Sr,Ca)AlSiN₃:Eu²⁺ phosphor (FH Münster, Mitsubishi Chemical Corporation, Type: BR-102/SB) in 10.0 mL (10.75 g) of Ceramabind® 880 under stirring. The suspension was stirred for 10 min using a magnetic stirrer, ultrasonically treated for approximately 40 min, transferred into a spray gun, and sprayed at a pressure of 1.2 bar onto six aluminum substrates (3.0 cm × 1.5 cm) without a screen layer.

20250812-16: A suspension was prepared in a 30 mL test tube by suspending 0.4 g of (Sr,Ca)AlSiN₃:Eu²⁺ phosphor (FH Münster, Mitsubishi Chemical Corporation, Type: BR-102/SB) in 10.0 mL (10.75 g) of Ceramabind® 880 under stirring. The suspension was stirred for 10 min using a magnetic stirrer, ultrasonically treated for approximately 40 min, transferred into a spray gun, and sprayed at a pressure of 1.2 bar onto six aluminum substrates (3.0 cm × 1.5 cm) without a screen layer.

20260129-1: The preparation procedure was identical to that used for sample **20250812-16**. However, in addition to the aluminum substrates, steel substrates (1.2 cm × 1.2 cm), likewise without a screen layer, were also coated.

20260129-2: The preparation procedure was identical to that used for sample **20250812-15**. However, in addition to the aluminum substrates, steel substrates (1.2 cm × 1.2 cm), likewise without a screen layer, were also coated.

K₃LuSi₂O₇:Eu²⁺ Coating

All samples in this series were prepared using a phosphor with the stoichiometry K₃LuSi₂O₇:0.03 Eu²⁺.

20251119-1: Four aluminum substrates (1.2 cm × 1.2 cm) were coated using 0.6 g of $\text{K}_3\text{LuSi}_2\text{O}_7:0.03 \text{Eu}^{2+}$ phosphor (FH Münster, sample: DLR-MA-2025-FR-024) and 2.5 mL (2.69 g) of Ceramabind® 880. Only half of a spray pass could be applied, resulting in a comparatively rough coating.

20260129-3: A suspension was prepared in a 30 mL test tube by suspending 0.4 g of $\text{K}_3\text{LuSi}_2\text{O}_7:0.03 \text{Eu}^{2+}$ phosphor (FH Münster, sample: DLR-MA-2025-FR-052) in 10.0 mL (10.75 g) of Ceramabind® 880 under stirring. The suspension was stirred for 10 min using a magnetic stirrer, ultrasonically treated for approximately 40 min, transferred into a spray gun, and sprayed at a pressure of 1.2 bar onto four aluminum substrates and two steel substrates (all 1.2 cm × 1.2 cm) without a screen layer.

20260129-4: A suspension was prepared in a 30 mL test tube by suspending 1.5 g of $\text{K}_3\text{LuSi}_2\text{O}_7:0.03 \text{Eu}^{2+}$ phosphor (FH Münster, sample: DLR-MA-2025-FR-052) in 6.0 mL (6.45 g) of Ceramabind® 880 under stirring. The suspension was stirred for 10 min using a magnetic stirrer, ultrasonically treated for approximately 40 min, transferred into a spray gun, and sprayed at a pressure of 1.2 bar onto four aluminum substrates and two steel substrates (all 1.2 cm × 1.2 cm) without a screen layer.

Table 2: Overview of the coated samples, including the corresponding phosphor-to-binder mass ratio.

Sample number	Phosphor	Phosphor mass / g	Ceramabind® 880 mass / g	Ratio phosphor : binder
20250812-15	SCASN	2.5	10.75	1:4
20250812-16	SCASN	0.4	10.75	1:27
20260129-1	SCASN	0.4	10.75	1:27
20260129-2	SCASN	2.5	10.75	1:4
20251119-1	KLPE	0.6	2.69	1:4
20260129-3	KLPE	0.4	10.75	1:27
20260129-4	KLPE	1.5	6.45	1:4

Photographs of the coated samples are provided in **section A.1.2**.

3.2 Measurements

3.2.1 X-ray Powder Diffraction

X-ray powder diffraction measurements were performed with an X'Pert Pro MPD diffractometer (Malvern Panalytical GmbH) using copper K_α radiation ($\lambda = 0.154184$ nm) at 40 mA and 40 kV in Bragg–Brentano geometry. All samples were prepared on aluminum sample holders and measured over the range $2\theta = 10$ to 80° using a step width of 0.17° .

3.2.2 Diffuse Reflectance Spectroscopy

Diffuse reflectance spectra were recorded using a modified FLS 920 spectrometer (Edinburgh Instruments Ltd.) equipped with an integrating sphere coated with highly reflective polytetrafluoroethylene (PTFE, Spectralon[®]) and a 450 W xenon arc lamp (Model: XBO 450 W, Osram GmbH). The instrument comprises two monochromators (Czerny-Turner configuration, model: TMS300, Bentham Instruments Ltd.) with echelle gratings (1800 F mm^{-1}) and a photomultiplier tube (Model: R928, Hamamatsu Photonics GmbH), which was cooled to -20°C using Peltier elements. For reflectance measurements, BaSO_4 (99.99 %, Sigma Aldrich) was used as a white reference standard. All samples were prepared on PTFE sample holders.

3.2.3 Photoluminescence Spectroscopy

Room temperature excitation and emission spectra in the visible range were recorded on another FLS 920 spectrometer (Edinburgh Instruments Ltd.). This setup was likewise equipped with a xenon arc lamp (Model: XBO 450 W, Osram GmbH), two monochromators (Czerny-Turner configuration, model: TMS300, Bentham Instruments Ltd.) with echelle gratings (1800 F mm^{-1}), and a photomultiplier tube (Model: R2658P, Hamamatsu Photonics GmbH), which was cooled to -20°C using Peltier elements. Intensity fluctuations of excitation source were compensated using a simultaneously operated reference detector, whereas the wavelength-dependent detector sensitivity was corrected using a correction file obtained from a calibrated tungsten incandescent lamp certified by the National Physical Laboratory (NPL). All samples were prepared on black anodized aluminum sample holders.

From internal quality assurance measurements performed with a standard sample, namely $\text{Y}_3\text{Al}_5\text{O}_{12}:\text{Ce}^{3+}$, the relative measurement uncertainty of the emission spectra and the corresponding emission integrals at room temperature is estimated to be $\pm 3\%$. Nevertheless, this

value should be regarded only as a rough figure of merit. Accordingly, no error bars are included in the corresponding plots, since this uncertainty was not specifically validated for the phosphors investigated in the present work.

3.2.4 Decay Curve Measurements

The room temperature decay curves were recorded using the same spectrometer described in **section 3.2.3**. Excitation was provided by a pulsed laser diode ($\lambda_{\text{Ex}} = 445 \text{ nm}$) operating in the picosecond range (Model: EPL Series, Edinburgh Instruments Ltd.). The decay times were determined by fitting the decay curves to the function described in **equation (2)**, using either $i = 1$ for mono-exponential decay processes or $i = 2$ for bi-exponential decay processes.

3.2.5 Temperature-Dependent Spectroscopy

Temperature-Dependent Emission Spectra

The temperature-dependent emission spectra were recorded using the same spectrometer and excitation source as described in **section 3.2.3**.

For measurements in the range of 77 to 500 K, a liquid nitrogen-cooled cryostat (Model: Microstat N2, Oxford Instruments GmbH) was used. The spectra from 100 to 500 K were recorded in 50 K intervals, with a temperature tolerance of $\pm 3 \text{ K}$.

For measurements in the range from room temperature to 480 °C, a custom-built water-cooled sample holder was employed. This setup contained a copper block, which was heated by a heating wire positioned beneath the copper block and on which the samples were mounted. For the coated samples, a small amount of conductive silver varnish (L100, Kemo-Electronic GmbH) was applied in order to ensure efficient heat transfer between the sample and the copper block. The temperature was controlled using a temperature controller (Model: ENDA EUC442), with the temperature sensor inserted into a hole drilled into the copper block directly below the sample. The spectra from 80 to 480 °C were recorded in 50 °C intervals, with a temperature tolerance of $\pm 3 \text{ °C}$. For both temperature ranges, the pressure inside the sample holders was reduced to approximately 100 mbar using a vacuum pump.

For selected measurements, the relative emission integrals of the recorded spectra were shown as a function of temperature and fitted using a non-linear function based on the *Struck-Fonger*

model (see **equation (4a)**). From this fit, the quenching temperature $T_{1/2}$ was determined, which corresponds to the temperature at which the emission integral decreases to 50 % of its value at $T = 0$ K.

As mentioned earlier, internal quality assurance measurements performed with a standard sample, namely $\text{Y}_3\text{Al}_5\text{O}_{12}:\text{Ce}^{3+}$, suggest a relative measurement uncertainty of approximately ± 3 % for the emission spectra and the corresponding emission integrals at room temperature. However, this value should only be regarded as a rough figure of merit for the present temperature-dependent measurements, since it was established exclusively for the standard sample and under room-temperature conditions. For this reason, no error bars are included in the corresponding plots.

Temperature-Dependent Decay Curves

The temperature-dependent decay curves were recorded using the same spectrometer setup described in **section 3.2.4**.

The same heating and cooling equipment, sample preparation, temperature ranges, and measurement intervals were used as described in **section 3.2.5**.

As before, the decay times were determined by fitting the decay curves to the function described in **equation (2)**, using either $i = 1$ for mono-exponential decay processes or $i = 2$ for bi-exponential decay processes. The standard deviations of the decay times and the reduced χ^2 values obtained from the fits are indicated alongside the results.

In selected cases, analogous to the procedure used for the temperature-dependent emission spectra, the resulting decay times were shown as a function of temperature and fitted using a non-linear function based on the *Struck-Fonger* model (see **equation (4b)**). Here, $T_{1/2}$ can likewise be determined, corresponding in this case to the temperature at which the decay time decreases to 50 % of its value at $T = 0$ K.

3.2.6 X-ray Excited Spectroscopy

X-ray excited emission spectra at room temperature were measured with a modified FLS 980 spectrometer (Edinburgh Instruments Ltd.), which is equipped with an emission monochromator (Czerny-Turner configuration, model: TMS300, Bentham Instruments Ltd.) and a photomultiplier tube (Model: R928P, Hamamatsu Photonics GmbH), cooled to -20 °C using Peltier elements.

X-ray excitation was provided by a water-cooled tungsten X-ray tube (Model: Neptune 5200, Oxford Instruments GmbH). The recorded emission spectra were again corrected using a sensitivity correction file obtained from a calibrated tungsten incandescent lamp certified by the National Physical Laboratory (NPL).

3.2.7 Particle Size Distribution

Particle size distribution (PSD) measurements were performed using the laser scattering particle sizer Partica LA-960V2 (HORIBA Europe GmbH). The measurements were carried out after 4 min of ultrasonic treatment within the device, and the PSD results are reported as volume-based distributions. For $\text{K}_3\text{LuSi}_2\text{O}_7:\text{Eu}^{2+}$ a refractive index of 1.80 was assumed, since literature data indicate a value of approximately 1.80 for the chemically very similar $\text{Lu}_2\text{Si}_2\text{O}_7$. However, it should be noted that these data are based on calculations rather than experimental measurements, although the authors report good agreement between calculated and experimental data for other materials [74]. A spherical particle shape was assumed. As water was used as the dispersing medium, the refractive index of the fluid was set to 1.33.

3.2.8 Further Photometric and Colorimetric Properties

Using the emission spectra as input, the luminous efficacy (LE) and CIE 1931 x/y color coordinates of the prepared samples were calculated with the OSRAM ColorCalculator v7.71. The centroid wavelength, defined as the wavelength that divides the emission spectrum into two equal integral halves, was calculated according to **equation (7)** [32]:

$$\lambda_c = \frac{\int_{\lambda_1}^{\lambda_2} \lambda \cdot S(\lambda) \cdot d\lambda}{\int_{\lambda_1}^{\lambda_2} S(\lambda) \cdot d\lambda} \quad (7)$$

with: λ_c : Centroid wavelength
 λ : Wavelength / nm
 $S(\lambda)$: Emission intensity at λ / cps

3.2.9 Layer Thickness

The layer thicknesses of the selected coated samples were measured using a QNix[®] 9500 (Automation Dr. Nix GmbH & Co. KG) in combination with a QNix[®] Probe 3.

3.2.10 Roughness

The roughness measurements of the coated samples were performed using a Perthometer S2 (Mahr GmbH). Each sample was measured in triplicate, each in x and y direction and using a total measurement length of 1.750 mm, divided into five intervals of 0.250 mm. From these measurements, the average roughness R_a , corresponding to the average deviation of the profile from its mean line, the mean maximum peak-to-valley height across the intervals R_z , and the maximum peak-to-valley height over the entire measurement length R_{max} were determined [75].

4 Results and Discussion

4.1 (Sr,Ca)AlSiN₃:Eu²⁺ Phosphor Powder

4.1.1 Luminescence Properties

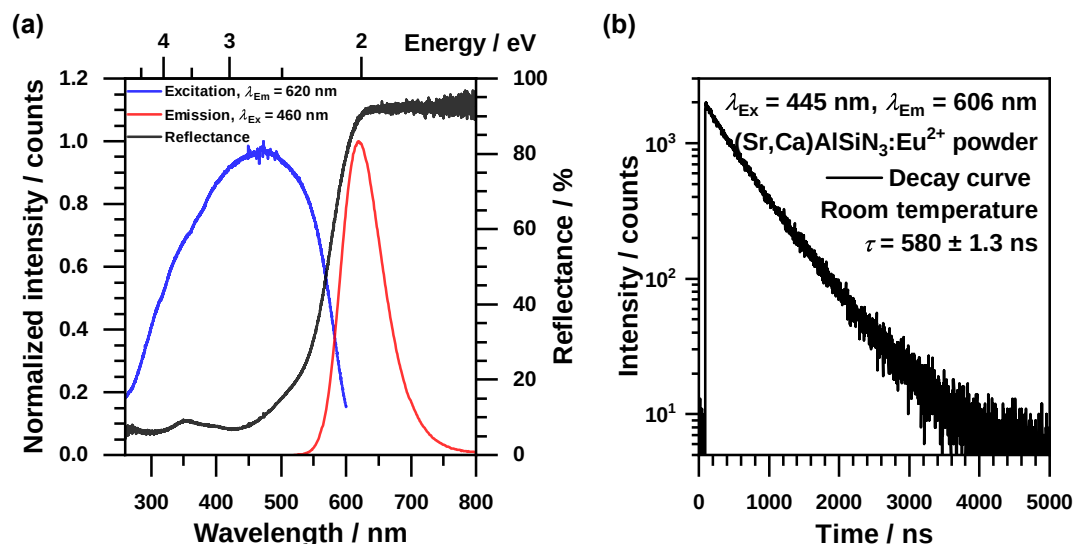


Figure 7: Excitation, emission, and reflectance spectra (a) together with the decay curve (b) of (Sr,Ca)AlSiN₃:Eu²⁺ powder at room temperature. The excitation spectrum was recorded at an emission wavelength of 620 nm, the emission spectrum at an excitation wavelength of 460 nm, and the decay curve at an excitation wavelength of 445 nm and an emission wavelength of 606 nm.

As already shown in the work of *Marlies Maroka*, the nitride Eu²⁺-activated phosphor (Sr,Ca)AlSiN₃:Eu²⁺ is a promising candidate for phosphor-based temperature-sensitive paints intended for higher-temperature applications. As shown in **figure 7(a)**, this phosphor exhibits an intense and broad excitation band with high intensity roughly between 350 to 500 nm. In addition, a somewhat narrower, yet still broad and intense, emission band is observed approximately between 600 to 650 nm, which is characteristic of Eu²⁺-activated phosphors. The diffuse reflectance spectrum further supports the strong optical absorption in the relevant excitation range. A closely related phosphor, namely CASN, has been reported to exhibit a high external quantum efficiency of 87 % [76], which further underlines the general suitability of the CASN/SCASN material class. Since the excitation maximum is located around 460 nm and the emission maximum around 620 nm, the large Stokes shift generally desired for TSP applications is clearly present. **Figure 7(b)** further shows that the decay time at room temperature, measured at an excitation wavelength of 445 nm and an emission wavelength of 606 nm, is (580.0 ± 1.3) ns. This value lies within the typical range for Eu²⁺-activated phosphors and, in principle, enables a fast response

when a lifetime-based TSP readout is employed. Overall, the luminescence properties of the SCASN phosphor powder are very favorable for the intended application.

4.1.2 Assessment of Possible Oxygen Quenching

For temperature-sensitive paints based on luminescent complexes, it is common to assess not only temperature sensitivity but also possible pressure sensitivity, since such systems often respond to both parameters [17]. For inorganic phosphors, pressure sensitivity is much less common. Nevertheless, cases have been reported in which inorganic phosphors exhibit pressure-dependent behavior due to oxygen quenching [77, 78]. For this reason, the possible influence of oxygen quenching on the SCASN phosphor powder was examined by recording the emission spectra under a 100 % oxygen atmosphere and, for comparison, under a 100 % nitrogen atmosphere. The corresponding spectra are shown in **figure 8**. No measurable difference between the two atmospheres can be observed under these conditions. It was therefore concluded that oxygen quenching does not play a significant role for this phosphor powder at room temperature. At the same time, this conclusion should not be generalized uncritically to elevated temperatures, for which the behavior may differ.

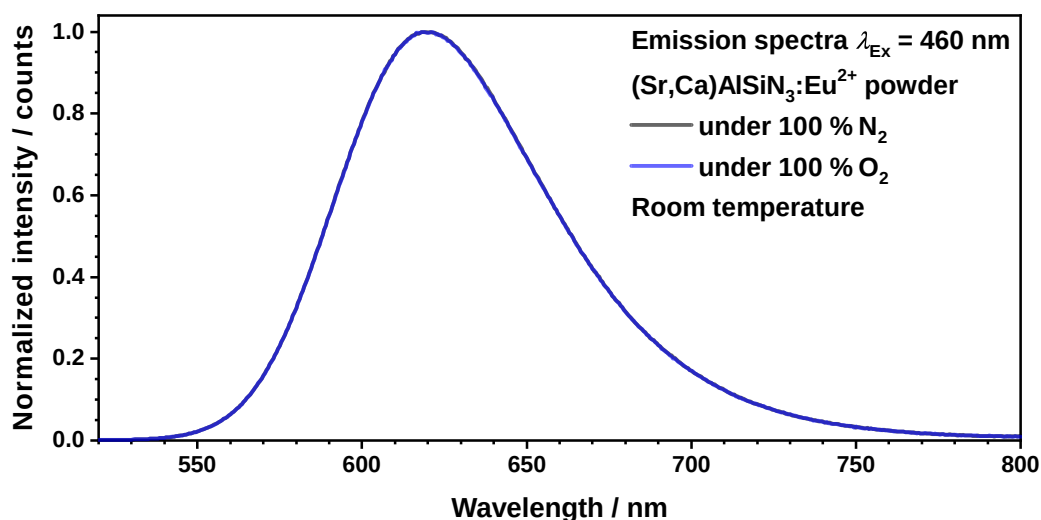


Figure 8: Emission spectra of (Sr,Ca)AlSiN₃:Eu²⁺ powder under 100 % N₂ atmosphere and 100 % O₂ atmosphere at room temperature, recorded at an excitation wavelength of 460 nm.

4.1.3 Thermal Degradation

Another important aspect investigated for the SCASN phosphor powder was its thermal degradation behavior. The aim was to determine the maximum temperature to which the powder can be exposed before an irreversible decrease in luminescence intensity occurs. This distinction is essential and should therefore be stated explicitly. Whereas thermal quenching refers to a reversible temperature-dependent decrease in a luminescence property, thermal degradation denotes an irreversible process that alters the chemical structure of the phosphor and thus causes a permanent loss in luminescence intensity.

For this purpose, the powder was heated in air for 2 h at 100 °C, after which the emission spectrum was measured at room temperature. The sample was then heated again for 2 h at 200 °C, measured once more, and this procedure was continued stepwise up to 600 °C. After each heating step, the phosphor powder was removed from the drying oven or furnace and allowed to cool naturally to room temperature. The resulting spectra are shown in **figure 9**. Up to 500 °C, no significant change in the emission spectra is observed. After treatment at 600 °C for 2 h, however, the emission intensity decreases markedly. The intensity loss amounts to roughly 30 % compared with the untreated SCASN powder.

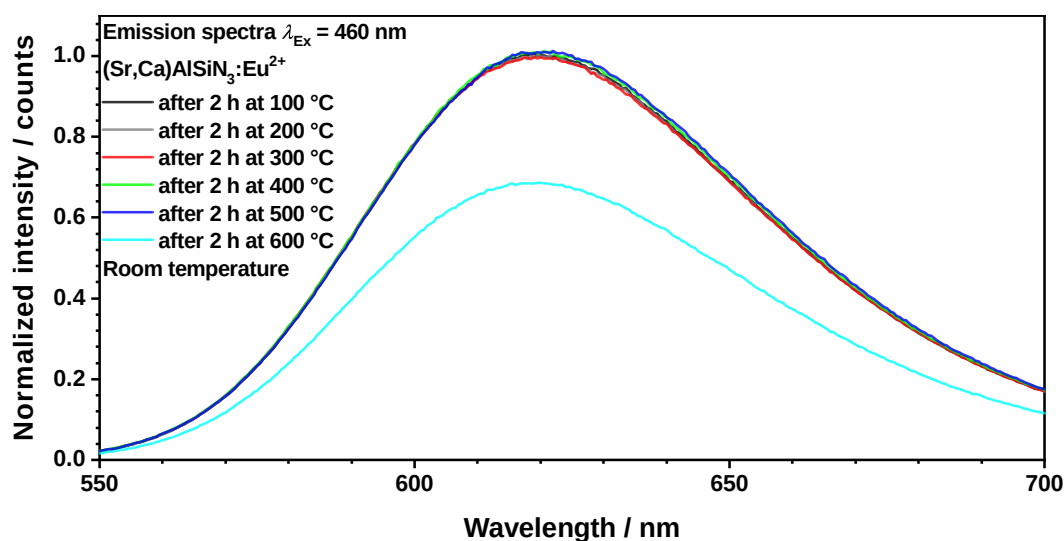


Figure 9: Emission spectra measured at room temperature of (Sr,Ca)AlSiN₃:Eu²⁺ after thermal treatment for 2 h at the indicated temperatures, recorded at an excitation wavelength of 460 nm.

This behavior is in good agreement with the work of *Oishi et al.*, who showed by thermogravimetric analysis coupled with mass spectrometry that, at around 600 °C, CASN powder starts to release N₂ and N₂O, even under inert helium atmosphere [79]. On this basis, it was concluded that the upper temperature limit for the application as a TSP is approximately 500 °C.

4.2 (Sr,Ca)AlSiN₃:Eu²⁺ Coating

4.2.1 Temperature-Dependent Emission Spectra

Building on the results obtained for the SCASN phosphor powder, the next step was to investigate the temperature-dependent behavior of the coated samples. As a first step, temperature-dependent emission spectra were recorded in order to assess the suitability of the coatings for intensity-based readout.

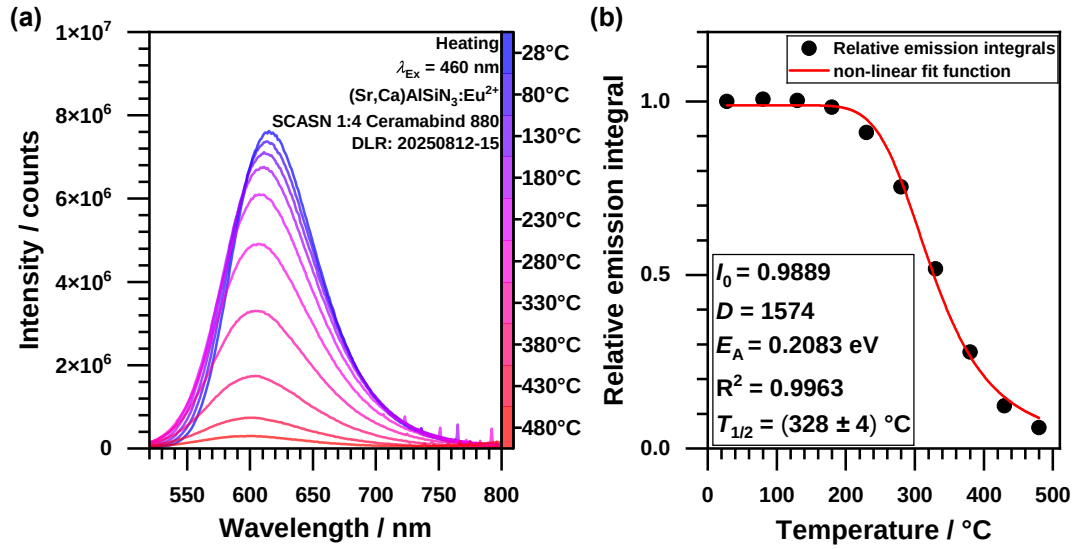


Figure 10: Emission spectra recorded upon heating (a) and the corresponding relative emission integrals together with the non-linear fit function (b) of sample 20250812-15 (SCASN 1:4 Ceramabind[®] 880) in the temperature range from 28 to 480 °C at an excitation wavelength of 460 nm.

Figure 10(a) shows the temperature-dependent emission spectra recorded upon heating sample 20250812-15 (SCASN 1:4 Ceramabind[®] 880) in the temperature range from 28 to 480 °C, whereas **figure 10(b)** presents the corresponding relative emission integrals together with a non-linear fit based on **equation (4a)**. With increasing temperature, the decrease in emission intensity due to thermal quenching is clearly observed. In addition, a temperature-dependent broadening of the emission band occurs, which is a phenomenon already discussed in **section 2.4.3** and can be rationalized by **equation (6)**. Another important observation is the blue shift of the emission band with increasing temperature. This behavior is well known for Eu²⁺-activated phosphors and is commonly attributed to thermally induced expansion of the host lattice, which modifies the local environment of the activator ion and thereby weakens the crystal field acting on the 5d states [13, 55, 80]. As a consequence, the emission of the excited 5d states is shifted to higher energy, resulting in a shift of the emission maximum toward shorter wavelengths [55, 80]. Overall, the observed spectral changes are therefore fully consistent with the expected

temperature-dependent behavior.

With the non-linear fit based on the *Struck-Fonger* model, a good agreement with the temperature dependence of the relative emission integrals was achieved. As mentioned earlier, this fit allows the quenching temperature $T_{1/2}$ to be determined, that is, the temperature at which the emission integral decreases to 50 % of the value extrapolated for $T = 0$ K. For sample 20250812-15, a value of $(328 \pm 4)^\circ\text{C}$ was obtained. The relative temperature sensitivity $S_{r,I}$ was calculated to be $0.39\% \text{ K}^{-1}$ in the temperature range from 28 to 480°C .

Figure 11 shows the same type of data as discussed above, but now for sample 20250812-16 (SCASN 1:27 Ceramabind[®] 880), that is, a coating containing a lower phosphor concentration. Nevertheless, the same temperature-dependent trends are observed. These include the decrease in emission intensity due to thermal quenching as well as the broadening and blue shift of the emission band, although the absolute emission intensity is lower throughout the entire temperature range. The corresponding $T_{1/2}$ value was again determined from the non-linear fit and amounts to $(330 \pm 3)^\circ\text{C}$. This value is in very good agreement with that of sample 20250812-15, for which $T_{1/2} = (328 \pm 4)^\circ\text{C}$ was obtained. The relative temperature sensitivity $S_{r,I}$ was calculated to be $0.36\% \text{ K}^{-1}$ in the temperature range from 26 to 480°C . This value is likewise very similar to that of sample 20250812-15, for which $0.39\% \text{ K}^{-1}$ was obtained.

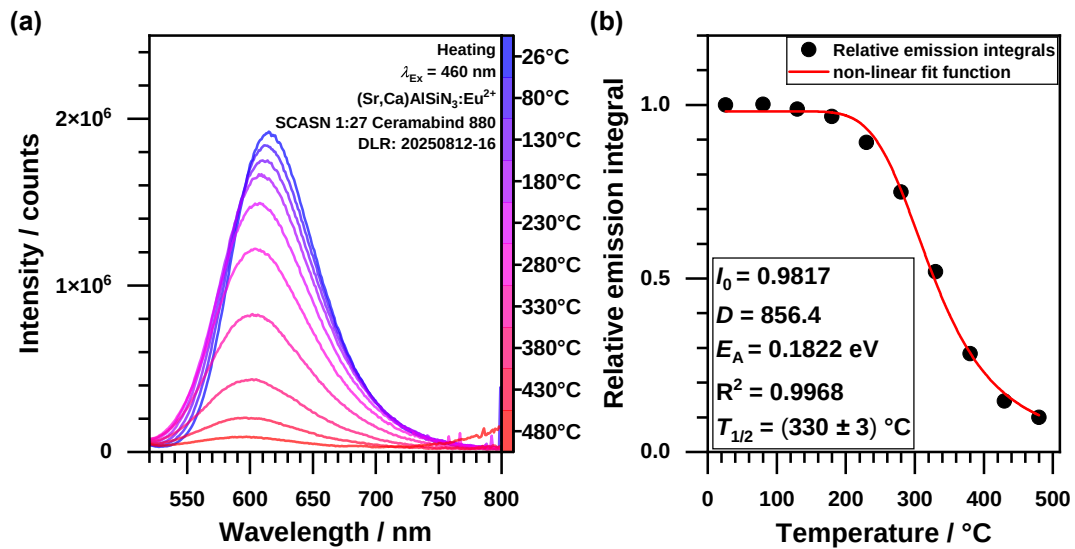


Figure 11: Emission spectra recorded upon heating (a) and the corresponding relative emission integrals together with the non-linear fit function (b) of sample 20250812-16 (SCASN 1:27 Ceramabind[®] 880) in the temperature range from 26 to 480°C at an excitation wavelength of 460 nm.

Comparison of Different Phosphor Concentrations

Since both coatings exhibit very similar temperature-dependent behavior, a more direct comparison of their absolute emission intensities is useful. For this purpose, the ratio between the absolute emission integrals of both samples was calculated according to the following expression:

$$R_{I,21} = \frac{I_2}{I_1} \cdot 100 \quad (8)$$

with: $R_{I,21}$: Absolute emission integral of sample 2 relative to that of sample 1 / %
 I_2 : Absolute emission integral of sample 2
 I_1 : Absolute emission integral of sample 1

The results of this calculation are presented in **table 3**:

Table 3: Absolute emission integrals and the corresponding relative emission ratio of samples 20250812-15 (SCASN 1:4 Ceramabind[®] 880) and 20250812-16 (SCASN 1:27 Ceramabind[®] 880) are compared. The relative emission ratio $R_{I,21}$ is defined according to **equation (8)**.

Temperature / °C	Absolute emission integral I_1 20250812-15	Absolute emission integral I_2 20250812-16	Emission ratio $R_{I,21}$ / %
28 / 26	6.695×10^8	1.703×10^8	25.4
80	6.737×10^8	1.707×10^8	25.3
130	6.711×10^8	1.682×10^8	25.1
180	6.584×10^8	1.647×10^8	25.0
230	6.094×10^8	1.519×10^8	24.9
280	5.043×10^8	1.276×10^8	25.3
330	3.465×10^8	8.846×10^7	25.5
380	1.856×10^8	4.817×10^7	26.0
430	8.214×10^7	2.492×10^7	30.3
480	4.001×10^7	1.692×10^7	42.3

It becomes evident that the emission ratio remains remarkably constant over a wide temperature

range. From room temperature up to 380 °C, $R_{l,21}$ varies only between 24.9 to 26.0 %. This shows that reducing the phosphor concentration lowers the absolute signal level, but has only a very minor influence on the temperature-dependent emission behavior in this range. In other words, both coatings exhibit nearly identical thermal quenching characteristics after normalization, while the lower-loaded sample retains approximately one quarter of the absolute emission intensity of the more highly loaded sample.

At 430 °C and especially at 480 °C, however, the ratio increases noticeably. This deviation should be interpreted with caution. At these temperatures, the absolute emission integrals have already decreased substantially for both samples, so that even comparatively small absolute fluctuations lead to much larger relative deviations in the calculated ratio. Accordingly, the apparent increase in $R_{l,21}$ at the highest temperatures most likely reflects the declining signal level and the associated increase in relative uncertainty rather than a fundamental change in the thermal quenching mechanism.

Overall, these results indicate that a substantial reduction in phosphor concentration is possible without significantly affecting either $T_{1/2}$ or $S_{r,l}$. At the same time, the much lower absolute emission intensity of the less concentrated coating has to be taken into account. Within the practically relevant range up to about 380 °C, the lower phosphor loading therefore appears feasible. Above this temperature, however, the intensity-based readout method shows markedly larger deviations between both coatings, indicating that the results are less comparable and less reproducible than in the lower-temperature range.

Comparison of Heating and Cooling

Another important aspect is the comparison of the emission behavior during heating and during cooling, because deviations between both branches may occur, a phenomenon commonly referred to as temperature hysteresis [2]. This point is particularly relevant for practical applications, since reliable temperature readout over full heating and cooling cycles requires sufficiently reproducible behavior in both directions.

For sample 20250812-15 (SCASN 1:4 Ceramabind® 880), the emission spectra recorded during heating and cooling are shown in **figure 12**. Due to technical problems with the heated sample holder, this comparison could only be carried out in the temperature range from 25 to 380 °C. Even within this restricted interval, however, a pronounced decrease in emission intensity during cooling is observed relative to the corresponding heating spectra.

The corresponding relative emission integrals are shown in **figure 13**. The difference between heating and cooling is clearly visible over the entire investigated range. A plausible explanation is

that irreversible degradation processes already occur in the coated samples at lower temperatures than in the phosphor powder. Compared with the bulk powder, the coating contains only a thin phosphor layer and therefore provides a much larger effective surface area. Moreover, the phosphor is deposited on an aluminum substrate, which ensures efficient heat transfer. In addition, the degradation behavior discussed in **section 4.1.3** involves release of N₂ and N₂O during thermal treatment, and this process may be further promoted by the reduced pressure inside the sample holder, as described in **section 3.2.5**.

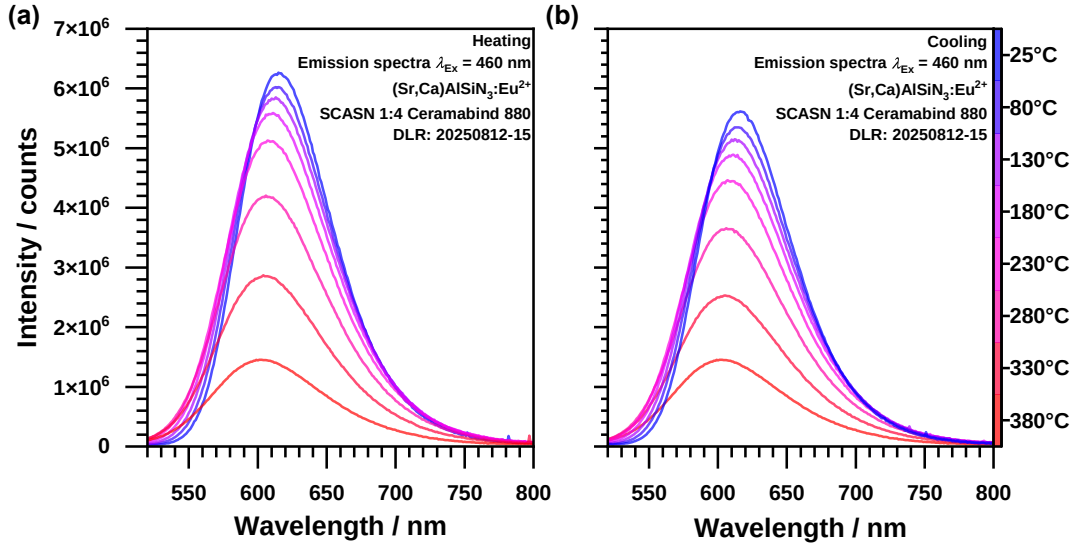


Figure 12: Emission spectra recorded upon heating (a) and cooling (b) of sample 20250812-15 (SCASN 1:4 Ceramabind® 880) in the temperature range from 25 to 380 °C at an excitation wavelength of 460 nm.

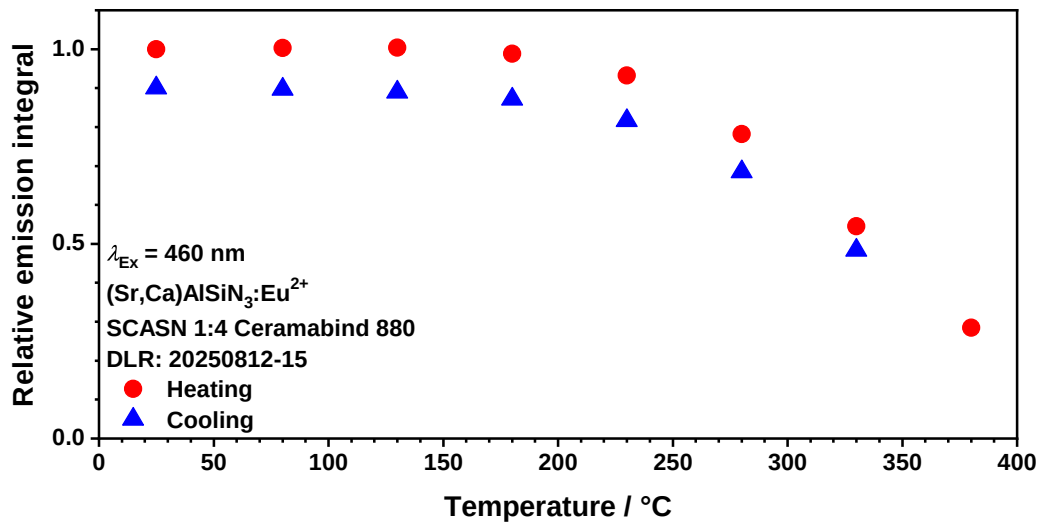


Figure 13: Relative emission integrals of sample 20250812-15 (SCASN 1:4 Ceramabind® 880) obtained from the emission spectra recorded upon heating and cooling in the temperature range from 25 to 380 °C at an excitation wavelength of 460 nm.

To quantify the difference between heating and cooling, the following relative changes were calculated. For the emission intensity, **equation (9a)** was used. Since the decay time will be analyzed analogously later, **equation (9b)** is given here as well in order to emphasize the close correspondence between both metrics.

$$\Delta I_{\text{rel, hc}} = \frac{I_{\text{cooling}} - I_{\text{heating}}}{I_{\text{heating}}} \cdot 100 \quad (9a) \quad \Delta \tau_{\text{rel, hc}} = \frac{\tau_{\text{cooling}} - \tau_{\text{heating}}}{\tau_{\text{heating}}} \cdot 100 \quad (9b)$$

with:

- $\Delta I_{\text{rel, hc}}$: Relative change of the relative emission integral during cooling compared to the corresponding heating value / %
- I_{heating} : Relative emission integral during heating at temperature T
- I_{cooling} : Relative emission integral during cooling at temperature T
- $\Delta \tau_{\text{rel, hc}}$: Relative change of the decay time during cooling compared to the corresponding heating value / %
- τ_{heating} : Decay time during heating at temperature T
- τ_{cooling} : Decay time during cooling at temperature T

Here, the relative change was intentionally calculated asymmetrically, that is, normalized to the heating value rather than to the mean of heating and cooling. This definition emphasizes the deviation of the cooling branch from the heating branch, which is the relevant quantity in the present context.

For sample 20250812-15, the resulting values of $\Delta I_{\text{rel, hc}}$ lie between –9.94 to –12.44 %, as presented in **table 4**. Thus, the cooling branch is consistently lower than the corresponding heating branch by about 10 to 12 %, which represents a pronounced hysteresis effect.

Table 4: Relative emission integrals of sample 20250812-15 (SCASN 1:4 Ceramabind® 880) obtained during heating and cooling are compared. The relative change $\Delta I_{\text{rel, hc}}$ is defined according to **equation (9a)**.

Temperatur / °C	Relative emission integral I_{heating}	Relative emission integral I_{cooling}	Relative change $\Delta I_{\text{rel, hc}}$ / %
25	1.000	0.901	–9.94
80	1.003	0.897	–10.59
130	1.004	0.890	–11.39
180	0.988	0.871	–11.84
230	0.932	0.816	–12.44
280	0.782	0.685	–12.40
330	0.545	0.483	–11.31

Temperatur / °C	Relative emission integral I_{heating}	Relative emission integral I_{cooling}	Relative change $\Delta I_{\text{rel, hc}} / \%$
380	0.285		

The same procedure was applied to sample 20250812-16 (SCASN 1:27 Ceramabind[®] 880). In this case, the comparison could only be performed between 80 to 480 °C, because the room temperature data point was affected by a technical error. In contrast to sample 20250812-15, **figure 14** reveals an increase in emission intensity during cooling relative to heating.

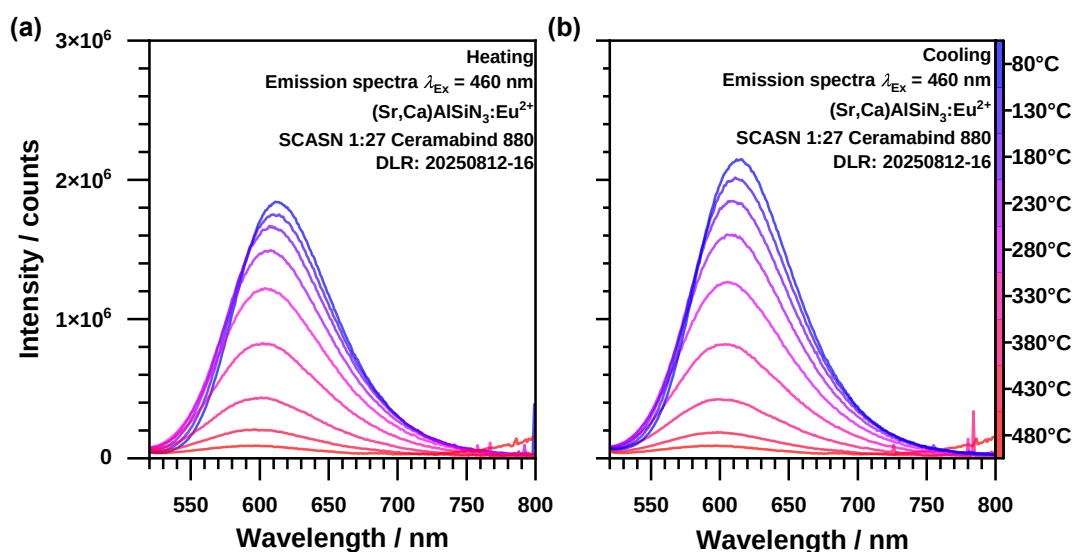


Figure 14: Emission spectra recorded upon heating (a) and cooling (b) of sample 20250812-16 (SCASN 1:27 Ceramabind[®] 880) in the temperature range from 80 to 480 °C at an excitation wavelength of 460 nm.

This behavior is also clearly reflected in the relative emission integrals shown in **figure 15**.

The calculated values of $\Delta I_{\text{rel, hc}}$ show that the relative change becomes increasingly positive toward lower temperatures and reaches up to 17.64 % at 80 °C, as shown in **table 5**.

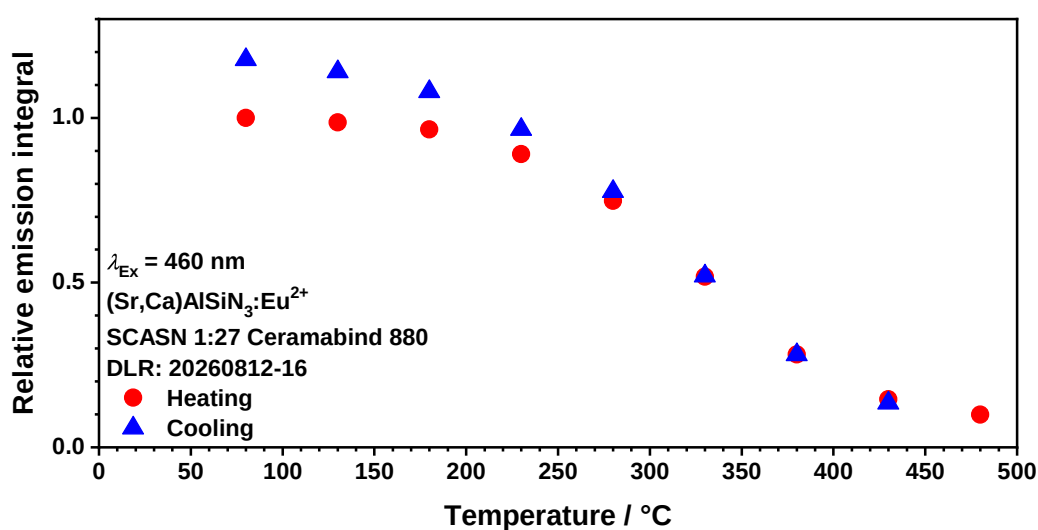


Figure 15: Relative emission integrals of sample 20250812-16 (SCASN 1:27 Ceramabind[®] 880) obtained from the emission spectra recorded upon heating and cooling in the temperature range from 80 to 480 °C at an excitation wavelength of 460 nm.

Table 5: Relative emission integrals of sample 20250812-16 (SCASN 1:27 Ceramabind[®] 880) obtained during heating and cooling are compared. The relative change $\Delta I_{\text{rel, hc}}$ is defined according to **equation (9a)**.

Temperatur / °C	Relative emission integral I_{heating}	Relative emission integral I_{cooling}	Relative change $\Delta I_{\text{rel, hc}} / \%$
80	1.000	1.176	17.64
130	0.986	1.140	15.60
180	0.965	1.079	11.86
230	0.890	0.965	8.39
280	0.748	0.777	3.86
330	0.518	0.520	0.23
380	0.282	0.281	-0.45
430	0.146	0.134	-8.13
480	0.099		

This behavior cannot be explained with certainty. One possible reason is that, during this particular measurement, the vacuum pump did not reduce the pressure inside the sample holder sufficiently, so that the sample may have been exposed to a substantially higher pressure, possibly even close to ambient pressure. Under such conditions, the degradation pathway

proposed above, namely thermally induced release of N₂ and N₂O, may have been suppressed. Instead, as discussed in more detail in **section 4.2.1**, the thermal treatment may even have led to a partial increase in emission intensity.

For the sake of clarity, the corresponding heating and cooling spectra of sample 20260129-2 (SCASN 1:4 Ceramabind[®] 880) are moved to the Appendix (see **figure A1**). Even though sample 20260129-2 was prepared in the same way as sample 20250812-15, including the phosphor concentration (see **section 3.1.2**), both samples behaved quite differently during thermal cycling. In the case of sample 20260129-2, visible damage to the coating surface occurred, including severe delamination and microcrack formation (see **figure A11**), the possible reasons for this behavior will be discussed later in **section 4.2.3**. The corresponding values are therefore included only for completeness, since the measured intensity is influenced not only by possible thermal degradation but also by mechanical damage to the coating. Nevertheless, **figure 16** shows the relative emission integrals of sample 20260129-2 during heating and cooling in the temperature range from 24 to 400 °C, again revealing lower intensities during cooling.

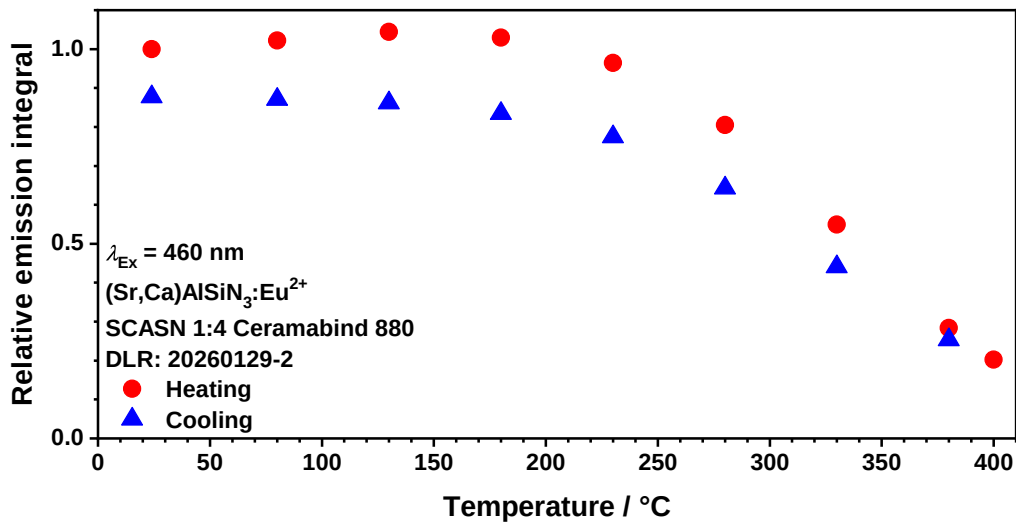


Figure 16: Relative emission integrals of sample 20260129-2 (SCASN 1:4 Ceramabind[®] 880) obtained from the emission spectra recorded upon heating and cooling in the temperature range from 24 to 400 °C at an excitation wavelength of 460 nm.

For this sample, the calculated values of $\Delta I_{\text{rel, hc}}$ are negative over the entire investigated range and lie between -10.53 to -20.16 % (see **table 6**).

Table 6: Relative emission integrals of sample 20260129-2 (SCASN 1:4 Ceramabind® 880) obtained during heating and cooling are compared. The relative change $\Delta I_{\text{rel, hc}}$ is defined according to **equation (9a)**.

Temperatur / °C	Relative emission integral I_{heating}	Relative emission integral I_{cooling}	Relative change $\Delta I_{\text{rel, hc}} / \%$
24	1.000	0.877	-12.29
80	1.022	0.871	-14.79
130	1.044	0.861	-17.49
180	1.029	0.835	-18.90
230	0.965	0.774	-19.76
280	0.805	0.643	-20.16
330	0.549	0.441	-19.67
380	0.283	0.254	-10.53
400	0.203		

Likewise, the heating and cooling spectra of sample 20260129-1 (SCASN 1:27 Ceramabind® 880) are shown in the Appendix for the sake of clarity (see **figure A2**). Although sample 20260129-1 was prepared analogously to sample 20250812-16, including the phosphor concentration (see **section 3.1.2**), its behavior during thermal cycling differed markedly. Again, visible coating damage occurred in the form of partial delamination and microcracks (see **figure A12**), the possible reasons for this behavior will again be discussed later in **section 4.2.3**. For this reason, the corresponding values are included only for completeness and should not be interpreted in the same way as those of the undamaged coatings. Still, **figure 17** shows that the deviation between heating and cooling is even more pronounced for this sample.

Here, the calculated values of $\Delta I_{\text{rel, hc}}$ remain negative throughout the full temperature range and vary between -15.57 to -34.14 % (see **table 7**). Whether this stronger deviation is primarily caused by more severe mechanical damage, by an increased susceptibility of the lower-loaded coating to degradation, or by a combination of both effects cannot be decided with certainty on the basis of the present data.

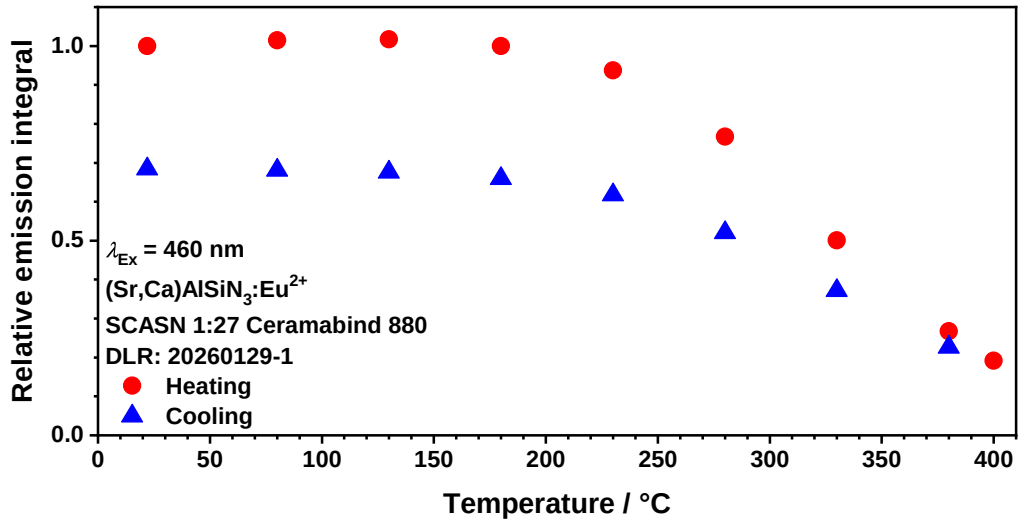


Figure 17: Relative emission integrals of sample 20260129-1 (SCASN 1:27 Ceramabind® 880) obtained from the emission spectra recorded upon heating and cooling in the temperature range from 22 to 400 °C at an excitation wavelength of 460 nm.

Table 7: Relative emission integrals of sample 20260129-1 (SCASN 1:27 Ceramabind® 880) obtained during heating and cooling are compared. The relative change $\Delta I_{\text{rel, hc}}$ is defined according to **equation (9a)**.

Temperatur / °C	Relative emission integral I_{heating}	Relative emission integral I_{cooling}	Relative change $\Delta I_{\text{rel, hc}} / \%$
22	1.000	0.684	−31.60
80	1.014	0.680	−32.93
130	1.017	0.676	−33.54
180	1.000	0.659	−34.09
230	0.937	0.617	−34.14
280	0.767	0.521	−32.14
330	0.501	0.372	−25.83
380	0.267	0.226	−15.57
400	0.191		

Overall, all heating-cooling comparisons except that of sample 20250812-16 show a pronounced hysteresis in emission intensity, even within the reduced temperature interval up to 380 °C or 400 °C. Taken together, these results indicate that intensity-based readout is not sufficiently reliable for measurements involving both heating and cooling cycles. By contrast, an intensity-

based approach may still be feasible when the measurement is restricted to heating only and no subsequent cooling branch has to be evaluated.

Thermal Degradation

Analogous to the investigation of the SCASN phosphor powder, the thermal degradation behavior of a coated SCASN sample was examined as well. For this purpose, the coated sample was heated in air for 2 h at 100 °C, after which the emission spectrum was recorded at room temperature. The sample was then heated again for 2 h at 200 °C, measured once more, and this procedure was continued stepwise up to 600 °C. After each heating step, the sample was removed from the drying oven or furnace and allowed to cool naturally to room temperature.

The resulting emission spectra are shown in **figure 18(a)**, while the corresponding relative emission integrals are given in **figure 18(b)**. Up to 300 °C, no significant change in either the spectral shape or the relative emission integral is observed. After thermal treatment at 400 °C for 2 h, however, the emission intensity increases markedly. At higher temperatures, this behavior reverses, and after treatment at 500 °C the intensity decreases strongly, indicating that irreversible thermal degradation has set in under these conditions.

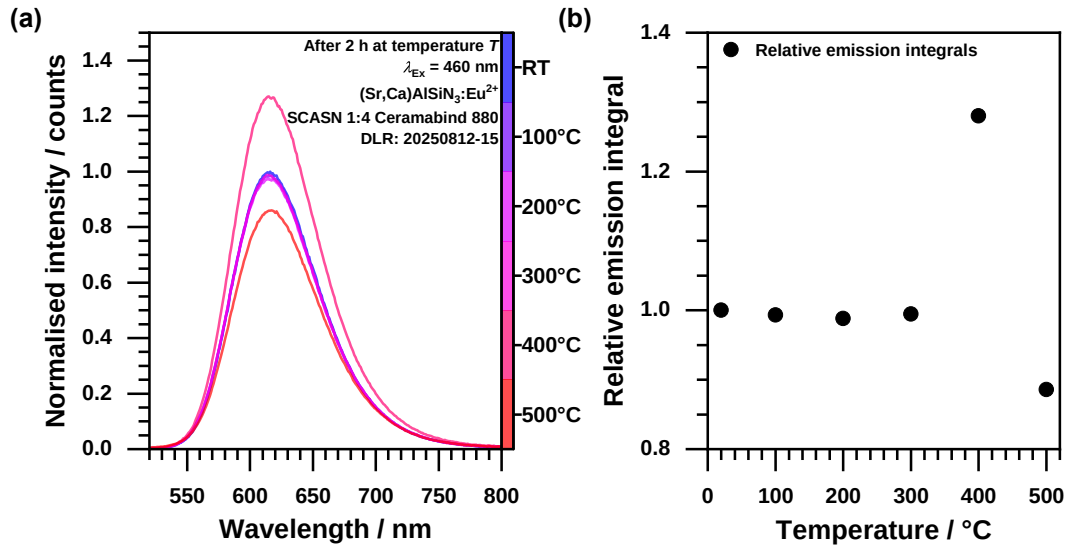


Figure 18: Emission spectra measured at room temperature after thermal treatment for 2 h at the indicated temperatures (a) and the corresponding relative emission integrals (b) of sample 20250812-15 (SCASN 1:4 Ceramabind® 880) at an excitation wavelength of 460 nm.

The pronounced increase of 28.0 % after 2 h at 400 °C is particularly noteworthy. To exclude a measurement artifact, a second piece of sample 20250812-15 was measured at room temperature and then remeasured after the same thermal treatment at 400 °C for 2 h. In this

repeat experiment, an increase of 26.5 % was obtained. Since this value is very close to the initially observed increase, the effect is considered reproducible and is therefore not attributed to measurement error.

A plausible explanation is the formation of a thin silica- or silicate-rich surface layer on the phosphor particles during thermal treatment. Such an interfacial layer may originate from the Ceramabind[®] binder, which is based on a silicone resin, and may therefore generate SiO₂-containing species upon heating. In addition, thermally induced reaction products of the phosphor itself cannot be excluded. For CASN, the formation of Ca₂Si₂AlO₇ during heat treatment has been reported [79]. Moreover, under conditions involving hydrolysis, phases such as CaAl₂Si₂O₈ and SrAl₂SiO₈ have been discussed for related systems [18].

Such a surface layer could improve transmission and reducing reflection losses at the particle-matrix interface. This interpretation is supported by literature reports showing that silica-based surface modifications or related interfacial layers can increase the observable emission intensity [81, 82], even explicitly for SCASN [83]. This explanation is also physically plausible in view of the refractive indices involved. SCASN has a comparatively high refractive index of about 2.2 [84], whereas SiO₂ exhibits a much lower value of about 1.46 [85]. Likewise, CaAl₂Si₂O₈ shows a refractive index of roughly 1.6 [86]. The formation of such a lower-index interfacial layer could therefore enhance transmission at the particle-matrix interface, thereby improving both excitation-light incoupling and emission-light outcoupling.

This interpretation may also help to rationalize the previously discussed behavior of sample 20250812-16 during heating and cooling, for which an increase in emission intensity during cooling was observed. In that case, it is possible that the vacuum pump did not operate as intended and that the sample holder was therefore closer to ambient pressure than expected. Under such conditions, thermal treatment may likewise have promoted the formation of a similar silica- or silicate-rich interfacial layer, thereby leading to the observed increase in emission intensity during cooling.

By contrast, this behavior was not observed for the other coated samples. A possible reason is that, under reduced pressure, the aforementioned thermally induced release of N₂ and N₂O may be accelerated, which would promote irreversible degradation and at the same time counteract or mask any beneficial effect of interfacial-layer formation. Accordingly, the final emission behavior of the coated samples may result from a competition between degradation processes and thermally induced formation of an optically favorable surface layer.

Alternative Substrate

Since a possible practical TSP application may involve complex models made of steel, steel was also tested as an alternative substrate. For the sake of clarity, the corresponding emission spectra of sample 20260129-2 on steel substrate (SCASN 1:4 Ceramabind[®] 880) recorded during heating and cooling are shown in the Appendix (see **figure A3**).

Sample 20260129-2 on steel was prepared analogously to the corresponding coating on aluminum substrate, with the substrate being the only intended difference. After thermal cycling, however, the coating slight microcrack formation (see **figure A13**), the possible reasons for this behavior will again be discussed later in **section 4.2.3**. The corresponding results should therefore be regarded primarily as qualitative, since the measured intensity is affected not only by the temperature-dependent luminescence behavior but also by mechanical damage of the coating.

Nevertheless, **figure 19** shows the relative emission integrals of sample 20260129-2 on steel substrate during heating and cooling in the temperature range from 25 to 400 °C. A clear deviation between the heating and cooling branches is observed, analogous to the behavior already discussed for the corresponding sample on aluminum substrate.

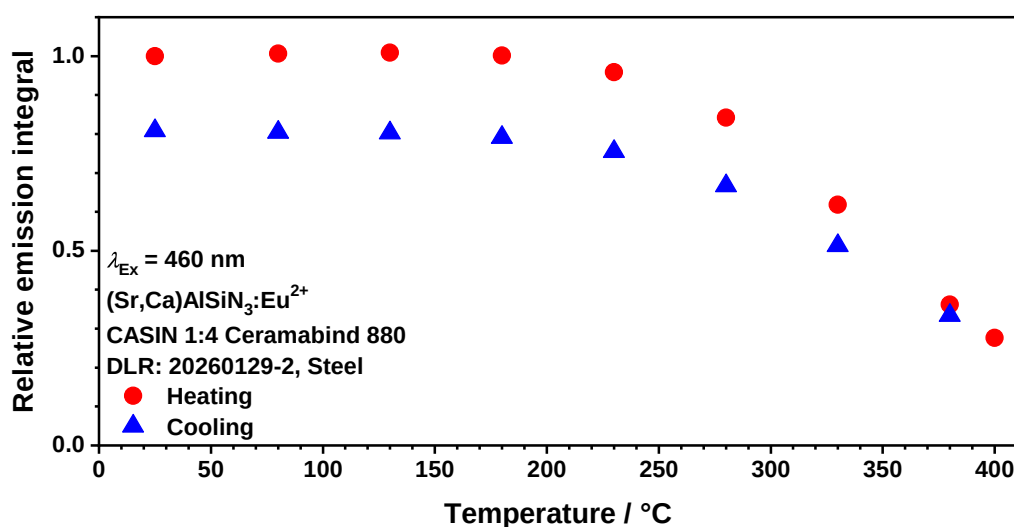


Figure 19: Relative emission integrals of sample 20260129-2 (SCASN 1:4 Ceramabind[®] 880, steel substrate) obtained from the emission spectra recorded upon heating and cooling in the temperature range from 25 to 400 °C at an excitation wavelength of 460 nm.

The values of the calculated relative change $\Delta I_{\text{rel, hc}}$ given in **table 8** remain negative over the entire investigated temperature range and lie between -7.76 to -21.26 %. This range is indeed very similar to that obtained for sample 20260129-2 on aluminum substrate, for which values

between –10.53 to –20.16 % were found.

Table 8: Relative emission integrals of sample 20260129-2 (SCASN 1:4 Ceramabind[®] 880, steel substrate) obtained during heating and cooling are compared. The relative change $\Delta I_{\text{rel, hc}}$ is defined according to **equation (9a)**.

Temperatur / °C	Relative emission integral I_{heating}	Relative emission integral I_{cooling}	Relative change $\Delta I_{\text{rel, hc}} / \%$
25	1.000	0.808	–19.19
80	1.006	0.804	–20.09
130	1.009	0.802	–20.47
180	1.001	0.791	–21.04
230	0.959	0.755	–21.26
280	0.842	0.667	–20.75
330	0.618	0.513	–16.95
380	0.361	0.333	–7.76
400	0.276		

The same comparison was also carried out for sample 20260129-1 on steel substrate (SCASN 1:27 Ceramabind[®] 880). For the sake of clarity, the corresponding emission spectra of sample 20260129-1 on steel substrate (SCASN 1:4 Ceramabind[®] 880) recorded during heating and cooling are shown in the Appendix (see **figure A4**).

Again, the coating on steel was prepared analogously to the corresponding coating on aluminum substrate, with the substrate being the only intended difference. After thermal cycling, however, partial delamination and visible microcrack formation were again observed (see **figure A14**), the possible reasons for this behavior will again be discussed later in **section 4.2.3**. For this reason, these results should likewise be interpreted only with caution, since the measured intensity is influenced by both thermal effects and coating damage.

Nevertheless, **figure 20** shows the relative emission integrals of sample 20260129-1 on steel substrate during heating and cooling in the temperature range from 25 to 400 °C. Here as well, a pronounced deviation between the heating and cooling branches is observed.

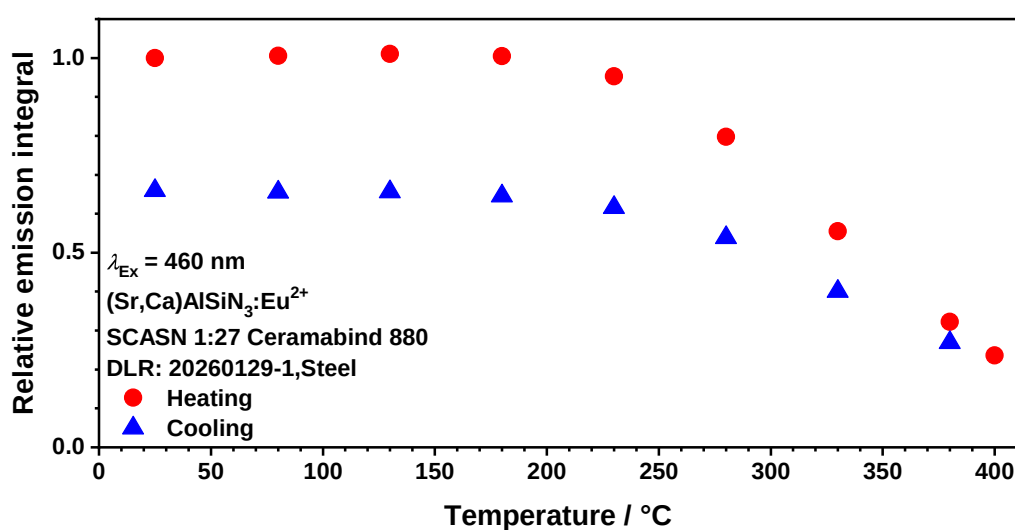


Figure 20: Relative emission integrals of sample 20260129-1 (SCASN 1:27 Ceramabind® 880, steel substrate) obtained from the emission spectra recorded upon heating and cooling in the temperature range from 25 to 400 °C at an excitation wavelength of 460 nm.

The calculated values of $\Delta I_{\text{rel, hc}}$ listed in **table 9** reveal a strongly negative relative change throughout the full temperature range, namely between -16.62 to -35.78 %. This is again very similar to the corresponding aluminum-based sample 20260129-1, for which values between -15.57 to -34.14 % were obtained.

Table 9: Relative emission integrals of sample 20260129-1 (SCASN 1:27 Ceramabind® 880, steel substrate) obtained during heating and cooling are compared. The relative change $\Delta I_{\text{rel, hc}}$ is defined according to **equation (9a)**.

Temperatur / °C	Relative emission integral I_{heating}	Relative emission integral I_{cooling}	Relative change $\Delta I_{\text{rel, hc}}$ / %
25	1.000	0.659	-34.07
80	1.006	0.655	-34.89
130	1.010	0.656	-35.04
180	1.005	0.645	-35.78
230	0.953	0.616	-35.38
280	0.798	0.538	-32.49
330	0.555	0.401	-27.74
380	0.322	0.268	-16.62

Temperatur / °C	Relative emission integral I_{heating}	Relative emission integral I_{cooling}	Relative change $\Delta I_{\text{rel, hc}} / \%$
400	0.236		

Overall, these initial tests indicate that the temperature-dependent behavior on steel substrate is broadly comparable to that observed for aluminum substrate. At the same time, the significance of these results remains limited, because all investigated coatings on steel showed partial delamination or microcracking. Consequently, a rigorous substrate-to-substrate comparison is not yet possible on the basis of the present measurements alone.

Alternative Integration Interval

One possible way to improve the temperature sensitivity of the intensity-based approach is to use a different integration interval, thereby making deliberate use of the blue shift of the emission band. As shown in **figure 10** for sample 20250812-15, the relative emission integrals remain nearly constant in the temperature range from 28 to 180 °C, even though the emission maximum itself already shows a visible decrease in intensity. This behavior can be attributed to the combined effects of band broadening and the temperature-dependent blue shift discussed above. If the full emission band is integrated, these spectral changes partly compensate each other. By contrast, when the integration interval is restricted to 600 to 800 nm, part of the short-wavelength, high-energy side of the emission band is excluded. As the band shifts to shorter wavelengths with increasing temperature, a larger fraction of the emission falls outside the integration window, which leads to a more pronounced decrease in the integrated intensity.

An example of this approach is shown for sample 20250812-15 in **figure 21**. Here, the same measurement as in **figure 10** was evaluated using an integration interval from 600 to 800 nm instead of 520 to 800 nm. As shown in **figure 21(b)**, the relative emission integrals now exhibit a noticeable decrease already in the lower-temperature region from 28 to 180 °C.

To quantify this effect, the relative temperature sensitivity $S_{r,I}$ was calculated for both integration intervals in this temperature range. For the interval from 520 to 800 nm, $S_{r,I}$ amounts to 0.02 % K⁻¹. For the interval from 600 to 800 nm, in contrast, $S_{r,I}$ increases to 0.12 % K⁻¹. Thus, although the absolute sensitivity remains comparatively low, it is enhanced by a factor of six in this temperature range.

For the full temperature range from 28 to 480 °C, the effect is much less pronounced. Here, $S_{r,I}$ increases only slightly, namely from 0.39 to 0.42 % K⁻¹.

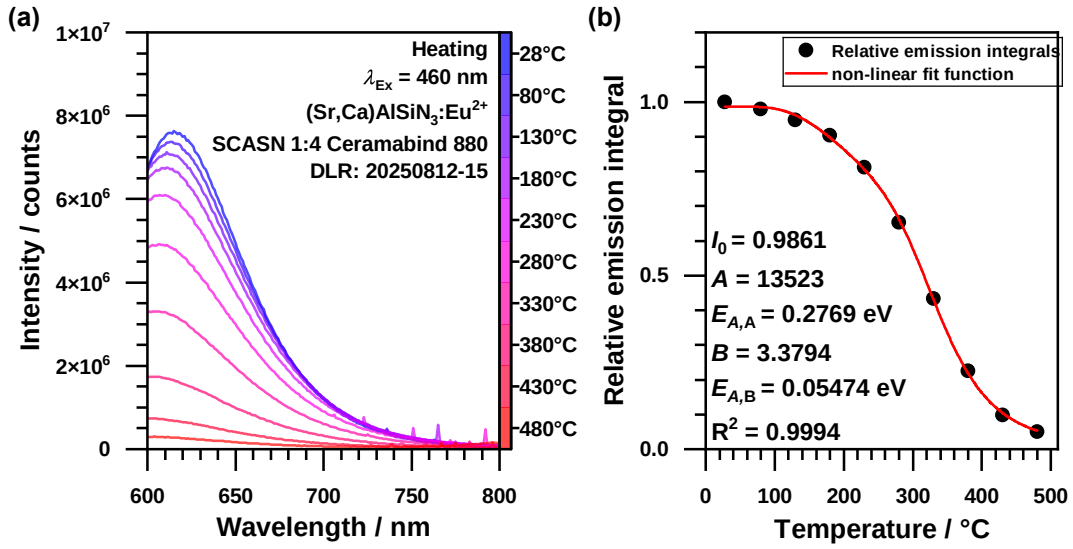


Figure 21: Emission spectra recorded upon heating (a) and the corresponding relative emission integrals together with the non-linear fit function (b) of sample 20250812-15 (SCASN 1:4 Ceramabind[®] 880) in the temperature range from 28 to 480 °C at an excitation wavelength of 460 nm, using an integration interval from 600 to 800 nm.

It should be noted that, for this evaluation, a different fit function was used. This was necessary because the resulting temperature dependence does not follow a classical sigmoidal shape, that is, it does not exhibit a clear upper and lower plateau to which the relative emission integrals converge. Instead, the following expression was used as a purely phenomenological fit function. It is important to emphasize that this approach does not imply the presence of two distinct non-radiative recombination channels. Although equations of this type are commonly applied to such cases [87], here they are used solely to achieve an adequate phenomenological description of the measured data.

$$I_T = \frac{I_0}{1 + A \exp\left(-\frac{E_{A,A}}{kT}\right) + B \exp\left(-\frac{E_{A,B}}{kT}\right)} \quad (10)$$

with:

- I_T : Emission integral at temperature T
- I_0 : Emission integral at temperature $T = 0$ K
- A : Dimensionless phenomenological fit parameter of contribution A
- B : Dimensionless phenomenological fit parameter of contribution B
- $E_{A,A}$: Effective activation energy associated with phenomenological contribution A / eV
- $E_{A,B}$: Effective activation energy associated with phenomenological contribution B / eV
- k : Boltzmann constant (8.6173×10^{-5} eV/K)
- T : Temperature

Overall, the use of an alternative integration interval proved to be a promising way to improve the intensity-based readout. Most notably, the temperature sensitivity $S_{r,I}$ could be increased by a factor of six in the temperature range from 28 to 180 °C, which had previously shown only very weak temperature dependence. From a practical point of view, this concept could be implemented easily by placing a 600 nm long-pass filter in front of the detector. It therefore represents an effective and experimentally accessible strategy to improve the intensity-based approach.

4.2.2 Temperature-Dependent Decay Curves

As the second step in assessing the temperature-dependent behavior of the coated samples, temperature-dependent decay curves were recorded in order to evaluate the suitability of the coatings for lifetime-based readout. For all samples, an excitation wavelength of 445 nm was selected because this was the available pulsed laser diode operating in the picosecond range closest to the previously used excitation wavelength of 460 nm. At the same time, SCASN still shows comparably strong excitation at 445 nm. Likewise, for all samples an emission wavelength of 606 nm was chosen, since this is the wavelength at which the highest SCASN emission intensity was measured before correction for the detector sensitivity.

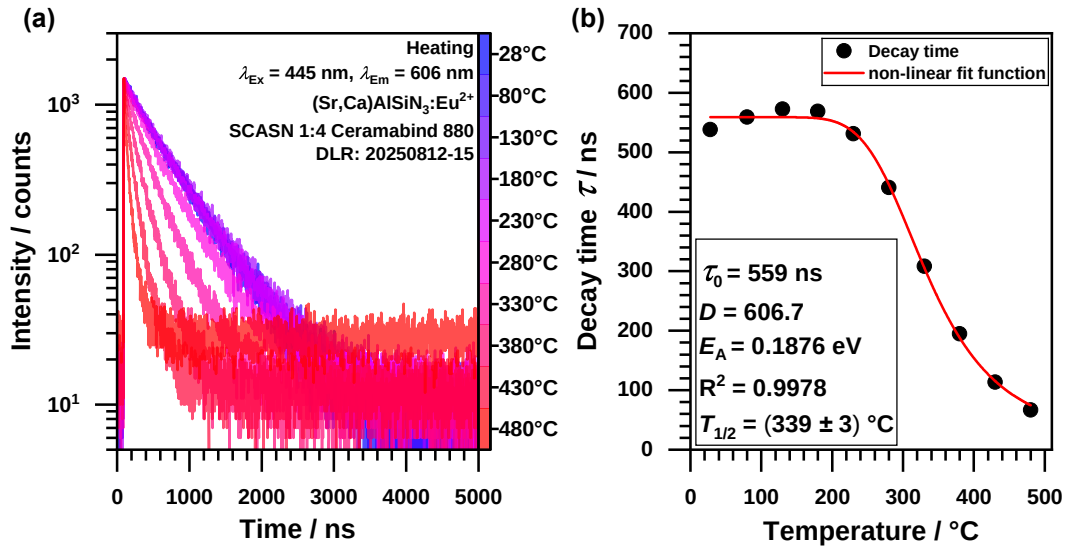


Figure 22: Decay curves recorded upon heating (a) and the corresponding decay times together with the non-linear fit function (b) of sample 20250812-15 (SCASN 1:4 Ceramabind[®] 880) in the temperature range from 28 to 480 °C at an excitation wavelength of 445 nm and a monitored emission wavelength of 606 nm. For clarity, error bars for the decay times are omitted, as the corresponding fit standard deviations are smaller than the symbol size.

Figure 22(a) shows the temperature-dependent decay curves recorded upon heating sample

20250812-15 (SCASN 1:4 Ceramabind® 880) in the temperature range from 28 to 480 °C, whereas **figure 22(b)** presents the corresponding decay times together with a non-linear fit based on **equation (4b)**. With increasing temperature, a pronounced shortening of the decay time is observed, which is consistent with thermally activated non-radiative relaxation processes. All decay curves, including those of the following samples unless stated otherwise, exhibit mono-exponential behavior and were therefore fitted accordingly. The resulting decay times are listed in **table 10**.

Table 10: Decay times of sample 20250812-15 (SCASN 1:4 Ceramabind® 880) obtained from the decay curves recorded upon heating in the temperature range from 28 to 480 °C are reported. The corresponding reduced χ^2 values of the fits are listed.

Temperatur / °C	Decay time τ / ns	Reduced χ^2
28	538 ± 1	1.284
80	559 ± 2	1.188
130	572 ± 2	1.209
180	568 ± 2	1.135
230	531 ± 1	1.184
280	440 ± 1	1.184
330	308 ± 1	1.305
380	195 ± 1	1.437
430	114 ± 1	1.415
480	67 ± 1	1.247

A noteworthy feature is the initial increase in decay time between 28 to 130 °C, before the expected decrease caused by thermal quenching sets in. This behavior has previously been reported for a few Eu²⁺-activated phosphors and is commonly attributed to thermal population of higher-lying excited 5d states with different spin character [36, 88]. According to this interpretation, thermally populated sextet-like excited states show lower radiative transition probabilities to the ⁸S_{7/2} ground state than the lower-lying octet-like states, which results in longer apparent decay times [36, 88]. As discussed earlier in **section 2.5**, the excited 5d state of Eu²⁺ indeed comprises states of mixed sextet and octet character. From a practical perspective, however, this initial increase is problematic, because similar decay times occur again at higher temperatures during the subsequent decrease. For example, the decay times at 28 and around 230 °C are of comparable magnitude and therefore cannot be uniquely distinguished by lifetime-based readout alone. Accordingly, either an additional complementary measurement, for example an intensity-based readout or a simple infrared thermometer, would be required to identify the relevant

temperature regime, or the experimental setup would need to be restricted to temperatures above about 130 °C, where the monotonic decrease in decay time begins.

Nevertheless, the non-linear fit based on the *Struck-Fonger* model reproduces the overall temperature dependence of the decay times well. As discussed earlier, this fit allows the quenching temperature $T_{1/2}$ to be determined, that is, the temperature at which the decay time decreases to 50 % of the value extrapolated for $T = 0$ K. For sample 20250812-15, a value of (339 ± 3) °C was obtained. The relative temperature sensitivity $S_{r,\tau}$ was calculated to be $0.34 \% K^{-1}$ in the temperature range from 28 to 480 °C.

Figure 23 shows the same type of data for sample 20250812-16 (SCASN 1:27 Ceramabind[®] 880), that is, for a coating with a lower phosphor concentration. Overall, the same temperature-dependent trends are observed. Again, the decay time first increases up to about 130 °C and then decreases markedly as thermal quenching becomes dominant. The corresponding decay times are given in **table 11**.

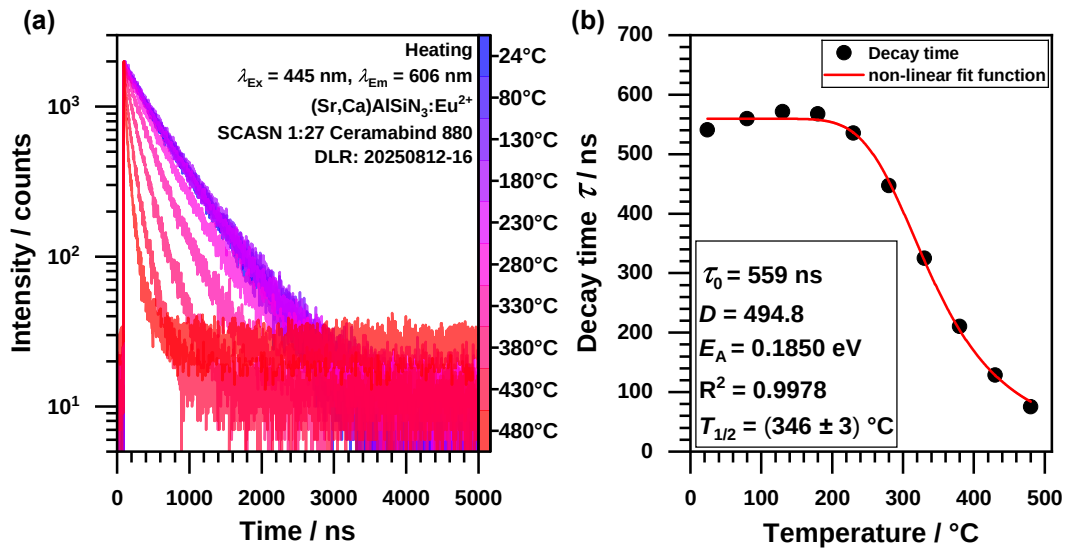


Figure 23: Decay curves recorded upon heating (a) and the corresponding decay times together with the non-linear fit function (b) of sample 20250812-16 (SCASN 1:27 Ceramabind[®] 880) in the temperature range from 24 to 480 °C at an excitation wavelength of 445 nm and a monitored emission wavelength of 606 nm. For clarity, error bars for the decay times are omitted, as the corresponding fit standard deviations are smaller than the symbol size.

Table 11: Decay times of sample 20250812-16 (SCASN 1:27 Ceramabind® 880) obtained from the decay curves recorded upon heating in the temperature range from 24 to 480 °C are reported. The corresponding reduced χ^2 values of the fits are listed.

Temperatur / °C	Decay time τ / ns	Reduced χ^2
24	541 ± 1	1.288
80	560 ± 1	1.287
130	572 ± 1	1.245
180	567 ± 1	1.164
230	535 ± 1	1.221
280	447 ± 1	1.208
330	325 ± 1	1.266
380	210 ± 1	1.464
430	128 ± 1	1.652
480	75 ± 1	1.347

The $T_{1/2}$ value determined from the non-linear fit is (346 ± 3) °C. This value is in good agreement with that of sample 20250812-15, for which $T_{1/2} = (339 \pm 3)$ °C was obtained. The relative temperature sensitivity $S_{r,\tau}$ was calculated to be $0.33 \% K^{-1}$ in the temperature range from 24 to 480 °C. Thus, the value is nearly identical to that of sample 20250812-15.

Comparison of Different Phosphor Concentrations

Since both coatings exhibit very similar temperature-dependent behavior with respect to the decay time, a direct comparison between the two phosphor concentrations is useful. For this purpose, the relative change between the decay times of both samples ($\Delta\tau_{\text{rel}, 21}$) was calculated according to **equation (11)**:

$$\Delta\tau_{\text{rel}, 21} = \frac{\tau_2 - \tau_1}{(\tau_1 + \tau_2) / 2} \cdot 100 \quad (11)$$

with:

- $\Delta\tau_{\text{rel}, 21}$: Relative change of the decay times of sample 2 and sample 1 / %
- τ_2 : Decay time of sample 2
- τ_1 : Decay time of sample 1

Here, the relative change was intentionally calculated symmetrically, that is, normalized to the mean value of both samples. This definition emphasizes the deviation from a common average and is therefore appropriate because the decay times of both samples are very similar over most of the investigated temperature range. The resulting values are presented in **table 12**.

Table 12: Decay times of samples 20250812-15 (SCASN 1:4 Ceramabind[®] 880) and 20250812-16 (SCASN 1:27 Ceramabind[®] 880), together with the corresponding relative change, are compared. The relative change $\Delta\tau_{\text{rel}, 21}$ is defined according to **equation (11)**.

Temperature / °C	Decay time τ_1 / ns	Decay time τ_2 / ns	Relative change
	20250812-15	20250812-16	$\Delta\tau_{\text{rel}, 21}$ / %
28 / 24	538 ± 1	541 ± 1	0.47
80	559 ± 2	560 ± 1	0.08
130	572 ± 2	572 ± 1	-0.13
180	568 ± 2	567 ± 1	-0.29
230	531 ± 1	535 ± 1	0.80
280	440 ± 1	447 ± 1	1.54
330	308 ± 1	325 ± 1	5.19
380	195 ± 1	210 ± 1	7.55
430	114 ± 1	128 ± 1	12.19
480	67 ± 1	75 ± 1	11.95

It becomes evident that the decay times remain remarkably similar up to about 280 °C, where the relative change reaches at most 1.54 %. At higher temperatures, the deviation increases and reaches 12.19 % at 430 °C, indicating that the decay time of sample 20250812-16 becomes noticeably longer than that of sample 20250812-15. At the same time, it should be kept in mind that the absolute decay times decrease strongly in this temperature range. Consequently, even comparatively small absolute differences translate into clearly larger relative deviations.

Overall, these results indicate that a substantial reduction in phosphor concentration is possible without significantly affecting either $T_{1/2}$ or $S_{r,\tau}$ in lifetime-based measurements. Within the range up to about 280 °C, the lower phosphor loading therefore appears fully feasible. This also highlights one of the central advantages of a lifetime-based readout method, namely its comparatively low sensitivity to phosphor concentration and, more generally, to moderate variations in signal amplitude. Above 280 °C, however, the deviations between both coatings become more pronounced, so that the quantitative agreement is clearly reduced compared with the lower-temperature range.

Comparison of Heating and Cooling

As in the case of the intensity-based evaluation, the comparison between heating and cooling is also important for the lifetime-based approach, because possible deviations between both branches would indicate hysteresis.

For sample 20250812-15 (SCASN 1:4 Ceramabind[®] 880), the decay curves recorded during heating and cooling are shown in **figure 24**. Due to technical problems with the heated sample holder, this comparison could again only be carried out in the temperature range from 24 to 380 °C. No obvious differences between the heating and cooling decay curves are visible.

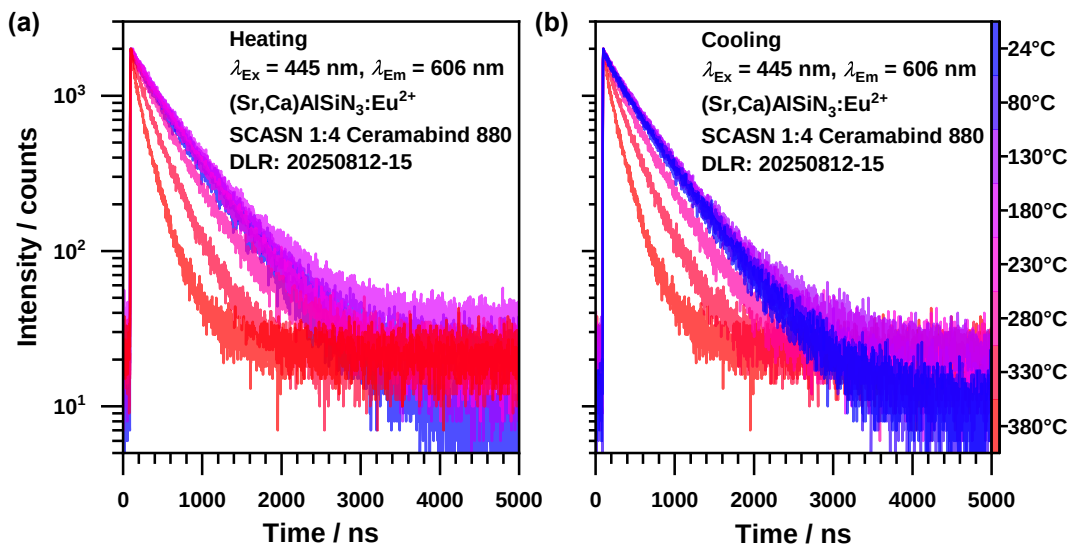


Figure 24: Decay curves recorded upon heating (a) and cooling (b) of sample 20250812-15 (SCASN 1:4 Ceramabind[®] 880) in the temperature range from 24 to 380 °C at an excitation wavelength of 445 nm and a monitored emission wavelength of 606 nm.

To visualize possible deviations more clearly, the corresponding decay times obtained during heating and cooling are plotted as a function of temperature in **figure 25**. Only very small differences become apparent. The corresponding values of $\Delta\tau_{\text{rel, hc}}$, calculated according to **equation (9b)**, are listed in **table 13**. For sample 20250812-15, they lie between -1.88 to 0.17 %. This range is well within the expected measurement uncertainty and therefore does not indicate any significant hysteresis effect.

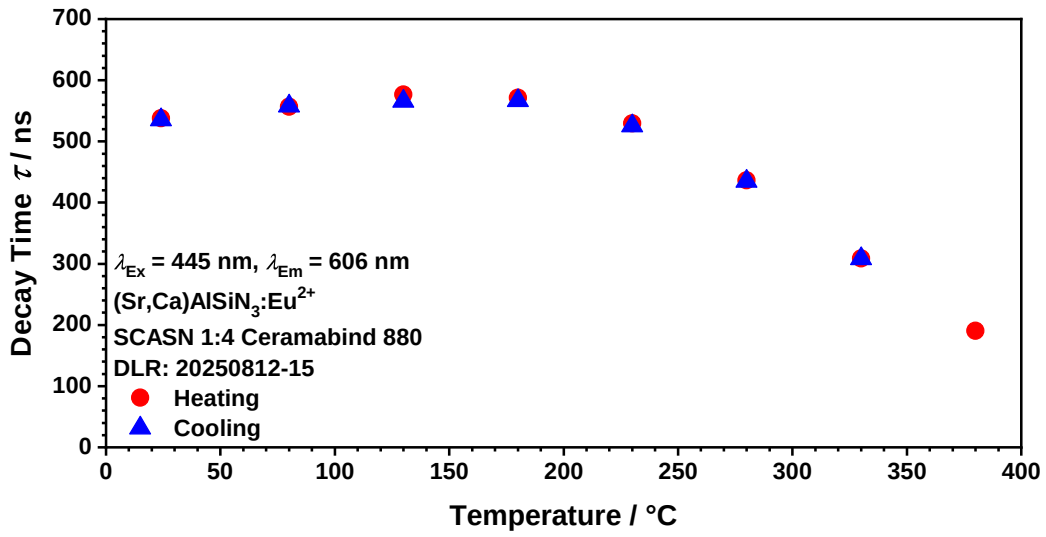


Figure 25: Decay times of sample 20250812-15 (SCASN 1:4 Ceramabind® 880) obtained from the decay curves recorded upon heating and cooling in the temperature range from 24 to 380 °C at an excitation wavelength of 445 nm and a monitored emission wavelength of 606 nm. For clarity, error bars for the decay times are omitted, as the corresponding fit standard deviations are smaller than the symbol size.

Table 13: Decay times of sample 20250812-15 (SCASN 1:4 Ceramabind® 880) obtained during heating and cooling, together with the corresponding relative change, are compared. The relative change $\Delta\tau_{\text{rel, hc}}$ is defined according to **equation (9b)**. The corresponding reduced χ^2 values of the fits are listed for heating and cooling.

Temperatur / °C	Decay time $\tau_{\text{heating}} / \text{ns}$	Decay time $\tau_{\text{cooling}} / \text{ns}$	Relative change $\Delta\tau_{\text{rel, hc}} / \%$	Reduced χ^2 (heating / cooling)
24	538 ± 1	535 ± 1	−0.49	1.333 / 1.309
80	557 ± 1	558 ± 1	0.17	1.154 / 1.196
130	576 ± 1	566 ± 1	−1.88	1.168 / 1.163
180	571 ± 2	567 ± 1	−0.85	1.298 / 1.149
230	530 ± 1	525 ± 1	−0.88	1.223 / 1.126
280	436 ± 1	435 ± 1	−0.31	1.243 / 1.227
330	309 ± 1	308 ± 1	−0.28	1.314 / 1.312
380	191 ± 1	—	—	1.444 / —

The same procedure was applied to sample 20250812-16 (SCASN 1:27 Ceramabind® 880) in the temperature range from 24 to 480 °C. In contrast to sample 20250812-15, **figure 26** reveals

a pronounced fast initial decay component at 24 °C during cooling. This feature will be discussed again in the following subsection dealing with thermal degradation.

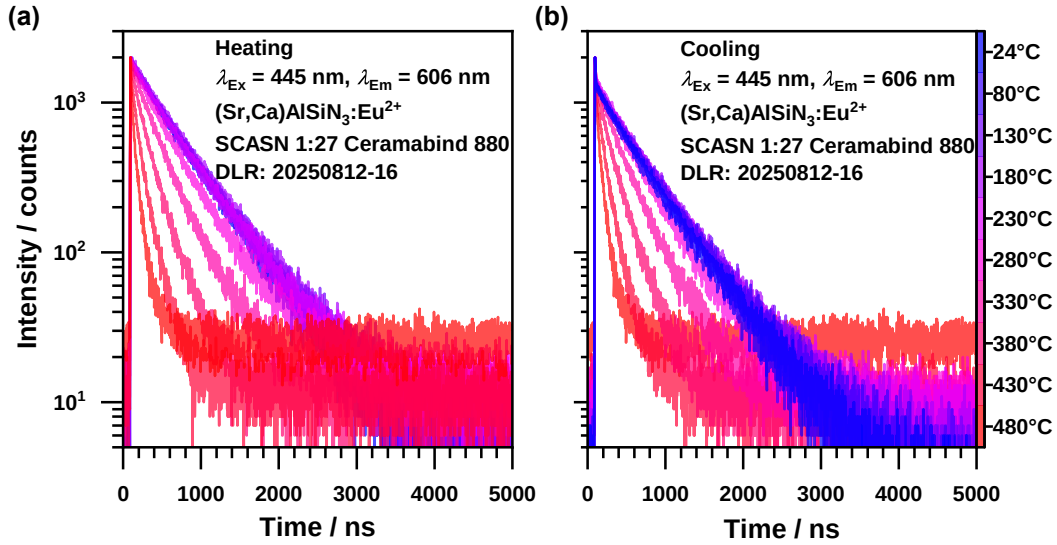


Figure 26: Decay curves recorded upon heating (a) and cooling (b) of sample 20250812-16 (SCASN 1:27 Ceramabind[®] 880) in the temperature range from 24 to 480 °C at an excitation wavelength of 445 nm and a monitored emission wavelength of 606 nm.

Despite this additional early-time component, the standardized mono-exponential evaluation of the decay curves shows essentially no relevant difference between heating and cooling, as illustrated in **figure 27**.

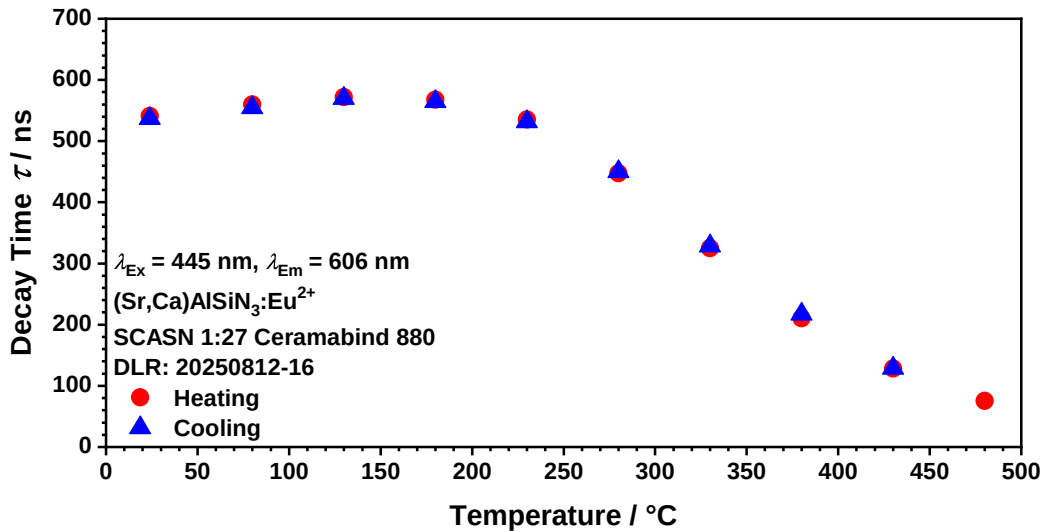


Figure 27: Decay times of sample 20250812-16 (SCASN 1:27 Ceramabind[®] 880) obtained from the decay curves recorded upon heating and cooling in the temperature range from 24 °C to 480 °C at an excitation wavelength of 445 nm and a monitored emission wavelength of 606 nm. For clarity, error bars for the decay times are omitted, as the corresponding fit standard deviations are smaller than the symbol size.

This is supported by the resulting values of $\Delta\tau_{\text{rel, hc}}$ listed in **table 14**, which lie in a narrow range from -0.96 to 1.10 %, except for the value at 380 °C, where $\Delta\tau_{\text{rel, hc}} = 3.34$ %. Since all other values remain within the expected uncertainty range, this isolated deviation is most reasonably regarded as an outlier. It is nevertheless worth noting that the pronounced fast initial decay component is observed only for the lower phosphor concentration, which may indicate that this behavior is indeed related to the reduced phosphor loading, as already suggested above.

Table 14: Decay times of sample 20250812-16 (SCASN 1:27 Ceramabind® 880) obtained during heating and cooling, together with the corresponding relative change, are compared. The relative change $\Delta\tau_{\text{rel, hc}}$ is defined according to **equation (9b)**. The corresponding reduced χ^2 values of the fits are listed for heating and cooling.

Temperatur / °C	Decay time $\tau_{\text{heating}} / \text{ns}$	Decay time $\tau_{\text{cooling}} / \text{ns}$	Relative change $\Delta\tau_{\text{rel, hc}} / \%$	Reduced χ^2 (heating / cooling)
24	541 ± 1	537 ± 1	-0.77	1.288 / 1.290
80	560 ± 1	554 ± 2	-0.96	1.287 / 1.289
130	572 ± 1	570 ± 2	-0.31	1.245 / 1.189
180	567 ± 1	565 ± 2	-0.37	1.164 / 1.203
230	535 ± 1	531 ± 1	-0.74	1.221 / 1.137
280	447 ± 1	450 ± 1	0.53	1.208 / 1.244
330	325 ± 1	328 ± 1	1.10	1.266 / 1.256
380	210 ± 1	217 ± 1	3.34	1.464 / 1.472
430	128 ± 1	129 ± 1	0.31	1.652 / 1.397
480	75 ± 1			1.347 / —

As discussed for the intensity-based measurements, both 20260129 samples showed partial delamination and visible microcrack formation after thermal cycling (see **figures A11 and A12**), the possible reasons for this behavior will again be discussed later in **section 4.2.3**. Their results should therefore be interpreted with some caution. Even so, the lifetime-based evaluation remains remarkably robust.

For sample 20260129-2 (SCASN 1:4 Ceramabind® 880), the corresponding decay curves are shown in the Appendix (see **figure A5**), while the decay times are plotted in **figure 28**. Only very small differences between heating and cooling are observed.

The corresponding values of $\Delta\tau_{\text{rel, hc}}$, listed in **table 15**, lie between -1.83 to 1.68 %. Thus, even in the presence of visible coating damage, no meaningful hysteresis effect is detected. This

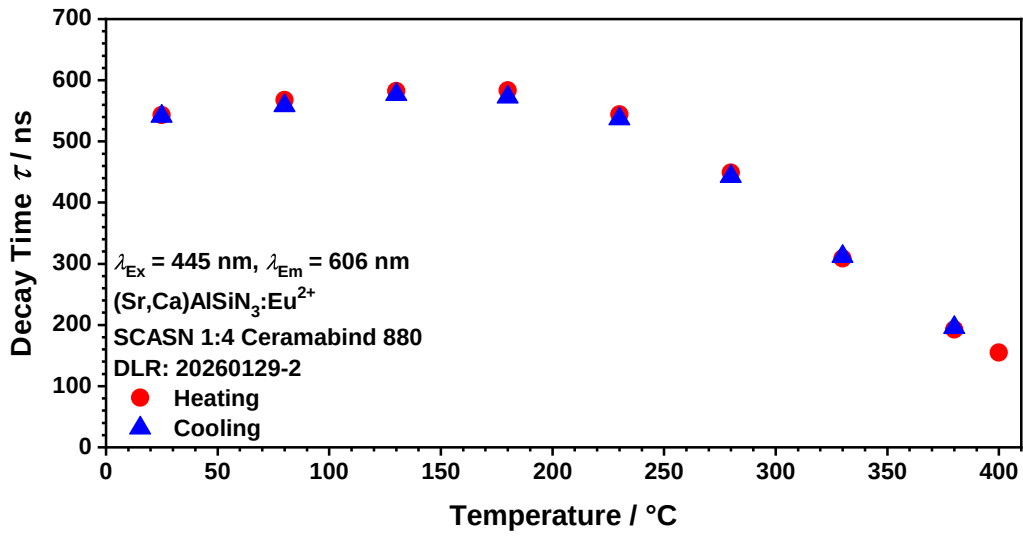


Figure 28: Decay times of sample 20260129-2 (SCASN 1:4 Ceramabind® 880) obtained from the decay curves recorded upon heating and cooling in the temperature range from 25 to 400 °C at an excitation wavelength of 445 nm and a monitored emission wavelength of 606 nm. For clarity, error bars for the decay times are omitted, as the corresponding fit standard deviations are smaller than the symbol size.

is an important result and again illustrates one of the strengths of lifetime-based thermometry. Even when the coating surface is not mechanically intact, the extracted decay times remain reproducible. Of course, for a spatially resolved TSP application, delamination and microcracks would still be highly problematic, because different parts of the surface would no longer behave uniformly. Nevertheless, compared with intensity-based readout, the lifetime-based method appears distinctly more robust.

Table 15: Decay times of sample 20260129-2 (SCASN 1:4 Ceramabind® 880) obtained during heating and cooling, together with the corresponding relative change, are compared. The relative change $\Delta\tau_{\text{rel, hc}}$ is defined according to **equation (9b)**. The corresponding reduced χ^2 values of the fits are listed for heating and cooling.

Temperatur / °C	Decay time $\tau_{\text{heating}} / \text{ns}$	Decay time $\tau_{\text{cooling}} / \text{ns}$	Relative change $\Delta\tau_{\text{rel, hc}} / \%$	Reduced χ^2 (heating / cooling)
25	543 ± 1	541 ± 1	-0.39	1.323 / 1.274
80	568 ± 1	558 ± 2	-1.66	1.288 / 1.268
130	582 ± 1	577 ± 1	-0.99	1.175 / 1.210
180	583 ± 2	573 ± 2	-1.83	1.097 / 1.146
230	544 ± 1	537 ± 1	-1.40	1.148 / 1.168
280	449 ± 1	443 ± 1	-1.35	1.305 / 1.223

Temperatur / °C	Decay time $\tau_{\text{heating}} / \text{ns}$	Decay time $\tau_{\text{cooling}} / \text{ns}$	Relative change $\Delta\tau_{\text{rel, hc}} / \%$	Reduced χ^2 (heating / cooling)
330	309 ± 1	312 ± 1	0.99	1.363 / 1.313
380	192 ± 1	196 ± 1	1.68	1.327 / 1.381
400	155 ± 1			1.453 / —

The same trend is found for sample 20260129-1 (SCASN 1:27 Ceramabind[®] 880). For clarity, the corresponding decay curves during heating and cooling are again shown in the Appendix (see **figure A6**), while the extracted decay times are plotted in **figure 29**. As for sample 20250812-16, a pronounced fast initial decay component is visible at the beginning of the decay curve. Even so, the resulting decay times during heating and cooling differ only minimally.

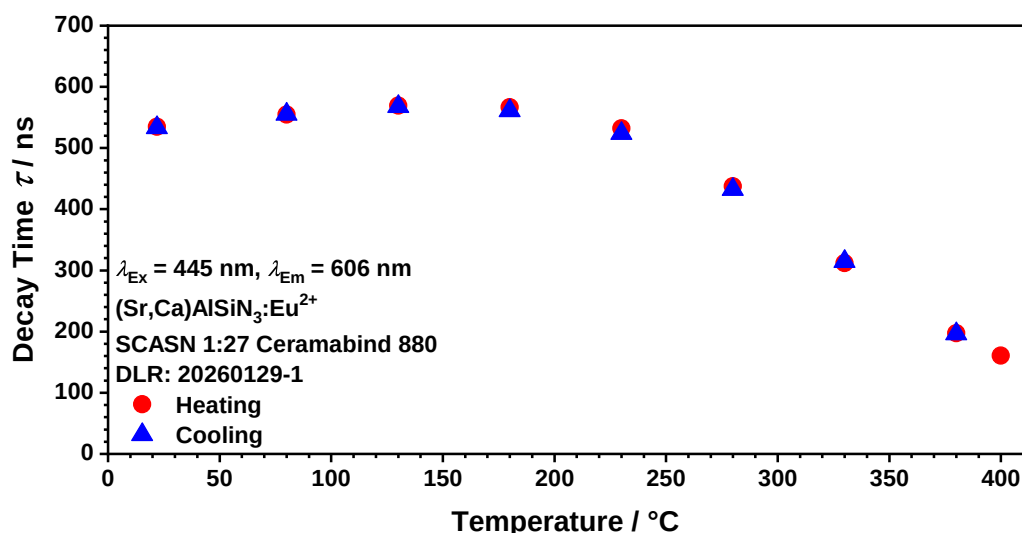


Figure 29: Decay times of sample 20260129-1 (SCASN 1:27 Ceramabind[®] 880) obtained from the decay curves recorded upon heating and cooling in the temperature range from 22 to 400 °C at an excitation wavelength of 445 nm and a monitored emission wavelength of 606 nm. For clarity, error bars for the decay times are omitted, as the corresponding fit standard deviations are smaller than the symbol size.

The corresponding values of $\Delta\tau_{\text{rel, hc}}$ listed in **table 16** lie between –1.50 to 0.84 %. Hence, no relevant hysteresis effect is observed here either.

Table 16: Decay times of sample 20260129-1 (SCASN 1:27 Ceramabind® 880) obtained during heating and cooling, together with the corresponding relative change, are compared. The relative change $\Delta\tau_{\text{rel, hc}}$ is defined according to **equation (9b)**. The corresponding reduced χ^2 values of the fits are listed for heating and cooling.

Temperatur / °C	Decay time $\tau_{\text{heating}} / \text{ns}$	Decay time $\tau_{\text{cooling}} / \text{ns}$	Relative change $\Delta\tau_{\text{rel, hc}} / \%$	Reduced χ^2 (heating / cooling)
22	535 ± 1	533 ± 2	−0.30	1.278 / 1.153
80	555 ± 1	555 ± 2	0.08	1.234 / 1.078
130	569 ± 1	567 ± 2	−0.28	1.236 / 1.266
180	567 ± 1	561 ± 2	−1.09	1.209 / 1.080
230	532 ± 1	524 ± 2	−1.50	1.151 / 1.113
280	438 ± 1	432 ± 2	−1.27	1.228 / 1.204
330	312 ± 1	315 ± 1	0.84	1.341 / 1.296
380	197 ± 1	196 ± 1	−0.47	1.369 / 1.298
400	161 ± 1			1.323 / —

As already suggested on the basis of sample 20250812-15, the occurrence of this pronounced fast initial decay component may be associated with the lower phosphor concentration. It is absent for sample 20260129-2 (SCASN 1:4 Ceramabind® 880), but becomes visible again here for sample 20260129-1 (SCASN 1:27 Ceramabind® 880).

Overall, all heating-cooling comparisons yield very promising results for the lifetime-based readout, even up to 480 °C for samples 20250812-15 and 20250812-16. For nearly all samples, the relative change $\Delta\tau_{\text{rel, hc}}$ remains within the expected measurement uncertainty. Taken together, these results indicate that the lifetime-based approach is much more promising than the intensity-based approach for measurements involving both heating and cooling cycles.

Thermal Degradation

Analogous to the investigations of the SCASN phosphor powder and of the coated SCASN sample by means of intensity-based readout, the thermal degradation behavior was also examined for the lifetime-based method. The same coated sample was used as in the intensity-based study. Accordingly, the sample was heated in air for 2 h at 100 °C, after which the decay curve was recorded at room temperature. The sample was then heated again for 2 h at 200 °C, measured once more, and this procedure was continued stepwise up to 600 °C. After each heating step,

the sample was removed from the drying oven or furnace and allowed to cool naturally to room temperature.

The resulting decay curves are shown in **figure 30(a)**, while the corresponding decay times as a function of temperature are presented in **figure 30(b)**. Over the entire investigated range, the extracted decay times remain very close to each other and scatter only around a mean value of (546.5 ± 1.9) ns. This corresponds to a relative deviation of only 0.35 %. However, after thermal treatment at 500 °C for 2 h, a pronounced fast initial decay component becomes visible at the beginning of the decay curve, similar to the behavior already observed for the lower-loaded samples 20250812-16 and 20260129-1.

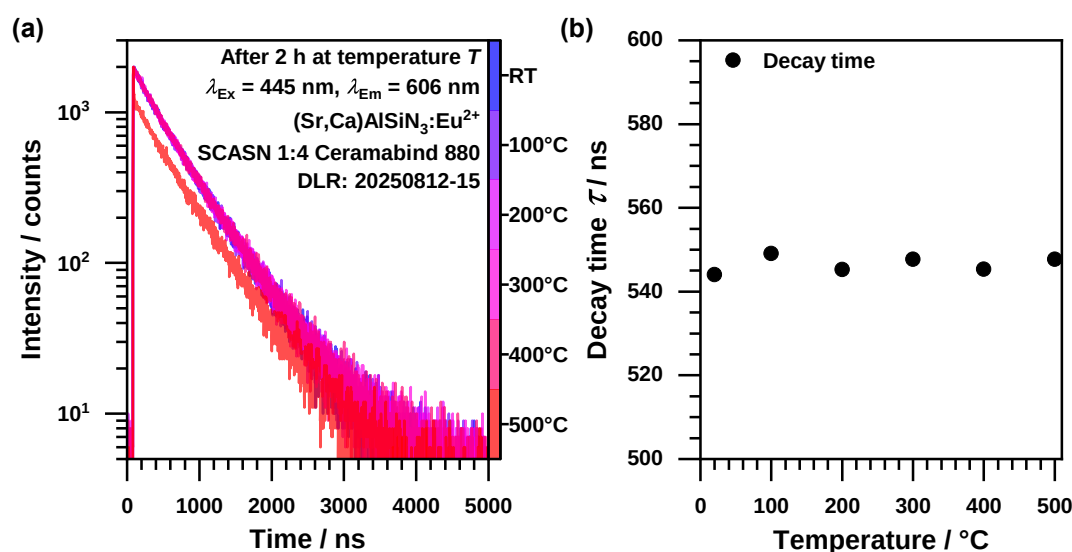


Figure 30: Decay curves measured at room temperature after thermal treatment for 2 h at the indicated temperatures (a) and the corresponding decay times (b) of sample 20250812-15 (SCASN 1:4 Ceramabind® 880) at an excitation wavelength of 445 nm and a monitored emission wavelength of 606 nm. For clarity, error bars for the decay times are omitted, as the corresponding fit standard deviations are smaller than the symbol size.

To visualize this feature more clearly, the corresponding decay curve is shown separately in **figure 31**, together with a magnified view of the initial time range. Since this additional fast initial decay component appears only after heating to the highest investigated temperature, it is reasonable to associate it with thermally induced degradation. A plausible interpretation is that defect-related non-radiative pathways are introduced during degradation and give rise to an additional very fast decay contribution. It is also noteworthy that, in the regular temperature-dependent measurements, this feature was observed only for the coatings with lower phosphor concentration. This is plausible, since at lower phosphor loading the individual particles are more strongly exposed and degradation-related surface effects may therefore become more pronounced.

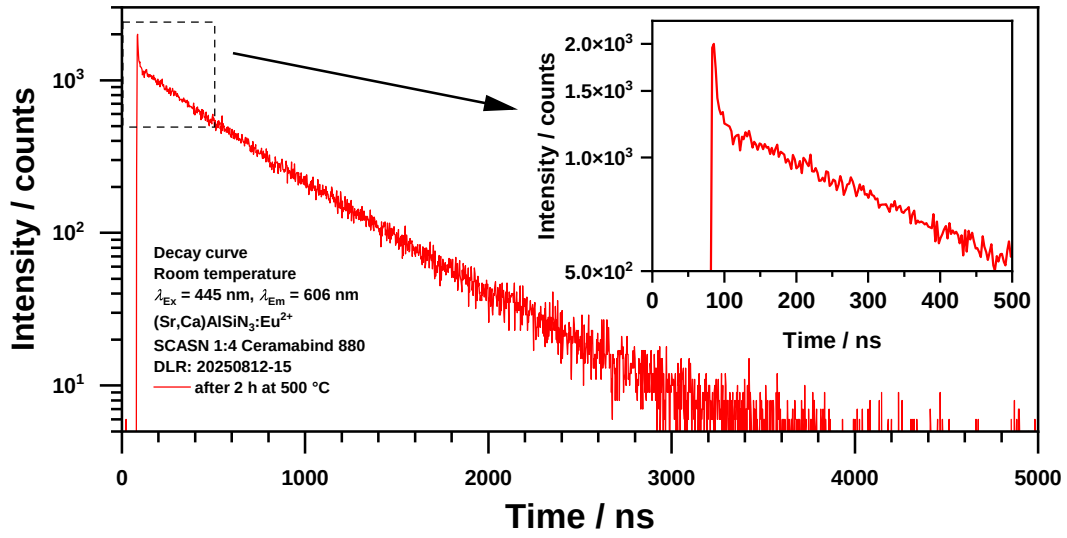


Figure 31: Decay curve of sample 20250812-15 (SCASN 1:4 Ceramabind® 880) measured at room temperature after thermal treatment for 2 h at 500 °C, including a magnified view of the initial part of the decay curve, at an excitation wavelength of 445 nm and a monitored emission wavelength of 606 nm.

To assess the influence of this additional fast component on the lifetime-based evaluation, the corresponding decay curve, which had previously been fitted mono-exponentially like all other decay curves, was also fitted bi-exponentially. This analysis yielded a first partial decay time of $\tau_1 = (10.4 \pm 1.1)$ ns with a fraction of only 0.89 %, whereas the second partial decay time was $\tau_2 = (548 \pm 2)$ ns with a fraction of 99.11 %. The resulting average decay time τ_x is therefore 543.2 ns, which is very close to the mono-exponential result of 547.7 ns. The absolute difference between both approaches is only 4.5 ns, corresponding to a relative deviation of 0.83 % with respect to the mean value of both. Thus, the influence of the additional fast decay component is sufficiently small that a mono-exponential treatment still provides highly reliable results. This finding explains why even for samples showing this degradation-related feature no substantial deviation in the extracted decay times was observed.

It should also be emphasized that a decay contribution with such a short lifetime can only be detected with sufficiently sensitive time-resolved equipment. Since TSP applications are often based on camera systems, it cannot be excluded that such a fast component would not be resolved at all in practice. For the intended application, this again underlines the robustness and practical attractiveness of the lifetime-based approach.

Alternative Substrate

As for the intensity-based method, an alternative substrate in the form of steel was also investigated for lifetime-based readout. For the sake of clarity, the corresponding decay curves of sample 20260129-2 on steel substrate (SCASN 1:4 Ceramabind[®] 880) recorded during heating and cooling are shown in the Appendix (see **figure A7**).

Sample 20260129-2 on steel was prepared analogously to the corresponding coating on aluminum substrate, with the substrate being the only intended difference. After thermal cycling, however, slight microcrack formation (see **figure A13**), the possible reasons for this behavior will again be discussed later in **section 4.2.3**.

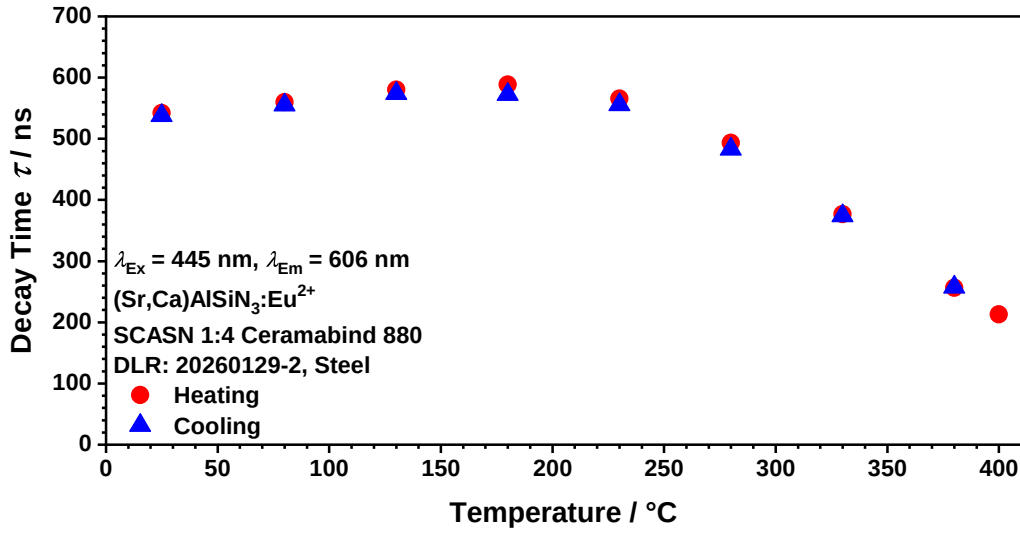


Figure 32: Decay times of sample 20260129-2 (SCASN 1:4 Ceramabind[®] 880, steel substrate) obtained from the decay curves recorded upon heating and cooling in the temperature range from 25 to 400 °C at an excitation wavelength of 445 nm and a monitored emission wavelength of 606 nm. For clarity, error bars for the decay times are omitted, as the corresponding fit standard deviations are smaller than the symbol size.

Figure 32 shows the corresponding decay times. Only minimal deviations between heating and cooling are observed. The resulting values of $\Delta\tau_{\text{rel, hc}}$, listed in **table 17**, lie between -2.73 to 0.38 %. Although this spread is slightly larger than for the corresponding coating on aluminum substrate, it still does not indicate any hysteresis effect comparable to that observed for the intensity-based method.

Table 17: Decay times of sample 20260129-2 (SCASN 1:4 Ceramabind® 880, steel substrate) obtained during heating and cooling, together with the corresponding relative change, are compared. The relative change $\Delta\tau_{\text{rel, hc}}$ is defined according to **equation (9b)**. The corresponding reduced χ^2 values of the fits are listed for heating and cooling.

Temperatur / °C	Decay time $\tau_{\text{heating}} / \text{ns}$	Decay time $\tau_{\text{cooling}} / \text{ns}$	Relative change $\Delta\tau_{\text{rel, hc}} / \%$	Reduced χ^2 (heating / cooling)
25	542 ± 1	538 ± 1	−0.77	1.288 / 1.207
80	560 ± 1	555 ± 1	−0.83	1.128 / 1.192
130	580 ± 1	574 ± 1	−1.11	1.204 / 1.246
180	589 ± 2	573 ± 2	−2.73	1.163 / 1.186
230	566 ± 1	556 ± 1	−1.77	1.166 / 1.249
280	494 ± 1	483 ± 1	−2.22	1.246 / 1.146
330	377 ± 1	374 ± 1	−0.67	1.269 / 1.227
380	256 ± 1	257 ± 1	0.38	1.416 / 1.366
400	213 ± 1			1.444 / —

The same comparison was also performed for sample 20260129-1 on steel substrate (SCASN 1:27 Ceramabind® 880). Again, the coating on steel was prepared analogously to the corresponding coating on aluminum substrate, with the substrate being the only intended difference. After thermal cycling, partial delamination and visible microcrack formation were again observed (see **figure A14**), the possible reasons for this behavior will again be discussed later in **section 4.2.3**. For clarity, the corresponding decay curves during heating and cooling are shown in the Appendix (see **figure A8**). As in the analogous aluminum-based sample with lower phosphor loading, a pronounced fast initial decay component is observed during cooling.

Figure 33 shows the corresponding decay times of sample 20260129-1 on steel substrate during heating and cooling in the temperature range from 25 to 400 °C. Due to a technical error, the decay curve and the corresponding decay time for 130 °C during cooling is not available.

The calculated values of $\Delta\tau_{\text{rel, hc}}$, given in **table 18**, lie between −1.32 to 0.90 %. Accordingly, the deviations remain very small and do not indicate temperature hysteresis.

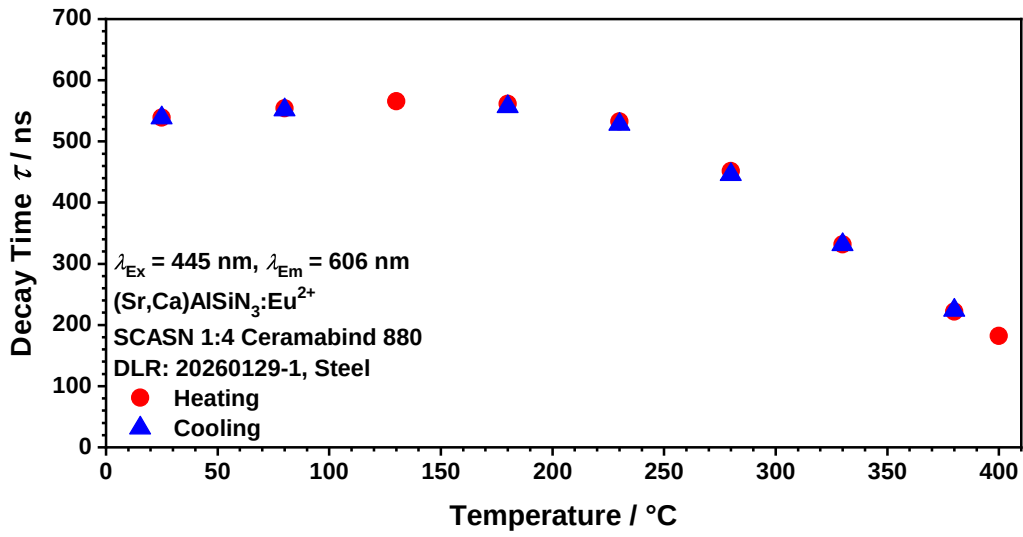


Figure 33: Decay times of sample 20260129-1 (SCASN 1:27 Ceramabind[®] 880, steel substrate) obtained from the decay curves recorded upon heating and cooling in the temperature range from 25 to 400 °C at an excitation wavelength of 445 nm and a monitored emission wavelength of 606 nm. For clarity, error bars for the decay times are omitted, as the corresponding fit standard deviations are smaller than the symbol size.

Table 18: Decay times of sample 20260129-1 (SCASN 1:27 Ceramabind[®] 880, steel substrate) obtained during heating and cooling, together with the corresponding relative change, are compared. The relative change $\Delta\tau_{\text{rel, hc}}$ is defined according to **equation (9b)**. The corresponding reduced χ^2 values of the fits are listed for heating and cooling.

Temperatur / °C	Decay time $\tau_{\text{heating}} / \text{ns}$	Decay time $\tau_{\text{cooling}} / \text{ns}$	Relative change $\Delta\tau_{\text{rel, hc}} / \%$	Reduced χ^2 (heating / cooling)
25	539 ± 1	539 ± 2	−0.10	1.196 / 1.208
80	554 ± 1	551 ± 2	−0.47	1.276 / 1.081
130	566 ± 1	—	—	1.171 / —
180	561 ± 1	557 ± 2	−0.82	1.156 / 1.167
230	533 ± 1	528 ± 2	−0.88	1.251 / 1.115
280	452 ± 1	446 ± 1	−1.32	1.228 / 1.207
330	332 ± 1	331 ± 1	−0.22	1.276 / 1.283
380	222 ± 1	224 ± 1	0.90	1.420 / 1.324
400	182 ± 1	—	—	1.392 / —

Overall, these initial tests with steel as an alternative substrate indicate that the temperature-dependent behavior is very similar to that observed for aluminum substrate. At the same time, the most notable result is once more the robustness of the lifetime-based readout. Even for coatings that exhibited partial delamination or microcracking, the extracted decay times remained highly reproducible.

Alternative Monitored Wavelengths for Decay Measurements

Since the measured decay times become very short at elevated temperatures, for example below 70 ns at 480 °C, their experimental determination becomes increasingly challenging. This is especially relevant with respect to a possible camera-based TSP setup, because time- and spatially resolved measurements in such a regime require both a very short-pulse excitation source in the picosecond range and a sufficiently fast detector. The shorter the decay time, the more demanding the measurement becomes. Accordingly, longer decay times would be advantageous from a practical point of view.

One possible route to achieve this can be rationalized on the basis of the Einstein coefficients. Following the work of *Einstein* [89], the coefficients describing absorption, stimulated emission, and spontaneous emission were introduced. In the present context, the Einstein coefficient A_{21} corresponds to the rate of spontaneous emission. Using the usual frequency-based definition of the Einstein coefficients, it is related to the coefficient B_{21} for stimulated emission by [90]:

$$A_{21} = B_{21} \frac{8\pi hf^3}{c^3} = B_{21} \frac{8\pi h}{\lambda^3} \quad (12)$$

where h is the Planck constant, f is the frequency of the transition, c is the speed of light in vacuum, and λ is the corresponding vacuum wavelength. Additionally, the radiative lifetime τ is given by [90]:

$$\tau = \frac{1}{A_{21}} \quad (13)$$

Combining **equation (12)** and **equation (13)** yields

$$\tau = \frac{\lambda^3}{8\pi h B_{21}} \propto \lambda^3 \quad (14)$$

Thus, if the transition strength represented by B_{21} remains comparable, the lifetime increases with increasing emission wavelength proportionally to λ^3 .

This behavior is indeed observed experimentally and is shown for SCASN powder in **figure 34**. The decay time increases systematically with increasing monitored emission wavelength, that is, toward the red side of the emission band. According to the relation given above, increasing the monitored wavelength from 600 to 650 nm should lead to an increase in lifetime of about 27.1 %. Experimentally, an increase of 29.6 % is observed, namely from 562 ns at 600 nm to 728 ns at 650 nm. This agreement is remarkably good.

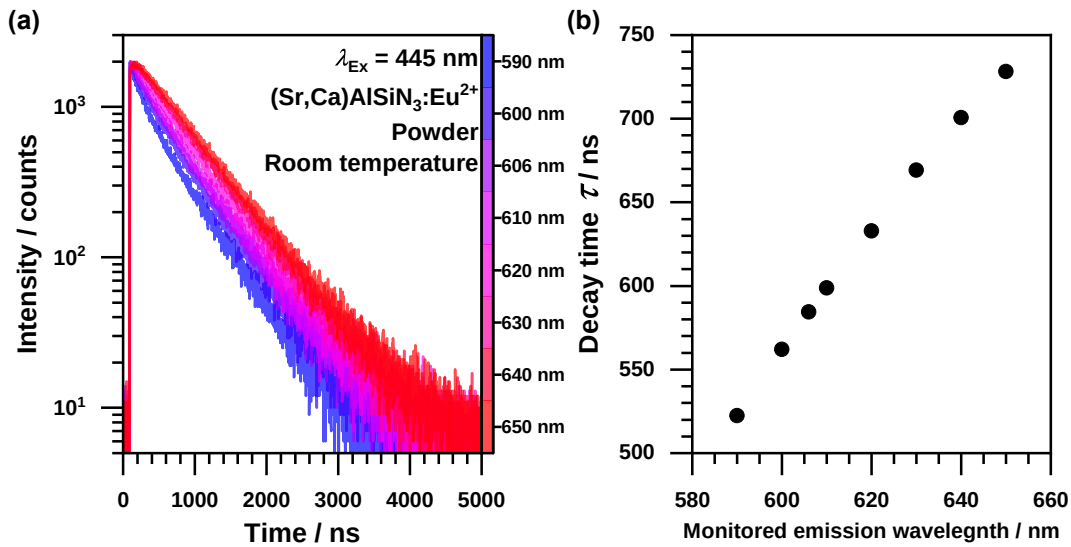


Figure 34: Decay curves recorded at monitored emission wavelengths from 590 to 650 nm (a) and the corresponding decay times (b) of SCASN powder at room temperature and an excitation wavelength of 445 nm. For clarity, error bars for the decay times are omitted, as the corresponding fit standard deviations are smaller than the symbol size.

The same analysis was also carried out for an actual coating, as shown in **figure 35**. Again, a clear increase in decay time with increasing monitored emission wavelength is observed. Here, the agreement with the theoretical estimate is even slightly better, with an experimental increase of 25.8 %, namely from 531 ns at 600 nm to 669 ns at 650 nm. The generally shorter decay times of the coating compared with the powder are plausibly attributed to additional non-radiative relaxation pathways arising from interaction with the ceramic binder.

Overall, this provides a simple and effective route to increase the measured decay times of the coatings by roughly 26 %. In practical terms, this could be realized by employing an optical filter that preferentially selects the longer-wavelength side of the emission band.

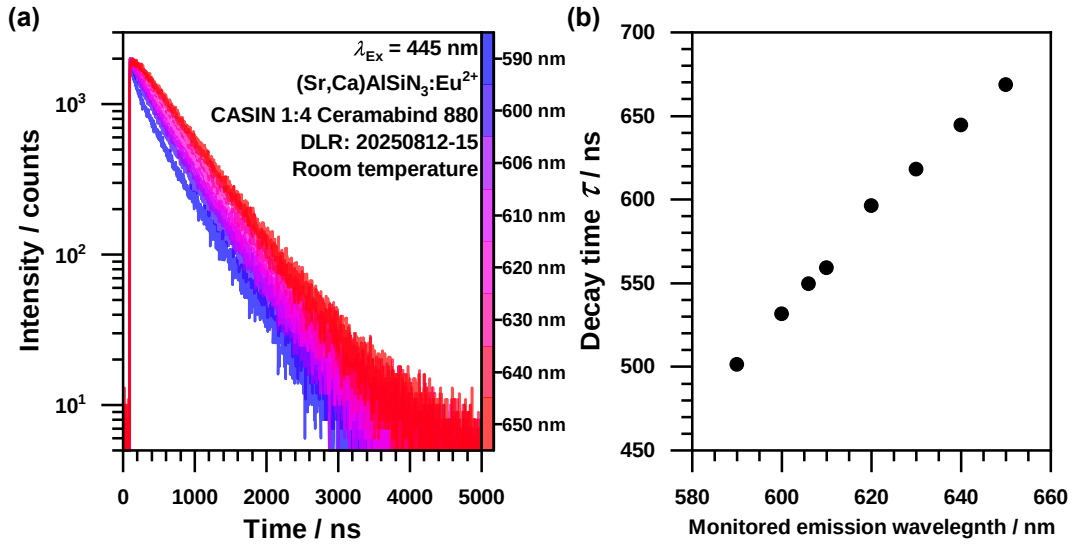


Figure 35: Decay curves recorded at monitored emission wavelengths from 590 to 650 nm (a) and the corresponding decay times (b) of sample 20250812-15 (SCASN 1:4 Ceramabind[®] 880) at room temperature and an excitation wavelength of 445 nm. For clarity, error bars for the decay times are omitted, as the corresponding fit standard deviations are smaller than the symbol size.

4.2.3 Layer Thickness, Roughness and Coating Quality

The approximate coating thicknesses of two selected coatings were measured before and after polishing. For sample 20250812-15 (SCASN 1:4 Ceramabind[®] 880), the layer thickness was approximately 60 μm in the unpolished state and about 30 μm after polishing. For sample 20250812-16 (SCASN 1:27 Ceramabind[®] 880), the corresponding values were approximately 30 μm before polishing and about 20 μm after polishing. Thus, polishing reduced the coating thickness noticeably in both cases, while the resulting layers still remained continuous and mechanically stable for the samples discussed here.

The roughness data of the unpolished coatings are summarized in **table 19**. Before polishing, both samples already exhibited comparatively moderate surface roughness. For sample 20250812-15, mean values of $R_a = 0.504 \mu\text{m}$, $R_z = 2.453 \mu\text{m}$, and $R_{\text{max}} = 3.080 \mu\text{m}$ were obtained. For sample 20250812-16, the corresponding mean values were $R_a = 0.560 \mu\text{m}$, $R_z = 2.267 \mu\text{m}$, and $R_{\text{max}} = 3.787 \mu\text{m}$. Accordingly, the unpolished coatings of both samples were of similar quality, although sample 20250812-16 showed slightly higher peak-to-valley extremes, as reflected by the larger R_{max} value.

Table 19: Roughness parameters of samples 20250812-15 (SCASN 1:4 Ceramabind® 880) and 20250812-16 (SCASN 1:27 Ceramabind® 880) in the unpolished state, including all individual measurements, the mean, and the standard deviation.

DLR 20250812-15 unpolished				DLR 20250812-16 unpolished			
Test	R_a / μm	R_z / μm	R_{max} / μm	Test	R_a / μm	R_z / μm	R_{max} / μm
x_1	0.567	2.750	3.800	x_1	0.643	2.530	3.110
x_2	0.527	2.660	3.490	x_2	0.594	2.310	4.570
x_3	0.437	2.270	2.720	x_3	0.480	2.080	4.170
y_1	0.451	2.070	2.280	y_1	0.596	2.360	3.110
y_2	0.487	2.400	2.960	y_2	0.492	2.140	3.370
y_3	0.552	2.570	3.230	y_3	0.552	2.180	4.390
Mean	0.504	2.453	3.080	Mean	0.560	2.267	3.787
Std. dev.	0.049	0.234	0.499	Std. dev.	0.058	0.152	0.607

Table 20: Roughness parameters of samples 20250812-15 (SCASN 1:4 Ceramabind® 880) and 20250812-16 (SCASN 1:27 Ceramabind® 880) in the polished state, including all individual measurements, the mean, and the standard deviation.

DLR 20250812-15 polished				DLR 20250812-16 polished			
Test	R_a / μm	R_z / μm	R_{max} / μm	Test	R_a / μm	R_z / μm	R_{max} / μm
x_1	0.433	1.860	2.190	x_1	0.064	0.340	0.520
x_2	0.402	1.860	2.080	x_2	0.098	0.650	0.920
x_3	0.370	1.690	1.880	x_3	0.065	0.390	0.720
y_1	0.408	1.860	2.160	y_1	0.047	0.330	0.500
y_2	0.369	1.790	2.080	y_2	0.094	0.540	1.090
y_3	0.410	1.670	1.850	y_3	0.047	0.270	0.630
Mean	0.399	1.788	2.040	Mean	0.069	0.420	0.730
Std. dev.	0.023	0.081	0.130	Std. dev.	0.020	0.132	0.213

The roughness values after polishing are listed in **table 20**. For sample 20250812-15, polishing led to a moderate but clearly measurable smoothing of the surface, with mean values of $R_a = 0.399 \mu\text{m}$, $R_z = 1.788 \mu\text{m}$, and $R_{\text{max}} = 2.040 \mu\text{m}$. For sample 20250812-16, the polishing step had a much stronger effect. Here, the mean roughness values decreased to $R_a = 0.069 \mu\text{m}$,

$R_z = 0.420\ \mu\text{m}$, and $R_{\text{max}} = 0.730\ \mu\text{m}$. Thus, the polished 1:27 coating exhibited a markedly smoother surface than the polished 1:4 coating.

From a practical point of view, these results suggest that application with spray gun initially requires a coating thickness of at least about $30\ \mu\text{m}$ in order to obtain a closed, continuous, and mechanically stable coating layer. Subsequent polishing can then be used to reduce the surface roughness to the values reported above. If required for aerodynamic applications, a further reduction of the coating thickness into the range of approximately $10\ \mu\text{m}$ may in principle be achieved by controlled polishing. However, this was not systematically investigated in the present work and would therefore require additional study.

A further important observation concerns the coating quality of samples 20260129-2 (SCASN 1:4 Ceramabind[®] 880) and 20260129-1 (SCASN 1:27 Ceramabind[®] 880), both on aluminum and on steel substrate. In contrast to samples 20250812-15 and 20250812-16, which were prepared with the same procedure (see **section 3.1.2**), these later samples showed visible coating damage, including partial delamination and microcrack formation (see **section A.1.2**). Since no layer-thickness measurements could be performed for these damaged samples, due to time limitations, a direct correlation cannot yet be established. Nevertheless, one plausible explanation is that local variations in layer thickness and, in particular, insufficient thickness homogeneity may have promoted stress formation during thermal cycling, thereby facilitating delamination and crack formation. At present, however, this interpretation remains tentative. Further work is therefore required to identify the underlying cause and, ultimately, to establish a standardized preparation route for reproducible, homogeneous, and mechanically stable coatings that do not exhibit partial delamination or microcracking during or after the measurement.

4.3 $K_3\text{LuSi}_2\text{O}_7:\text{Eu}^{2+}$ Phosphor Powder

4.3.1 X-ray Powder Diffraction

The powder X-ray diffraction patterns of the synthesized samples are shown in **figure 36**. All measured patterns are in good agreement with the reported reference pattern [91]. On this basis, it is assumed that all samples were successfully synthesized and crystallize in the hexagonal space group $P6_3/mmc$ (# 194). Nevertheless, at Eu^{2+} concentrations above $x = 0.01$, small additional reflections are observed in the range from 13 to 18° , indicating the presence of minor secondary phases. This suggests that incorporation of Eu^{2+} is limited and that secondary phase formation becomes more likely at higher dopant concentrations.

This interpretation is broadly consistent with the empirical substitution rules commonly associated

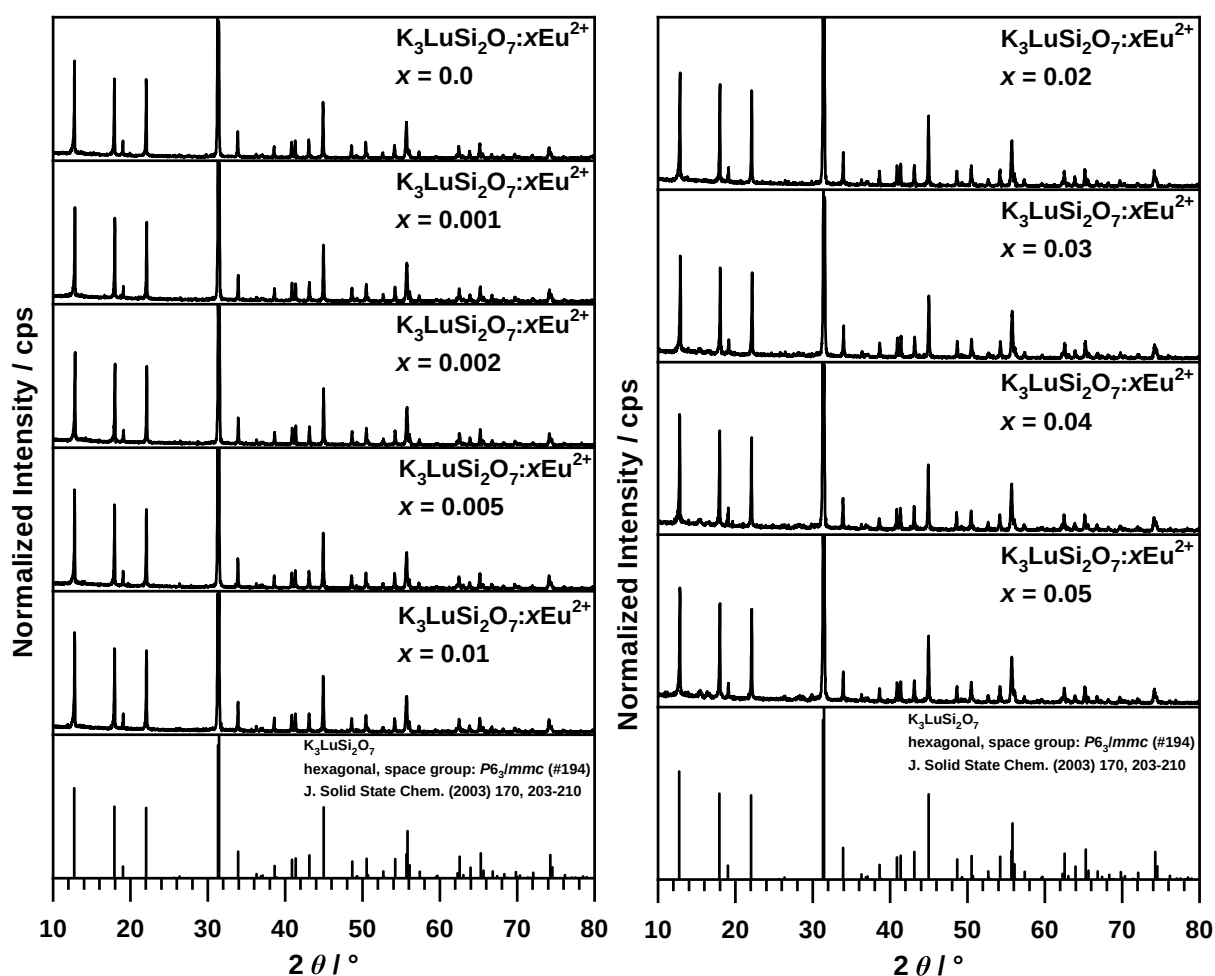


Figure 36: Powder X-ray diffraction pattern of $K_3LuSi_2O_7:xEu^{2+}$ ($x = 0.0-0.05$) powder in comparison with a reference pattern of $K_3LuSi_2O_7$ [91] taken from the Pearson's Crystal Database [92].

with *Goldschmidt*, according to which extensive substitution in ionic solids is generally favored only when the difference in ionic radii is small, often approximated as not exceeding about 15 % [93]. In the present discussion, the relative deviation is calculated with respect to the smaller ionic radius, following the classical formulation of the radius criterion. As suggested by *Vidican et al.*, $K_3LuSi_2O_7$ contains two K^+ sites, both with a coordination number (CN) of 8 [91]. *Qiao et al.*, in contrast, proposed two distinct K^+ sites, one with CN = 6 and one with CN = 9. In either structural description, however, **table 21** shows that the radius mismatch between Eu^{2+} and K^+ is slightly above the commonly cited empirical limit for extensive substitution. Partial substitution therefore appears plausible, whereas at higher concentrations structural instability and the formation of secondary phases, as observed here, appear plausible, particularly in view of the difference in charge.

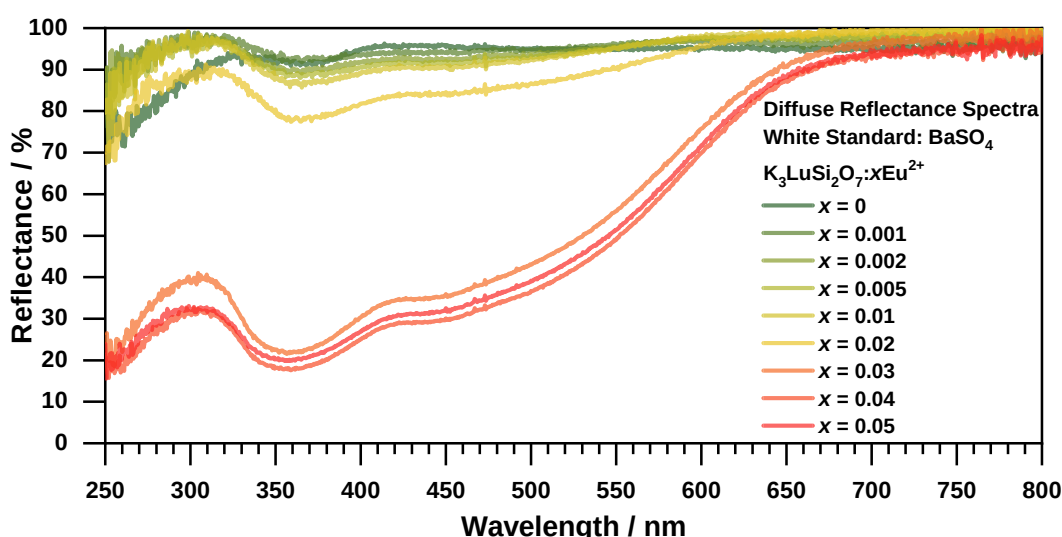
Table 21: Ionic radii of Eu^{2+} and K^+ for coordination numbers (CN) of 6, 8, and 9, and their relative deviation [94].

CN	Eu^{2+} / pm	K^+ / pm	Relative deviation / %
6	131	152	16.0
8	139	165	18.7
9	144	169	17.4

At this stage, it should also be noted that the Lu^{3+} site is octahedrally coordinated (CN = 6) and has an ionic radius of only 100.1 pm, making direct substitution by Eu^{2+} highly unlikely. In contrast, substitution by Eu^{3+} is considerably more plausible, since its ionic radius for CN = 6 is 108.7 pm and its charge matches that of Lu^{3+} .

4.3.2 Diffuse Reflectance Spectra

The diffuse reflectance spectra of all samples recorded in the wavelength range from 250 to 800 nm are shown in **figure 37**. The undoped $K_3LuSi_2O_7$ sample exhibits high reflectance, reaching roughly 95 % between 330 to 800 nm, which suggests good optical quality of the phosphor powder. Below 300 nm, the reflectance starts to decrease, which most likely marks the onset of band gap absorption, in agreement with the work of *Qiao et al.* [15].

**Figure 37:** Diffuse reflectance spectra of $K_3LuSi_2O_7:xEu^{2+}$ ($x = 0-0.05$) in comparison with a $BaSO_4$ white standard.

Upon introduction of Eu^{2+} into the host lattice, a very broad absorption band emerges, extending

approximately from 300 to 700 nm. This band becomes increasingly pronounced and broader with increasing Eu^{2+} concentration. It can be attributed to the expected Eu^{2+} absorption associated with the interconfigurational $4f^7 \rightarrow 4f^6 5d^1$ transitions discussed earlier. The increase in intensity and spectral width up to $x = 0.04$, followed by an apparent saturation, is consistent with the increasing number of Eu^{2+} ions available to participate in these transitions. It is furthermore noteworthy that above 700 nm the reflectance still reaches roughly 95 %, which likewise suggests good optical quality of the doped samples.

4.3.3 Excitation Spectra

The excitation spectra of all samples recorded at room temperature at an emission wavelength of 745 nm in the wavelength range from 250 to 620 nm are shown in **figure 38**. The emission wavelength of 745 nm was selected because it corresponds to the emission maximum, as will be shown in the following section. Except for the undoped sample, all compositions exhibit a very broad excitation band across nearly the entire recorded spectral range. A broad maximum is observed approximately between 420 to 520 nm, which makes this material well suited for excitation in the same spectral region as the SCASN system. This behavior can be attributed to the expected Eu^{2+} absorption associated with the interconfigurational $4f^7 \rightarrow 4f^6 5d^1$ transition discussed earlier. Moreover, the excitation band shows a pronounced dependence on europium concentration. The highest excitation intensity is observed for the sample with $x = 0.03$. At higher europium contents, however, the excitation intensity decreases significantly, indicating reduced excitation efficiency at higher europium concentrations, consistent with concentration quenching.

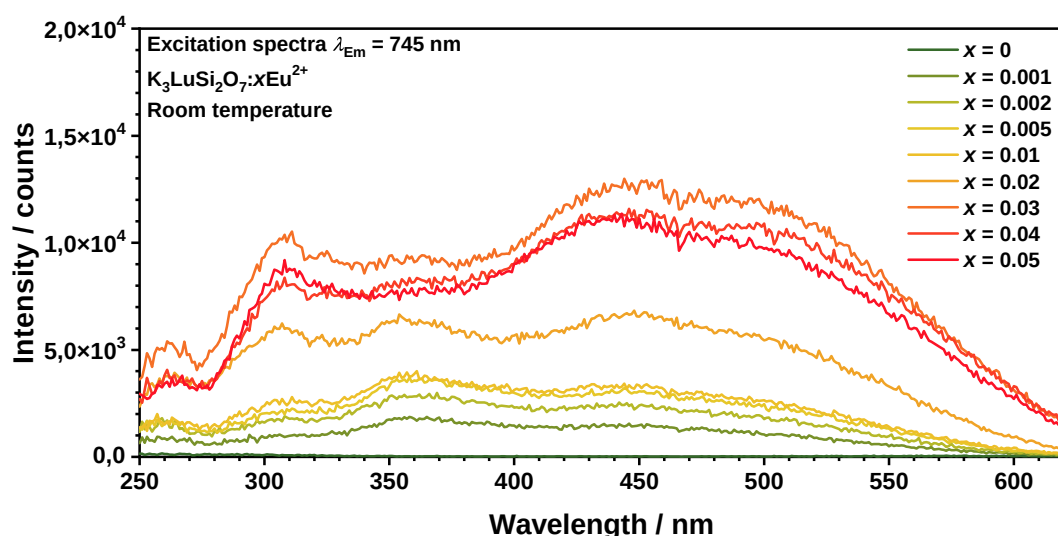


Figure 38: Excitation spectra of $K_3LuSi_2O_7:xEu^{2+}$ ($x = 0-0.05$) recorded at room temperature at an emission wavelength of 745 nm.

4.3.4 Emission Spectra

The emission spectra of all samples recorded at room temperature at an excitation wavelength of 460 nm in the wavelength range from 550 to 850 nm are shown in **figure 39**. The excitation wavelength of 460 nm was chosen to allow direct comparison with the investigated SCASN system. As expected, a very broad emission band, with a local maximum around 745 nm, is observed across the entire recorded spectral range, which is attributed to the $4f^65d^1 \rightarrow 4f^7$ transition discussed earlier. It should be noted that the two lines at 817 nm and 822 nm are artifacts. This assignment is supported by the fact that the undoped sample exhibits these lines as well. They are therefore most likely associated with the xenon arc lamp used for excitation, since xenon shows spectral lines at these wavelengths [95]. Similar to the excitation spectra, the emission intensity shows a pronounced dependence on europium concentration. The highest emission intensity is observed for the sample with $x = 0.03$. At higher europium contents, the emission decreases again, which is consistent with the onset of concentration quenching.

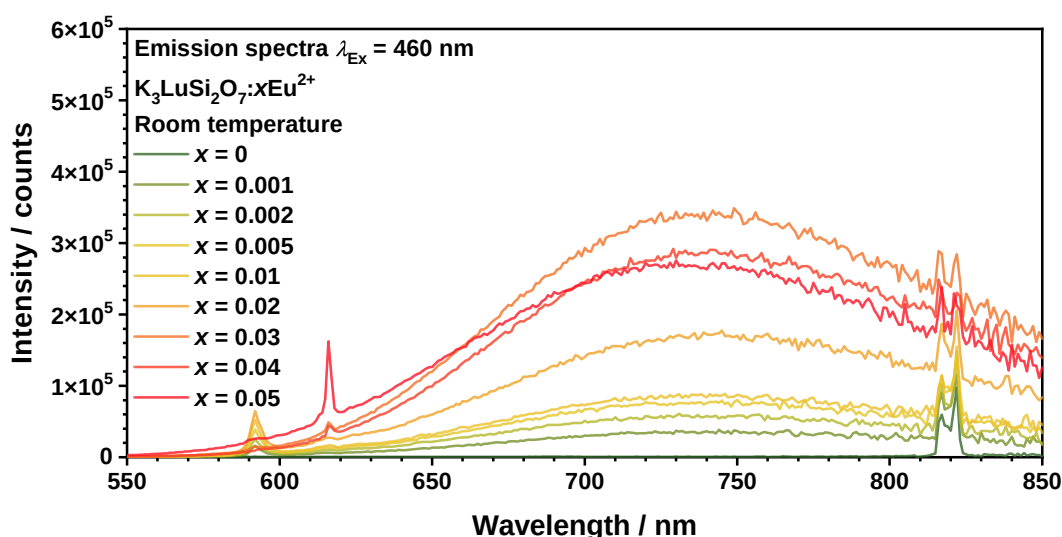


Figure 39: Emission spectra of $K_3LuSi_2O_7:xEu^{2+}$ ($x = 0-0.05$) recorded at room temperature at an excitation wavelength of 460 nm.

In addition to the expected broad Eu^{2+} emission, two narrow Eu^{3+} emission lines arising from intraconfigurational $4f^6 \rightarrow 4f^6$ transitions are observed at 591 nm and 615 nm. These lines are assigned to the $^5D_0 \rightarrow ^7F_1$ and $^5D_0 \rightarrow ^7F_2$ transitions of Eu^{3+} , respectively. As already discussed in **section 4.3.1**, substitution of Lu^{3+} by Eu^{3+} appears plausible, and the presence of these lines is consistent with that interpretation.

It is furthermore noteworthy that the $^5D_0 \rightarrow ^7F_1$ line increases in intensity with increasing europium concentration up to $x = 0.02$. For higher europium contents, this feature is no longer clearly distinguishable, while the $^5D_0 \rightarrow ^7F_2$ line becomes progressively more prominent and is especially pronounced at $x = 0.05$. These Eu^{3+} -related features will be discussed in more detail

later in **section 4.3.8**.

4.3.5 Temperature-Dependent Emission Spectra

For an initial assessment of the temperature-dependent luminescence behavior, emission spectra were recorded in the temperature range from 77 to 500 K at an excitation wavelength of 460 nm, as shown in **figure 40**. A sample with the composition $K_3LuSi_2O_7:0.03 Eu^{2+}$ was selected because this europium concentration showed the highest emission intensity.

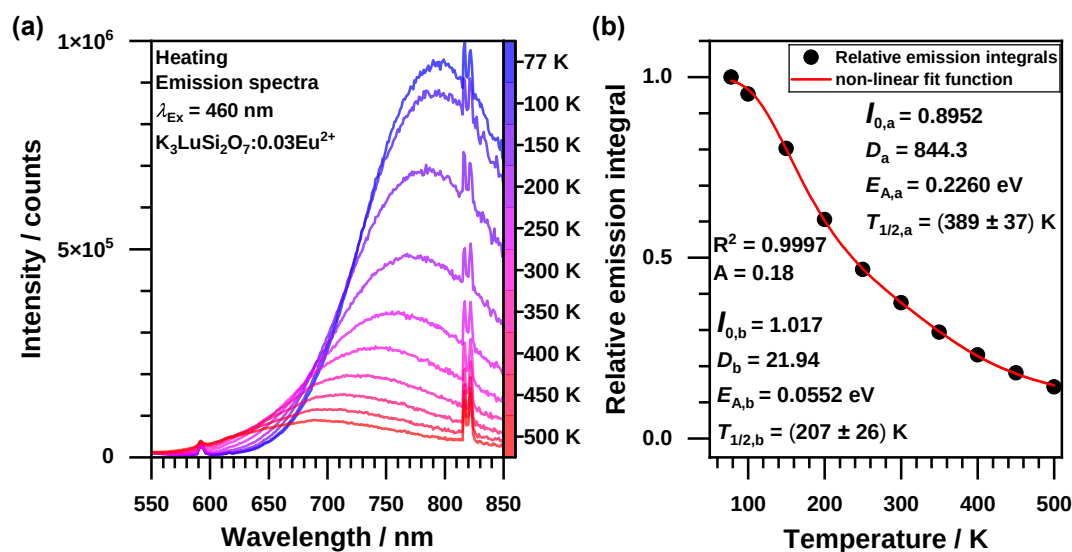


Figure 40: Emission spectra recorded upon heating (a) and the corresponding relative emission integrals together with the non-linear fit function (b) of $K_3LuSi_2O_7:0.03 Eu^{2+}$ in the temperature range from 77 to 500 K at an excitation wavelength of 460 nm.

A pronounced decrease in emission intensity due to thermal quenching is observed over the entire temperature range. In addition, the spectra exhibit the blue shift that was already discussed earlier for the SCASN system. Unlike SCASN, for which the relative emission integrals remained nearly constant up to about 130 °C, KLPE already shows a decrease between 77 to 100 K. Upon further heating, the relative emission integral drops rapidly and reaches only about 13 % of its initial value at 500 K. Moreover, the temperature dependence of the relative emission integrals does not follow the classical sigmoidal behavior observed for SCASN. For that reason, the data were fitted using a two-weighted model (**equation (5a)**).

A plausible explanation for this behavior is the presence of two distinct quenching processes associated with Eu^{2+} occupying different K^+ sites within the host lattice, as discussed in **section 4.3.1**. Because these crystallographic sites differ in their local chemical environment, slightly different energetic gaps from the conduction band may result. This, in turn, could lead to different activation energies for thermal quenching. A related interpretation has also been proposed for

KLPE by Qiao *et al.*, although those authors attributed the behavior to Eu^{2+} located on both K^+ and Lu^{3+} sites [15]. In view of the large difference in ionic radius, however, occupation of the Lu^{3+} site by Eu^{2+} appears questionable.

The two-weighted fit yields two different $T_{1/2}$ values, namely $T_{1/2,a} = (389 \pm 37) \text{ K}$ and $T_{1/2,b} = (207 \pm 26) \text{ K}$. The comparatively large uncertainties most likely arise because only a limited number of data points contribute to each partial quenching process. The relative temperature sensitivity $S_{r,I}$ was calculated to be $0.36 \% \text{ K}^{-1}$ in the temperature range from 77 to 500 K. Thus, the sensitivity of KLPE in powder form is essentially the same as that of the coated SCASN samples 20250812-15 ($0.39 \% \text{ K}^{-1}$) and 20250812-16 ($0.36 \% \text{ K}^{-1}$) in the temperature range from room temperature to 480°C .

Overall, these results suggest that KLPE is a promising candidate for phosphor-based temperature-sensitive paints using intensity-based readout. At the same time, its applicable temperature range is expected to be lower than that of SCASN because of the much steeper decrease in emission intensity. Accordingly, KLPE appears more suitable for measurements within the investigated range from 77 to 500 K with a relative temperature sensitivity of $0.36 \% \text{ K}^{-1}$.

4.3.6 Temperature-Dependent Decay Curves

As in the case of the temperature-dependent emission measurements, the temperature-dependent decay curves (see **figure 41(a)**) and the corresponding average decay times (see **figure 41(b)**) were investigated in the temperature range from 78 to 500 K. The measurements were carried out at an excitation wavelength of 445 nm and an emission wavelength of 745 nm.

The decay curves shown in **figure 41(a)** exhibit bi-exponential behavior. Accordingly, the partial decay times, their relative fractions, and the resulting average decay times are summarized in **table 22**.

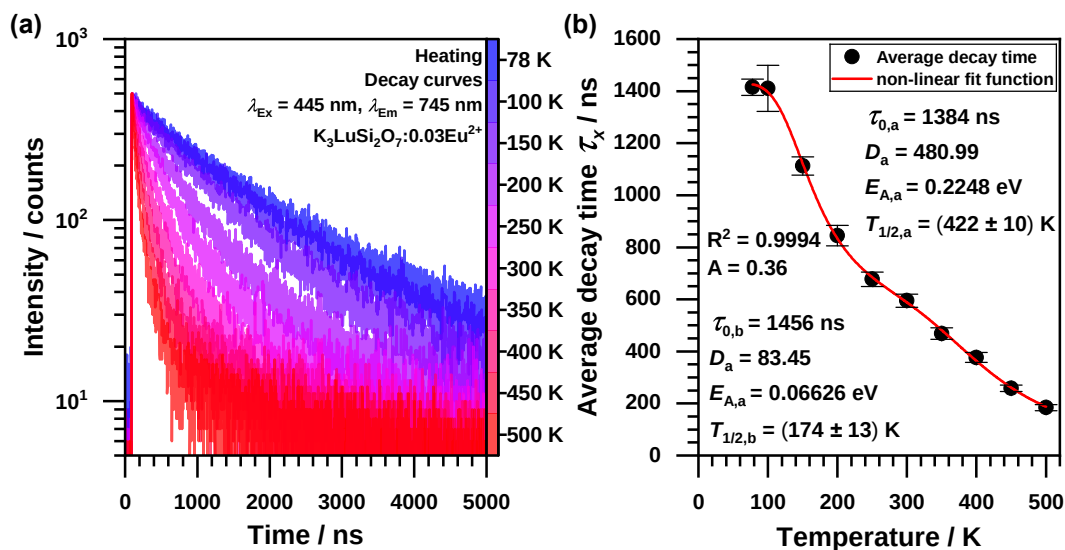


Figure 41: Decay curves recorded upon heating (a) and the corresponding average decay times, including the estimated error, together with the non-linear fit function (b) of $K_3LuSi_2O_7:0.03 Eu^{2+}$ in the temperature range from 78 to 500 K at an excitation wavelength of 445 nm and an emission wavelength of 745 nm.

Table 22: Partial decay times and corresponding average decay times of $K_3LuSi_2O_7:0.03 Eu^{2+}$ in the temperature range from 78 to 500 K. The columns for τ_1 and τ_2 are given together with their fit uncertainties, and the average decay time τ_x corresponds to the amplitude-weighted average decay time obtained from the bi-exponential fit. The uncertainty of τ_x was estimated by Gaussian error propagation from the standard deviations of the partial decay times.

Temperature / K	Decay time τ_1 / ns	Fraction of τ_1 / %	Decay time τ_2 / ns	Fraction of τ_2 / %	Average decay time τ_x / ns
78	306 ± 95	2.1	1439 ± 32	97.9	1415 ± 31
100	536 ± 105	10.5	1513 ± 98	89.5	1411 ± 89
150	385 ± 44	11.6	1208 ± 39	88.4	1113 ± 35
200	407 ± 26	32.9	1059 ± 58	67.1	845 ± 40
250	329 ± 11	48.7	1008 ± 53	51.3	677 ± 27
300	266 ± 6	60.1	1091 ± 62	39.9	595 ± 25
350	185 ± 4	56.9	842 ± 51	43.1	468 ± 22
400	138 ± 4	57.0	694 ± 45	43.0	377 ± 19
450	95 ± 4	50.3	424 ± 26	49.7	258 ± 13
500	73 ± 4	51.6	303 ± 25	48.4	184 ± 12

In a similar manner to the relative emission integrals, the average decay times do not follow a classical sigmoidal trend. This indicates that at least two quenching contributions are likely involved, which is why the data were fitted using a two-weighted model (**equation (5b)**). A plausible explanation is, once more, the presence of Eu^{2+} at different K^+ sites, which may exhibit different quenching behavior because of their distinct local environments. The average decay time of 1415 ns at 78 K is comparatively long for Eu^{2+} , since many Eu^{2+} -activated phosphors show decay times in the range from 100 to 1000 ns [61]. Furthermore, thermal quenching sets in rather early, becoming clearly apparent above 100 K. With increasing temperature, the average decay time decreases rapidly and reaches only 184 ns at 500 K. The two-weighted fit again yields two distinct $T_{1/2}$ values, namely $T_{1/2,a} = (422 \pm 10)$ K and $T_{1/2,b} = (174 \pm 13)$ K. The relative temperature sensitivity $S_{r,\tau}$ was again calculated to be $0.36 \% K^{-1}$ in the temperature range from 78 to 500 K. Thus, the sensitivity of KLPE in powder form for a lifetime-based method is essentially the same as that of the coated SCASN samples 20250812-15 ($0.34 \% K^{-1}$) and 20250812-16 ($0.34 \% K^{-1}$) in the temperature range from room temperature to 480 °C.

Overall, the same general conclusions as for the temperature-dependent emission spectra can be drawn. These results suggest that KLPE is also a promising candidate for phosphor-based temperature-sensitive paints using lifetime-based readout. At the same time, the steep decrease in decay time indicates that the practically useful temperature range is lower than that of SCASN. Accordingly, KLPE appears more suitable for measurements within the investigated range from 77 to 500 K with a relative temperature sensitivity of $0.36 \% K^{-1}$. Compared with conventional TSPs based on organic complexes, which typically exhibit temperature sensitivities of up to about $1 \% K^{-1}$ [1, 17] and, in some cases, can even reach $4 \% K^{-1}$ [17], this value is lower. However, an important distinction is that conventional TSPs usually operate only over a comparatively narrow temperature range and therefore often show higher sensitivities within that limited interval. By contrast, phosphor-based thermometry commonly covers much wider temperature ranges, although typically with lower sensitivities, often on the order of $0.5 \% K^{-1}$ when considered over broad intervals [21]. Against this background, a temperature sensitivity of $0.36 \% K^{-1}$ is comparatively modest, yet still sufficiently reliable for measurements over an extended temperature range.

4.3.7 Thermal Degradation

As in the case of the SCASN phosphor powder, the thermal stability of the KLPE phosphor powder was investigated. For this purpose, the powder was heated in air for 2 h at 100 °C, subsequently measured at room temperature, then heated again for 2 h at 200 °C, measured once more, and so on up to 600 °C. After each heating step, the phosphor powder was removed from the drying oven or furnace and allowed to cool naturally to room temperature. In contrast to the SCASN phosphor powder, both the emission spectra and the decay times were measured at

room temperature after each heating step.

Figure 42 shows the emission spectra together with the corresponding relative emission integrals. As becomes evident from these data, the relative emission integrals remain nearly constant from the initial room temperature measurement up to a treatment temperature of 300 °C. In this range, the relative standard deviation is only 2.6 %, which lies within the expected measurement uncertainty. Upon heating to 400 °C, however, a distinct increase in the relative emission integral is observed. This trend continues after treatment at 500 °C, where the relative emission integral is approximately 44 % higher than in the reference measurement. Thus, within this temperature range, thermal treatment does not yet lead to degradation, but instead appears to improve the luminescence properties. Only after thermal treatment at 600 °C does the emission intensity decrease again, indicating the onset of thermal degradation under these conditions.

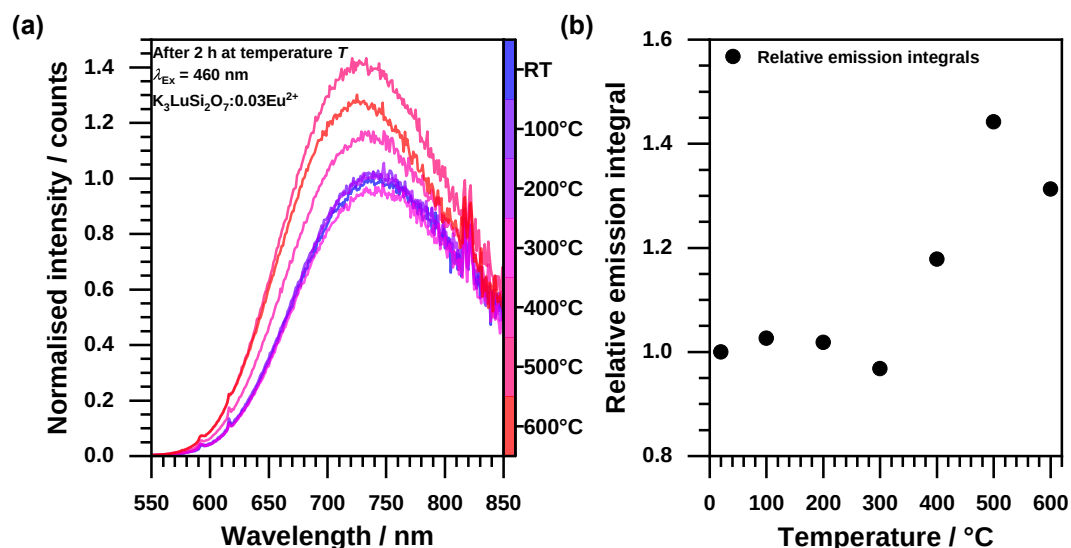


Figure 42: Emission spectra measured at room temperature after thermal treatment for 2 h at the indicated temperatures (a) and the corresponding relative emission integrals (b) of $K_3LuSi_2O_7:0.03Eu^{2+}$ at an excitation wavelength of 460 nm.

The pronounced increase in emission intensity after treatment at 400 and 500 °C is particularly noteworthy. A plausible explanation is the partial elimination of oxygen vacancies within the crystal lattice. As described in **section 3.1.1**, the calcination step during KLPE synthesis is carried out under a flowing reducing atmosphere of 90 % N_2 and 10 % H_2 in order to ensure reduction of Eu^{3+} to Eu^{2+} . Such conditions may also promote the formation of oxygen vacancies. A subsequent annealing step in air could reduce the concentration of these defects and thereby increase the emission intensity, as suggested in the literature [96]. In addition, oxygen vacancies have been reported to act as non-radiative recombination centers, thereby promoting luminescence quenching [97].

This interpretation is further supported by the decay curves and the corresponding average

decay times shown in **figure 43**, which were measured at room temperature at an excitation wavelength of 445 nm and an emission wavelength of 745 nm. Even the first three heating steps lead to a slight, nearly linear increase in the average decay time. After treatment at 500 °C, this increase becomes much more pronounced, while after 600 °C only a comparatively small further increase is observed. From the initial reference measurement to the sample treated for 2 h at 500 °C, the average decay time increases from 602 to 995 ns, corresponding to a relative increase of 65 %.

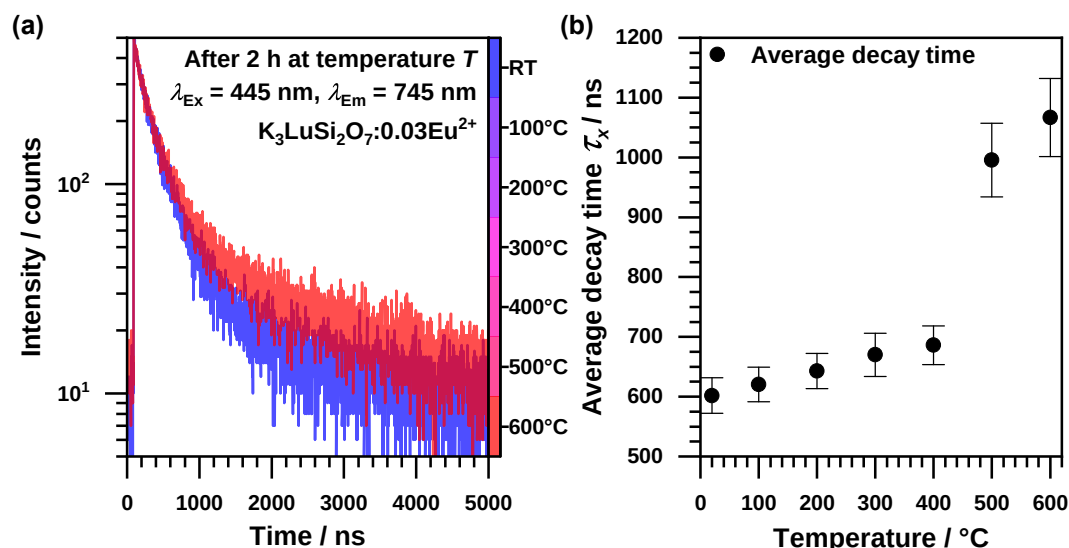


Figure 43: Decay curves measured at room temperature after thermal treatment for 2 h at the indicated temperatures (a) and the corresponding average decay times (b) of $K_3LuSi_2O_7:0.03 Eu^{2+}$ at an excitation wavelength of 445 nm and an emission wavelength of 745 nm. For improved visibility of the changes in the decay curves before and after thermal treatment, the curves obtained after treatment between 100 to 500 °C are omitted. The depicted uncertainty of τ_x was estimated by Gaussian error propagation from the standard deviations of the partial decay times.

The substantially longer average decay time is consistent with the proposed defect-related explanation. If oxygen vacancies indeed act as non-radiative recombination channels [97], their reduction would be expected to suppress non-radiative depopulation pathways. As a result, the excited state would decay more predominantly via radiative processes, leading to longer decay times and higher emission intensity.

Taken together, these results suggest that the obtained KLPE phosphor powder benefits from an annealing step in air at 500 °C for at least 2 h, since both the emission intensity and the average decay time increase markedly under these conditions. Hence, within the investigated range, thermal treatment is beneficial up to 500 °C. Only at 600 °C does the decrease in emission intensity indicate that the onset of thermal degradation has been reached.

4.3.8 X-ray Excited Emission Spectra

Although the measurement of the X-ray excited emission spectra is not of primary relevance to the application as a TSP, these spectra were nevertheless recorded for a more complete characterization of the phosphor and to better assess its general luminescent properties. Interestingly, under X-ray excitation no broad Eu^{2+} emission, as observed for excitation at 460 nm, is detected. Instead, the spectra are dominated by line emission (see **figure 44**). These lines are assigned to the parity-forbidden intraconfigurational transitions of the Eu^{3+} ion, corresponding to the intraconfigurational $[Xe]4f^6 \rightarrow [Xe]4f^6$ transitions. Such transitions are well known and are characteristic of trivalent europium. In slight contrast to the emission spectra recorded under 460 nm excitation, the emission intensity increases up to a concentration of $x = 0.02$ and decreases at higher concentrations, indicating that concentration quenching sets in at a lower concentration than for the Eu^{2+} emission.

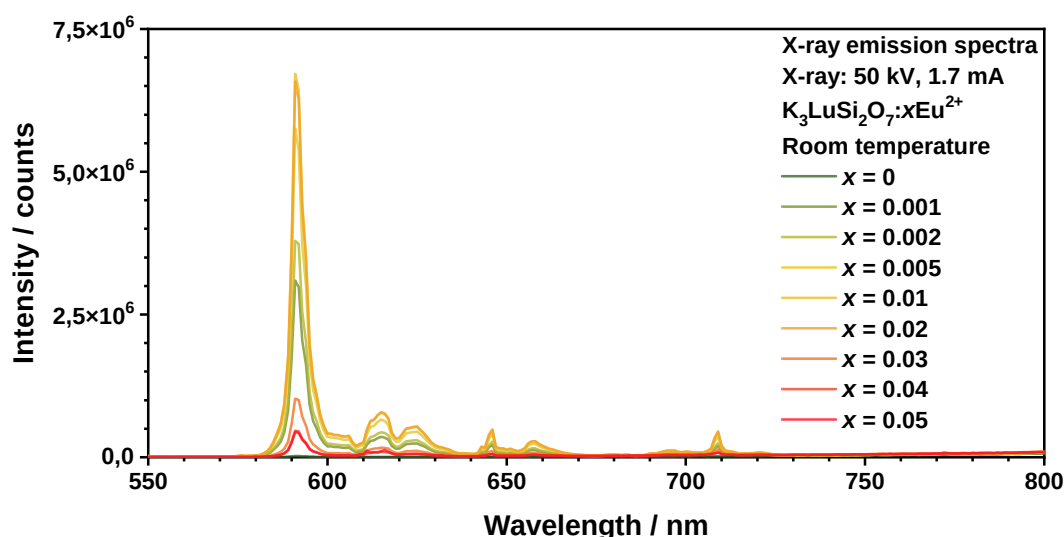


Figure 44: X-ray excited emission spectra of $K_3LuSi_2O_7:xEu^{2+}$ ($x = 0-0.05$) recorded at room temperature under X-ray excitation at 50 kV and 1.7 mA.

The transition assignments follow the work of *Dieke* and *Crosswhite*, as well as *Pier* and are summarized in **table 23** [98–100].

Table 23: Observed emission lines and the corresponding electronic transitions in the X-ray excited emission spectra of $K_3LuSi_2O_7:xEu^{2+}$ ($x = 0-0.05$).

Number	Wavelength / nm	Transition
1	591	$^5D_0 \rightarrow ^7F_1$
2	615, 624	$^5D_0 \rightarrow ^7F_2$

Number	Wavelength / nm	Transition
3	646, 657	$^5D_0 \rightarrow ^7F_3$
4	709	$^5D_0 \rightarrow ^7F_4$

A possible explanation is that high-energy excitation, such as X-ray irradiation, initially generates charge carriers in the host lattice in the form of electron-hole pairs. As shown by *Dorenbos* for a different phosphor system, the resulting emission is governed by trapping, detrapping, and recombination processes, which can lead to luminescence behavior that differs substantially from that observed under visible-light excitation [101]. In this context, Eu^{3+} -related centers may act as relatively efficient electron traps, potentially more readily than Eu^{2+} , due to the higher positive charge of Eu^{3+} . Subsequent recombination with thermalized holes could then populate excited Eu^{3+} states, thereby resulting in the observed intraconfigurational $[Xe]4f^6 \rightarrow [Xe]4f^6$ transitions.

Another notable observation is that the $^5D_0 \rightarrow ^7F_1$ transition is by far the most intense one. This suggests that the Eu^{3+} site likely possesses relatively high symmetry, since the $^5D_0 \rightarrow ^7F_1$ transition has magnetic-dipole character, whereas the other $^5D_0 \rightarrow ^7F_J$ transitions are electric-dipole transitions and are therefore much more sensitive to the *Laporte* rule in a centrosymmetric environment [100]. Again, the *Laporte* rule states that electric-dipole transitions must involve a change in parity, meaning in this case that $4f \rightarrow 4f$ transitions of this kind are strongly forbidden [32]. This explains why the magnetic-dipole transition remains intense, whereas the electric-dipole transitions are less pronounced. It is furthermore observed that, among the electric-dipole transitions, the $^5D_0 \rightarrow ^7F_3$ transition is comparatively weaker than the $^5D_0 \rightarrow ^7F_2$ and $^5D_0 \rightarrow ^7F_4$ transitions. This could likewise be rationalized by a comparatively symmetric local environment, since under such conditions the so-called *J*-mixing, as described within *Judd-Ofelt* theory, may relax the ΔJ selection rules primarily for $J = 2, 4, 6$ [100].

4.3.9 Simplified Energy-Level Scheme of $K_3LuSi_2O_7:Eu^{2+/3+}$

Based on the measurements performed and the corresponding results, a strongly simplified schematic energy-level diagram of $K_3LuSi_2O_7:Eu^{2+/3+}$ was constructed, as shown in **figure 45**. It should be emphasized that most features of this diagram represent only rough approximations and are not derived from directly measured absolute level positions. The optical band gap E_g of the $K_3LuSi_2O_7$ host was taken from the Supplementary Information of *Qiao et al.* and is reported to be 5.0 eV [15]. This value is in good agreement with the possible onset of band-gap absorption discussed in **section 4.3.2**.

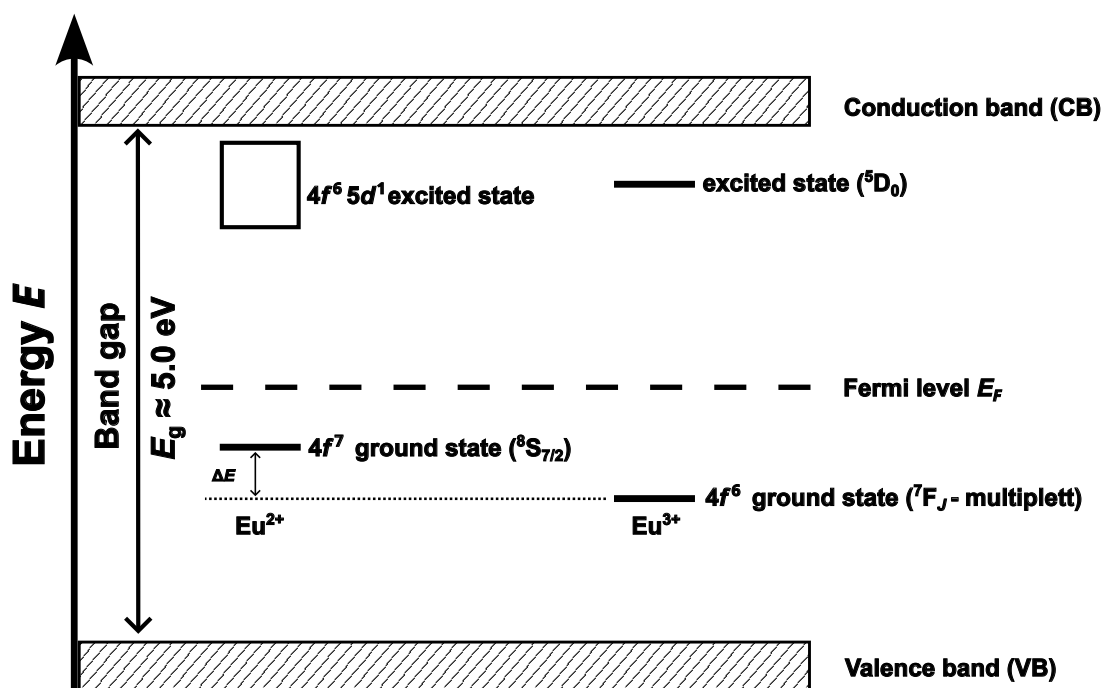


Figure 45: Strongly simplified schematic energy-level diagram of $K_3LuSi_2O_7:Eu^{2+/3+}$. The diagram is intended to illustrate the qualitative relation between the host material and selected Eu^{2+} and Eu^{3+} levels. All level positions are approximate and not to scale.

The energy separation between the $4f^7$ ground state and the $4f^6 5d^1$ excited state of Eu^{2+} was chosen to correspond approximately to the maximum of the emission band at 745 nm, that is, to an energy of about 1.66 eV. The $4f^6 5d^1$ excited state is depicted as a broad band, because it represents a d -derived excited state, as is also commonly done in the literature [15]. Moreover, it is placed close to the conduction band because, as discussed extensively above, thermal ionization is considered the most plausible dominant thermal-quenching mechanism in this Eu^{2+} -activated phosphor. This interpretation is further supported by the comparatively low activation energies obtained from the temperature-dependent emission spectra and decay times of $K_3LuSi_2O_7:Eu^{2+}$.

The Eu^{3+} ground state is shown below the Eu^{2+} ground state. For simplicity, the 7F_J multiplet is represented by a single line rather than by its individual components. It should nevertheless be noted that the true lowest level of this multiplet is 7F_0 , which corresponds to the actual ground state of Eu^{3+} .

The energy difference ΔE between the Eu^{3+} and Eu^{2+} ground states was estimated only very roughly from the standard potential of the Eu^{3+}/Eu^{2+} redox couple in water, reported as -0.35 V [102]. This approach is, of course, only a very crude approximation, since the corresponding energy difference in a solid host such as $K_3LuSi_2O_7$ will differ significantly from that in aqueous solution.

Nevertheless, the simplified energy-level diagram provides a useful qualitative framework for illustrating the relation between the host material and the selected Eu^{2+} and Eu^{3+} energy levels in the investigated phosphor.

4.3.10 Further Photometric and Colorimetric Properties

Again, the evaluation of further photometric and colorimetric properties is not of primary relevance to the application as a TSP. Nevertheless, these properties are considered here in order to provide a more complete characterization of the phosphor and to better assess its general optical properties.

The centroid wavelength values were calculated according to **equation (7)**. In addition, the luminous efficacy (LE), CIE 1931 color coordinates, and centroid wavelength values were derived from the emission spectra as described in **section 3.2.8** to further characterize the optical properties of KLPE under 460 nm and X-ray excitation. Since the LE depends on the overlap of the emission spectrum with the photopic luminosity function of the human eye, pronounced differences between both excitation modes are expected. For photopic vision, the eye sensitivity curve is normalized to 1 at 555 nm and already falls below 1 % of that value at 690 nm [32, 103, 104]. Consequently, emission in the deep-red and near-infrared spectral region contributes only weakly to the LE, whereas spectral components closer to the maximum of the eye sensitivity curve lead to markedly higher values.

For excitation at 460 nm, the emission is dominated by the broad Eu^{2+} band discussed above, with its maximum around 745 nm. Accordingly, the corresponding LE values are comparatively low, as summarized in **table 24**.

Table 24: Luminous efficacy, CIE 1931 color coordinates, and centroid wavelength values derived from the emission spectra at an excitation wavelength of 460 nm of the investigated $K_3LuSi_2O_7:xEu^{2+}$ are summarized.

x in $K_3LuSi_2O_7:xEu^{2+}$	Luminous efficacy $/ \frac{lm}{W_{vis}}$	CIE Color coordinate x	CIE Color coordinate y	Centroid wavelength / nm
0.001	38	0.645	0.355	725
0.002	36	0.653	0.347	724
0.005	37	0.653	0.347	724
0.01	39	0.652	0.348	724

x in $K_3LuSi_2O_7:xEu^{2+}$	Luminous efficacy $/ \frac{lm}{W_{vis}}$	CIE Color coordinate x	CIE Color coordinate y	Centroid wavelength / nm
0.02	31	0.662	0.338	727
0.03	21	0.686	0.314	729
0.04	21	0.686	0.314	729
0.05	37	0.667	0.333	720

As shown in **table 24**, the LE values for 460 nm excitation range from 21 to 39 $\frac{lm}{W_{vis}}$. At the same time, the centroid wavelengths lie between 720 to 729 nm, which confirms that the emission is strongly in the deep-red to NIR spectral region. This is fully consistent with the broad Eu^{2+} emission band observed in **figure 39**. Likewise, the CIE 1931 color coordinates are located in the red region of the chromaticity diagram. In this context, the red standard defined by the National Television System Committee (NTSC) is located at ($x = 0.670$, $y = 0.330$) [105, 106]. Among the investigated samples, the composition with $x = 0.05$ lies closest to this NTSC red point. By contrast, the samples with $x = 0.03$ and $x = 0.04$ are shifted slightly further toward deeper red coordinates, with higher x and lower y values. Overall, the photometric data therefore reflect the strongly red-shifted character of the Eu^{2+} emission under 460 nm excitation.

Under X-ray excitation, in contrast, the emission is dominated by Eu^{3+} line emission, with the most intense contribution located around 591 nm, as discussed in **section 4.3.8**. This spectral position overlaps much more strongly with the photopic eye sensitivity curve. As a result, the corresponding LE values are substantially higher, as summarized in **table 25**.

Table 25: Luminous efficacy, CIE 1931 color coordinates, and centroid wavelength values derived from the X-ray excited emission spectra of the investigated $K_3LuSi_2O_7:xEu^{2+}$ are summarized.

x in $K_3LuSi_2O_7:xEu^{2+}$	Luminous efficacy $/ \frac{lm}{W_{vis}}$	CIE Color coordinate x	CIE Color coordinate y	Centroid wavelength / nm
0.001	345	0.607	0.392	630
0.002	352	0.607	0.392	627
0.005	362	0.607	0.392	623
0.01	365	0.608	0.392	621
0.02	360	0.608	0.391	622

x in $K_3LuSi_2O_7:xEu^{2+}$	Luminous efficacy $/ \frac{lm}{W_{vis}}$	CIE Color coordinate x	CIE Color coordinate y	Centroid wavelength / nm
0.03	240	0.613	0.386	664
0.04	184	0.615	0.384	685
0.05	181	0.617	0.382	686

The data in **table 25** show LE values between 181 and 365 $\frac{lm}{W_{vis}}$. In addition, the centroid wavelengths are markedly shorter than for 460 nm excitation and fall between 621 nm and 686 nm. This shift toward shorter wavelengths indicates significantly better overlap with the human eye sensitivity curve and therefore explains the much higher LE values. The CIE 1931 color coordinates are correspondingly less deeply red than those obtained under 460 nm excitation. For low europium concentrations up to $x = 0.02$, the centroid wavelength remains in the range from 621 to 630 nm, and the LE reaches its highest values. At higher concentrations, however, the centroid wavelength shifts to longer wavelengths and the LE decreases noticeably. This behavior is consistent with the concentration-dependent changes in the X-ray excited emission spectra discussed in **section 4.3.8**.

Direct comparison of both excitation modes highlights the strong influence of the excitation mechanism on the resulting photometric properties. Under 460 nm excitation, the broad Eu^{2+} emission produces very red color coordinates but only low LE values, because most of the emission lies in a spectral region to which the human eye is only weakly sensitive. Under X-ray excitation, by contrast, the dominant Eu^{3+} line emission is located much closer to the maximum of the photopic luminosity function and therefore gives rise to substantially higher LE values. The centroid wavelength values reflect the same trend and confirm that the X-ray excited emission overlaps much more strongly with the eye sensitivity curve than the broad Eu^{2+} emission observed under 460 nm excitation.

4.4 $K_3LuSi_2O_7:Eu^{2+}$ Coating

The KLPE coating samples are highly uneven and inhomogeneous in appearance (see **section A.1.2**) and are therefore not comparable in quality to the coatings obtained for the SCASN system. This is most likely related to the particle size distribution, since the KLPE particles are considerably larger than those of SCASN, as will be discussed in the following section.

Nevertheless, the emission spectra were recorded for samples 20251119-1 (KLPE 1:4 Ce-

ramabind[®] 880), 20260129-3 (KLPE 1:27 Ceramabind[®] 880), and 20260129-4 (KLPE 1:4 Ceramabind[®] 880), and are shown in **figure 46**. Although the overall emission intensity is very low, the expected broad emission band is still clearly visible for all samples. As noted above, the two lines at 817 and 822 nm are artifacts originating from xenon lines of the xenon arc lamp used for excitation. Among the investigated coatings, sample 20260129-4 exhibits the highest emission intensity. Even for this sample, however, the intensity at 745 nm is only roughly one tenth of that of the phosphor powder with the same europium concentration, namely $K_3LuSi_2O_7:0.03 Eu^{2+}$.

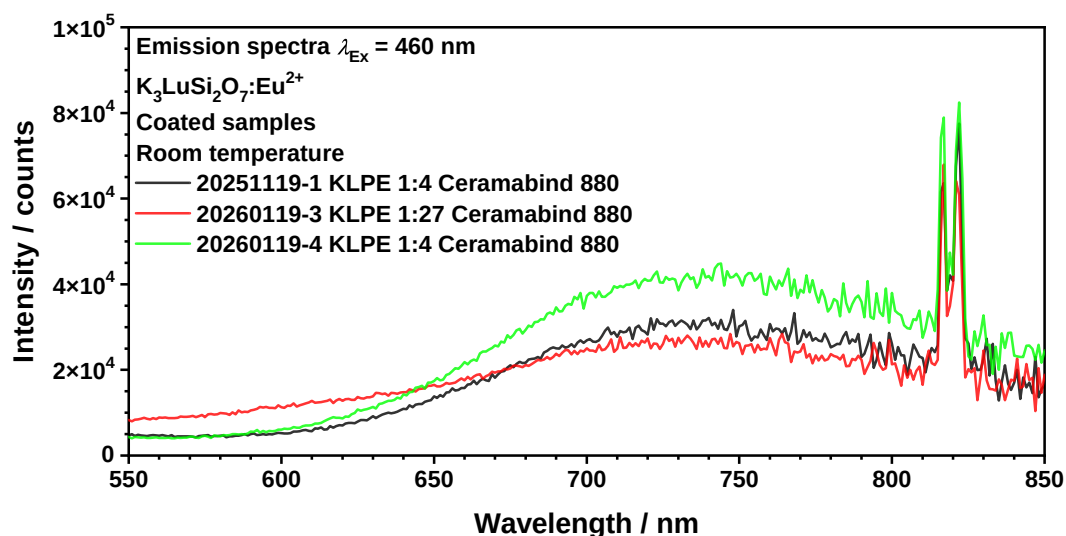


Figure 46: Emission spectra of the coated $K_3LuSi_2O_7:Eu^{2+}$ samples recorded at room temperature at an excitation wavelength of 460 nm. The spectra correspond to samples 20251119-1 (KLPE 1:4 Ceramabind[®] 880), 20260129-3 (KLPE 1:27 Ceramabind[®] 880), and 20260129-4 (KLPE 1:4 Ceramabind[®] 880).

To assess the general viability of the coatings for temperature-dependent emission measurements, sample 20260129-4 (KLPE 1:4 Ceramabind[®] 880) was heated from room temperature to 180 °C. The resulting emission spectra and relative emission integrals are shown in **figure 47**. It should be noted that, for this experiment, only the spectral range between 550 to 800 nm is displayed and only this interval was used for integration. This restriction was necessary because the xenon-lamp artifacts at 817 and 822 nm were comparatively intense relative to the weak emission of the coating samples and would therefore have introduced an undesired contribution.

As already discussed in **section 4.3.5**, thermal quenching in KLPE sets in at comparatively low temperatures, and the emission is already strongly quenched at room temperature. Even so, a clear temperature-dependent decrease in emission intensity is still observed within the investigated range. Because of this pronounced thermal quenching, the relative temperature sensitivity $S_{r,I}$ was calculated to be 0.56 % K⁻¹ in the temperature range from 25 to 180 °C. This

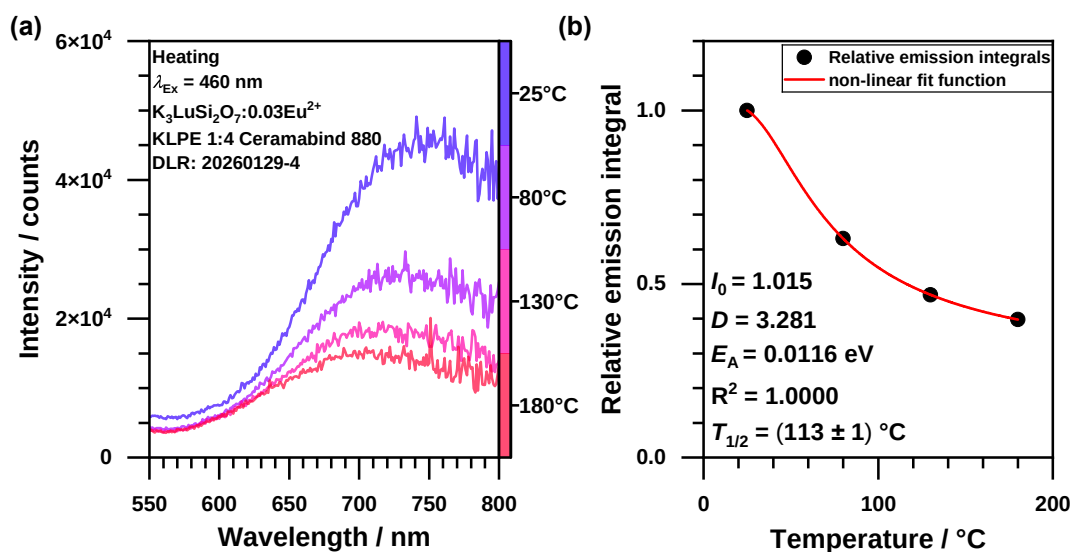


Figure 47: Emission spectra recorded upon heating (a) and the corresponding relative emission integrals together with the non-linear fit function (b) of sample 20260129-4 (KLPE 1:4 Ceramabind® 880) in the temperature range from 25 to 180 °C at an excitation wavelength of 460 nm. The relative emission integrals were obtained by integrating the spectra up to 800 nm.

value is comparatively high relative to the other systems investigated in the present work, although it applies only over a comparatively narrow temperature interval. These results demonstrate the general viability of this coating system for phosphor-based temperature-sensitive paint applications within the investigated range, that is, from room temperature up to at least 180 °C.

It should furthermore be noted that no decay curves could be measured reliably, neither at room temperature nor at elevated temperatures. This is attributed to the very low emission intensity of the coatings in combination with the limited detector sensitivity in this spectral region.

4.4.1 Possible Improvement of Coating

As discussed in the previous section, the quality and homogeneity of the KLPE coating samples are insufficient and clearly inferior to those of the SCASN coatings. This is most likely caused by the considerably larger particle size of KLPE, which hinders the formation of a homogeneous coating surface. As shown in **figure 48(a)**, the d_{50} value of a $K_3\text{LuSi}_2\text{O}_7:0.03\text{Eu}^{2+}$ sample prior to any treatment was 54 μm . By comparison, the SCASN phosphor powder used in this work exhibited a d_{50} value of 13 μm (data provided by Dr. Tim Pier, AG TOM, research group of Prof. Dr. Thomas Jüstel). Thus, the d_{50} value of KLPE was approximately four times higher.

For this reason, a top-down approach based on milling was pursued in order to obtain a smaller

and more monomodal particle size distribution. To do this, 5 g of the sample were milled for 12 h in a 125 mL polyethylene bottle with 30 mL of isopropanol on a roller bench at a speed of 2000 rpm. This approach proved successful, as shown in **figure 48(a)**. After milling, the particle size distribution became much more monomodal, and the d_{50} value decreased to 12 μm , which is very close to that of SCASN.

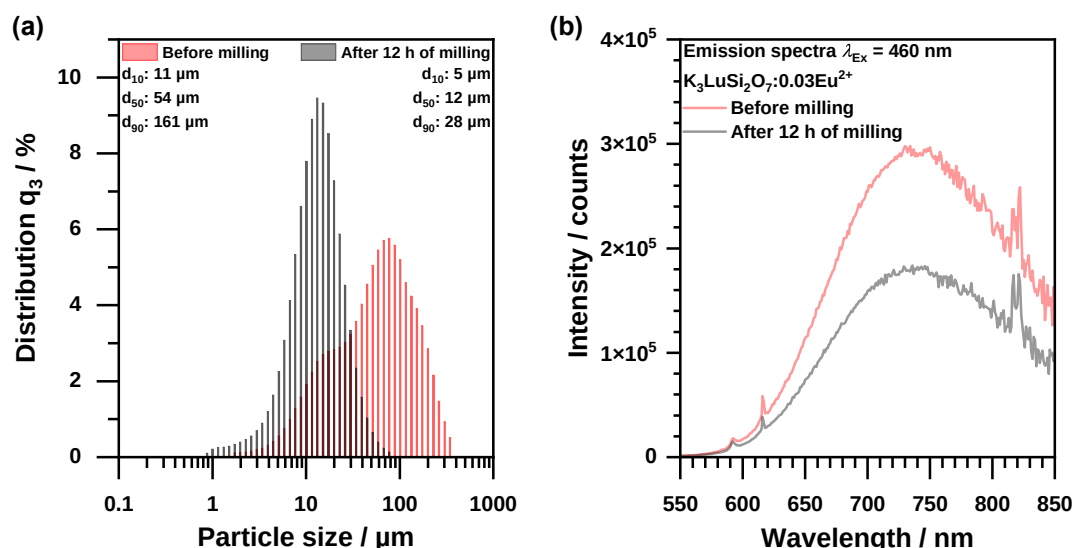


Figure 48: Particle size distributions (a) and the corresponding emission spectra (b) of $K_3\text{LuSi}_2\text{O}_7:0.03\text{Eu}^{2+}$ (DLR-MA-2025-FR-057) before milling and after 12 h of milling. The emission spectra were recorded at an excitation wavelength of 460 nm.

As shown in **figure 48(b)**, the emission intensity of the milled sample decreases by roughly 50 % compared with that of the sample before milling. This is a well-known consequence of surface-defect formation caused by the mechanical stress introduced during milling, which can lead to a reduction in emission intensity [107]. It has also been reported that a post-milling annealing step can partially restore the emission intensity [107, 108], most likely because of improved crystallinity and partial healing of milling-induced defects. Since the untreated KLPE phosphor powder likewise showed increased emission intensity after annealing at 500 $^{\circ}\text{C}$ for 2 h, as discussed in **section 4.3.7**, the milled sample was subjected to the same treatment.

As illustrated in **figure 49(a)**, the particle size distribution did not change noticeably after the additional annealing step. This is a favorable result, since no significant particle agglomeration occurred. At the same time, **figure 49(b)** shows that no recovery of the emission intensity was achieved. Instead, a slight further decrease was observed.

These results indicate that reducing the particle size distribution in order to improve the coating quality is indeed possible, but only at the expense of a substantial loss in emission intensity. Under the conditions tested here, this loss could not be recovered by the subsequent annealing step. Further studies are therefore required to determine whether a milling process involving

lower mechanical stress, for example at reduced rotational speed, could minimize the decrease in emission intensity, as suggested by the literature [108]. Likewise, the temperature and duration of the annealing step should be investigated in more detail in order to evaluate whether partial regeneration can still be achieved. In addition, alternative synthesis routes, such as sol-gel or combustion synthesis, could be explored in order to obtain smaller particle sizes without the need for post-processing [60].

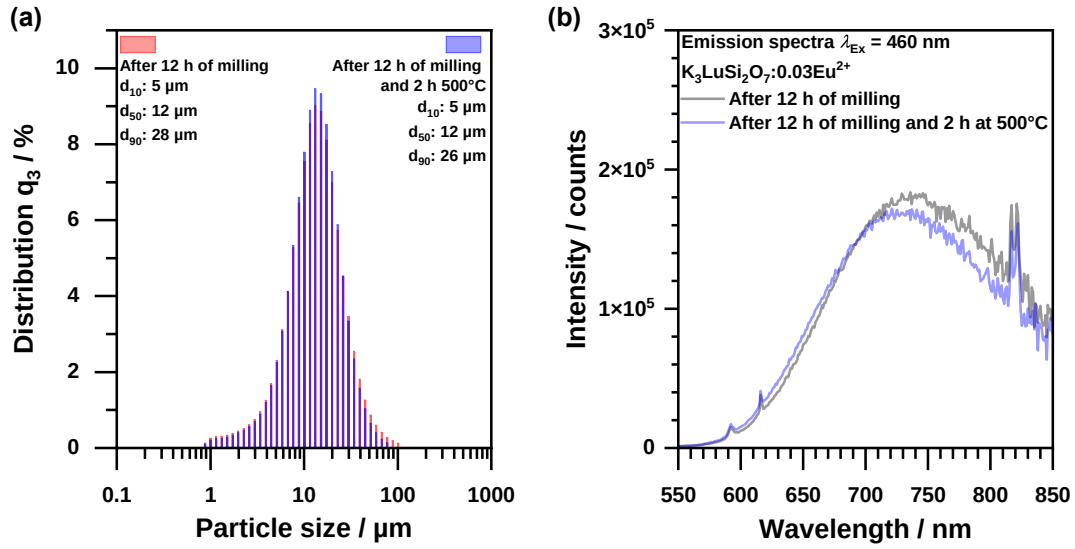


Figure 49: Particle size distributions (a) and the corresponding emission spectra (b) of $K_3LuSi_2O_7:0.03Eu^{2+}$ (DLR-MA-2025-FR-057) after 12 h of milling and after an additional thermal treatment for 2 h at 500 °C. The emission spectra were recorded at an excitation wavelength of 460 nm.

5 Conclusions

(Sr,Ca)AlSiN₃:Eu²⁺: As already shown in previous work, (Sr,Ca)AlSiN₃:Eu²⁺ exhibits strong excitation in the blue spectral region, intense broad-band emission around 620 nm, a large Stokes shift, and temperature-dependent thermal quenching, all of which are favorable for phosphor-based temperature-sensitive paints. In the present work, further characterization was carried out in order to gain a deeper understanding of the temperature-dependent behavior, especially of coated samples.

For intensity-based readout, the coatings showed very similar behavior during heating, with $T_{1/2}$ values of $(328 \pm 4)^\circ\text{C}$ and $(330 \pm 3)^\circ\text{C}$, and relative temperature sensitivities of 0.36 to 0.39 % K⁻¹ up to 480 °C. However, pronounced deviations between heating and cooling were observed for nearly all samples. Thus, intensity-based readout appears suitable mainly for heating-only measurements.

By contrast, the lifetime-based approach proved distinctly more robust. The decay times showed very similar temperature dependences for both phosphor concentrations, with $T_{1/2}$ values of $(339 \pm 3)^\circ\text{C}$ and $(346 \pm 3)^\circ\text{C}$, and relative temperature sensitivities of about 0.33 to 0.34 % K⁻¹. Most importantly, only very small differences between heating and cooling were observed, even up to 480 °C. In addition, lifetime-based readout was less sensitive to phosphor-concentration differences, coating inhomogeneities, and visible coating damage. Up to about 280 °C, even the phosphor concentration had only a minor influence on the decay times.

The results further showed that the sensitivity of the intensity-based approach can be improved by restricting the integration interval. For sample 20250812-15, the relative temperature sensitivity in the range from 28 to 180 °C increased from 0.02 to 0.12 % K⁻¹ when the integration interval was changed from 520 to 800 nm to 600 to 800 nm. Likewise, monitoring the decay at longer wavelengths is advantageous, since the decay time increased by about 26 % between 600 and 650 nm.

Overall, SCASN coatings appear especially promising for lifetime-based phosphor-based temperature-sensitive paints at elevated temperatures, in particular when reliable measurements during both heating and cooling are required. In this respect, the measured data provide a sound experimental basis for the future implementation and evaluation of a camera-based measurement setup.

K₃LuSi₂O₇:Eu²⁺: It was successfully possible to synthesize a phosphor in the desired deep-red to NIR spectral region. K₃LuSi₂O₇:Eu²⁺ can be excited efficiently in the blue spectral range and shows a broad emission band between about 650 and 800 nm, with a local maxi-

mum around 745 nm, resulting in a very large Stokes shift. Among the investigated samples, $\text{K}_3\text{LuSi}_2\text{O}_7:0.03\text{Eu}^{2+}$ showed the highest emission intensity.

Temperature-dependent measurements on the powder demonstrated that KLPE is in principle suitable for both intensity-based and lifetime-based thermometry. In the range from 78 to 500 K, both methods yielded a relative temperature sensitivity of $0.36\% \text{K}^{-1}$. At the same time, thermal quenching sets in much earlier than for SCASN, indicating that KLPE is better suited for lower-temperature applications.

This was also reflected in the coating measurements. For sample 20260129-4, intensity-based readout from room temperature to 180°C yielded a relative temperature sensitivity of $0.56\% \text{K}^{-1}$. However, the coating emission remained weak, and reliable decay measurements were not possible.

In addition, the KLPE coatings were markedly more inhomogeneous than the SCASN coatings. Through milling, the particle size was reduced significantly in order to achieve better and more homogeneous coatings, but this also caused a substantial loss in emission intensity that could not be recovered. This is in contrast to the untreated KLPE powder, for which a subsequent annealing step at 500°C for 2 h increased both the emission intensity and the average decay time markedly, which is plausibly related to a reduction of defect-related non-radiative pathways.

Overall, KLPE is a scientifically interesting and potentially useful phosphor for phosphor-based temperature-sensitive paints in the lower-temperature range. However, compared with SCASN, the low coating intensity and insufficient coating quality remain the main limitations at present.

6 Outlook

(Sr,Ca)AlSiN₃:Eu²⁺: For SCASN, one important next step is the development of a standardized coating process that avoids partial delamination, microcracking, and local thickness inhomogeneities. Equally important, camera-based measurements should be performed in order to verify whether the promising spectrometer-based results can be transferred to a practical spatially resolved TSP setup. The effects of optical filtering, monitored wavelength, coating thickness, and polishing should also be investigated further. In addition, the influence of thermal pretreatment should be studied systematically. In particular, coatings pretreated at 400°C for 2 h should be examined in more detail, since the present results indicate that such a treatment may increase the emission intensity, possibly due to the formation of an optically favorable interfacial layer, which could be beneficial for intensity-based measurements.

$\text{K}_3\text{LuSi}_2\text{O}_7\text{:Eu}^{2+}$: For KLPE, future work should first focus primarily on improving powder preparation, subsequent treatment and the resulting coating quality. Gentler milling conditions should be explored in order to reduce particle size while minimizing emission losses, and the subsequent annealing conditions should be optimized further. Moreover, alternative synthesis routes that intrinsically yield finer powders, like sol-gel or combustion methods, may represent a promising approach. On this basis, more homogeneous coatings should be prepared and evaluated. If the present limitations in coating quality and signal intensity can be overcome, KLPE may become a useful complementary system for lower-temperature phosphor-based TSP applications.

Alternative Phosphor for Temperature-Sensitive Paint

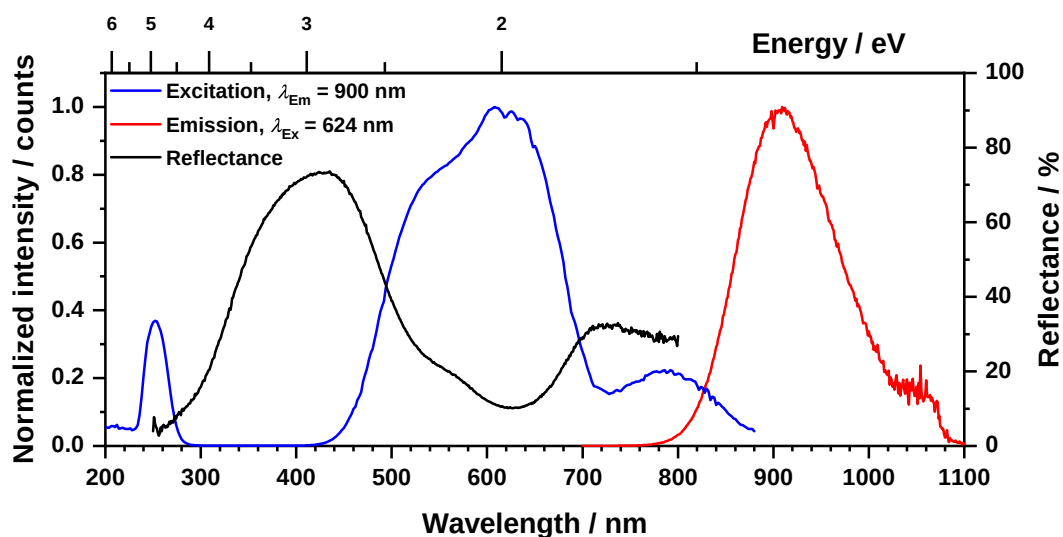


Figure 50: Excitation, emission, and reflectance spectra of $\text{CaCuSi}_4\text{O}_{10}$ powder at room temperature. The excitation spectrum was recorded at an emission wavelength of 900 nm and the emission spectrum at an excitation wavelength of 624 nm. Data provided by Dr. Julia Goldmann and Dr. Patrick Pues, AG TOM, research group of Prof. Dr. Thomas Jüstel.

In addition to further optimizing already established phosphors for temperature-sensitive paints operating at elevated temperatures, the search for alternative phosphors for this purpose is still ongoing. In this context, Egyptian blue, $\text{CaCuSi}_4\text{O}_{10}$, is of interest, especially because it represents a deep-red to near-infrared phosphor.

Egyptian blue is an ancient blue pigment that has been known for several thousand years and was already used in ancient Egypt [109]. Its photoluminescence, however was probably first described by *Pozza et al.* in 2000 [110]. From the perspective of phosphor-based thermometry, Egyptian blue appears attractive because it combines near-infrared emission with a comparatively high quantum yield [111] and excellent chemical stability [112].

As shown in **figure 50**, Egyptian blue exhibits a broad excitation band with high intensity extending roughly from 500 to 700 nm. In addition, a broad emission band is observed approximately between 860 and 980 nm, with a local maximum around 920 nm.

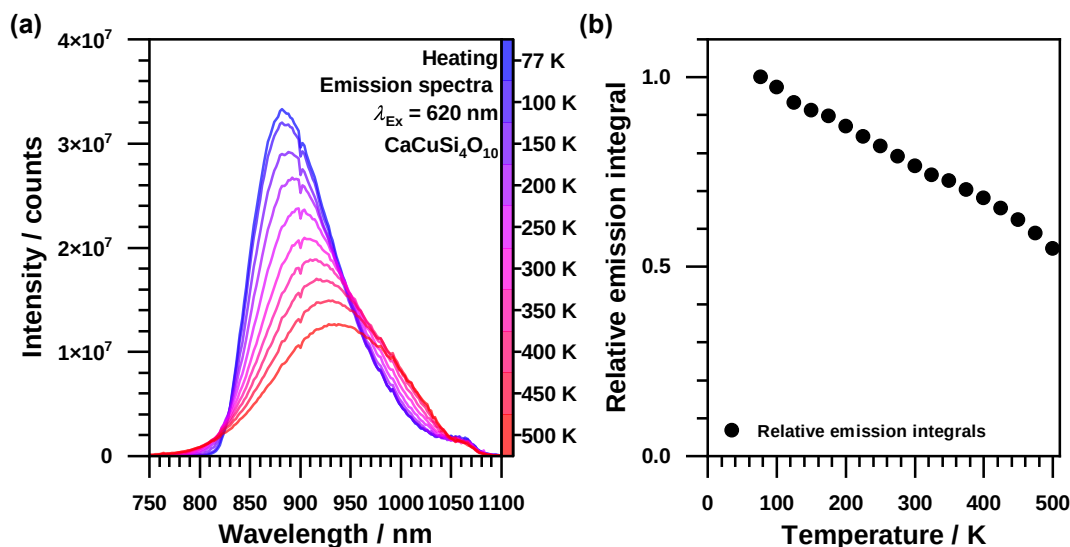


Figure 51: Emission spectra recorded upon heating (a) and the corresponding relative emission integrals (b) of $\text{CaCuSi}_4\text{O}_{10}$ powder in the temperature range from 77 to 500 K at an excitation wavelength of 620 nm. Data provided by Dr. Julia Goldmann and Dr. Patrick Pues, AG TOM, research group of Prof. Dr. Thomas Jüstel.

The temperature-dependent emission behavior shown in **figure 51** is likewise noteworthy. A clear thermal quenching of the emission intensity in the temperature range from 77 to 500 K is observed. At the same time, the relative emission integrals remain above 50 % even at 500 K, indicating that the material may remain usable even at temperatures beyond the range investigated here.

A similarly promising trend is observed for the temperature-dependent decay curves and the corresponding decay times shown in **figure 52**. One limitation, however, is that the decay time at room temperature is about 150 μs . This value is longer than the criterion formulated in the work of *Marlies Maroka*, according to which the lifetime should not exceed approximately 100 μs . Nevertheless, the literature suggests that partial substitution of Ca^{2+} by Ba^{2+} can reduce the decay time, which could provide a possible route toward improvement [113].

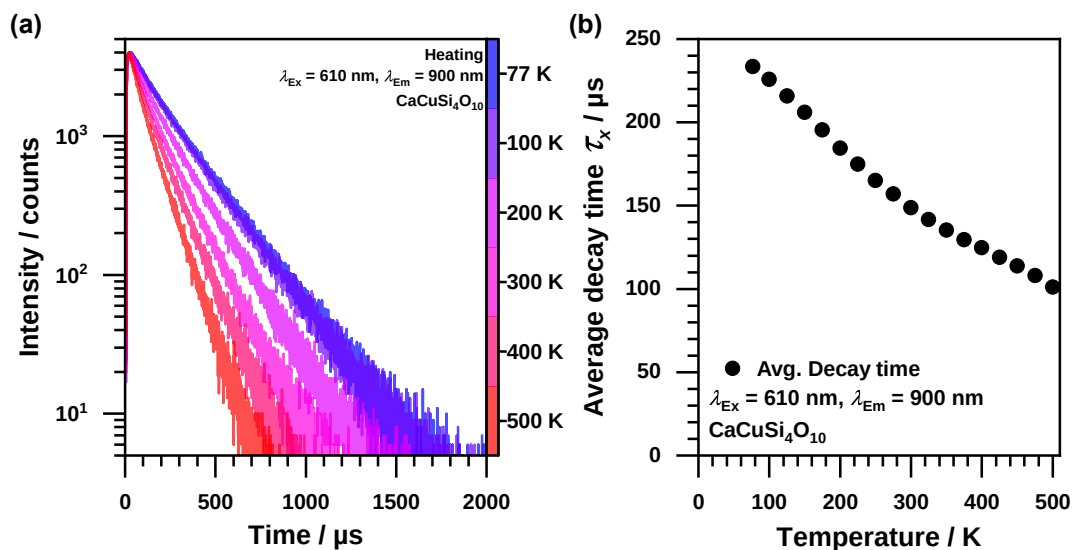


Figure 52: Decay curves recorded upon heating (a) and the corresponding average decay times (b) of $\text{CaCuSi}_4\text{O}_{10}$ powder in the temperature range from 77 to 500 K at an excitation wavelength of 610 nm and a monitored emission wavelength of 900 nm. Data provided by Dr. Julia Goldmann and Dr. Patrick Pues, AG TOM, research group of Prof. Dr. Thomas Jüstel. For clarity, error bars for the average decay times τ_x are omitted, as the corresponding fit standard deviations are smaller than the symbol size.

Overall, Egyptian blue appears to be a promising alternative phosphor for phosphor-based temperature-sensitive paints intended for higher-temperature applications. Accordingly, this material could represent an interesting starting point for future work, for example within a subsequent thesis.

7 References

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Appendix

A.1 Supplementary Material

A.1.1 Supplementary Figures

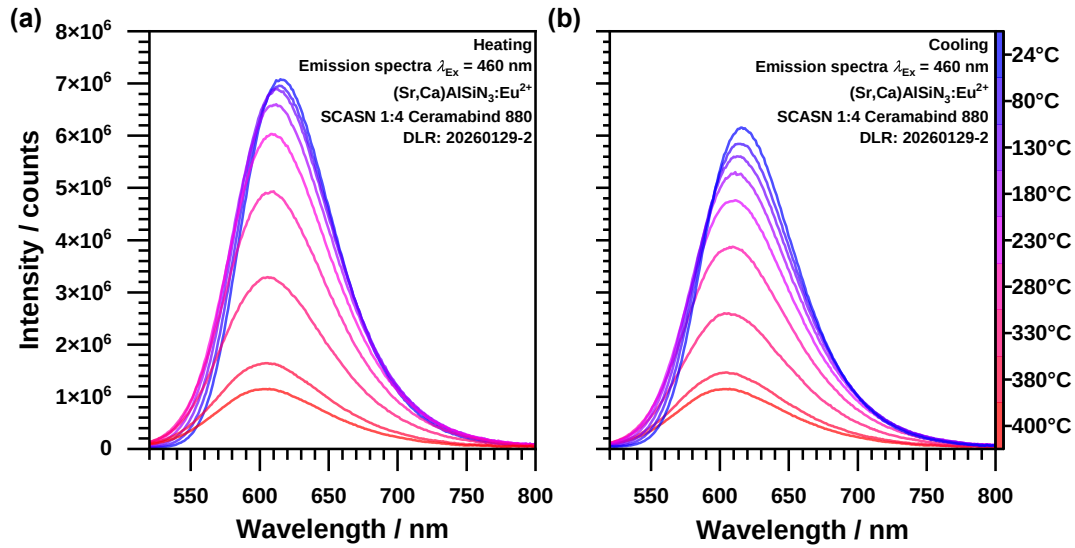


Figure A1: Emission spectra recorded upon heating (a) and cooling (b) of sample 20260129-2 (SCASN 1:4 Ceramabind[®] 880) in the temperature range from 24 to 400 °C at an excitation wavelength of 460 nm.

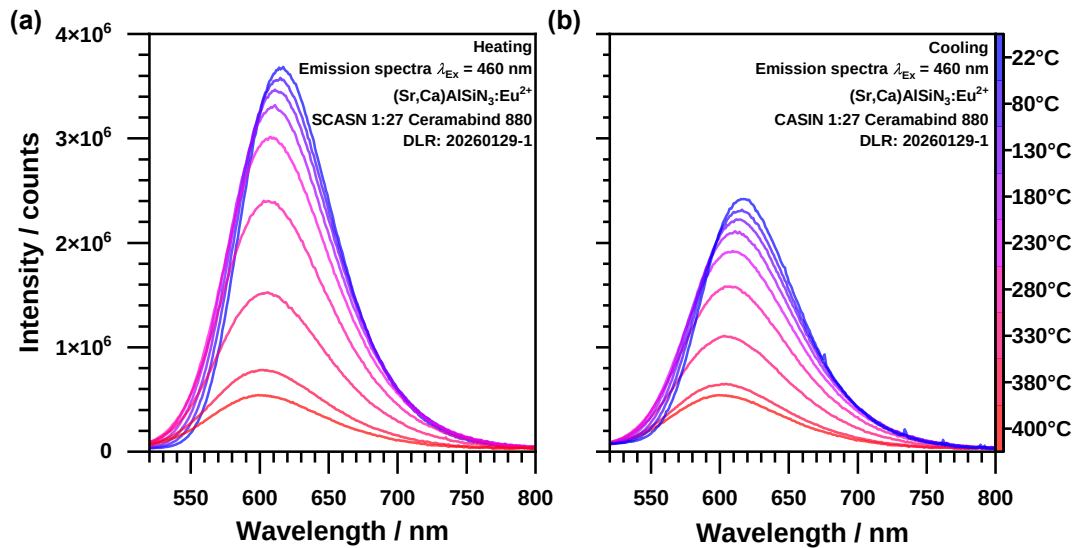


Figure A2: Emission spectra recorded upon heating (a) and cooling (b) of sample 20260129-1 (SCASN 1:27 Ceramabind[®] 880) in the temperature range from 22 to 400 °C at an excitation wavelength of 460 nm.

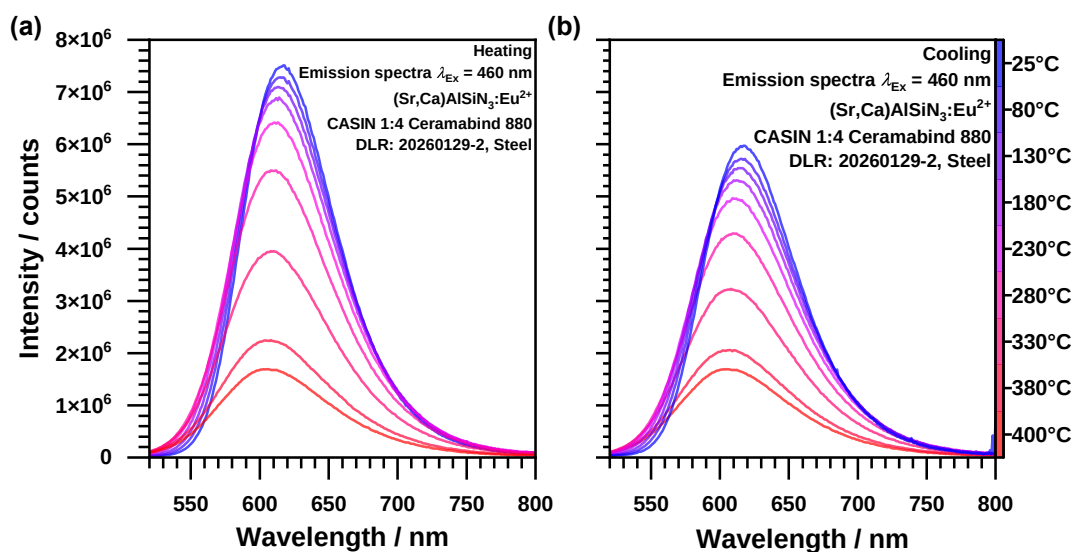


Figure A3: Emission spectra recorded upon heating (a) and cooling (b) of sample 20260129-2 (SCASN 1:4 Ceramabind[®] 880, steel substrate) in the temperature range from 25 to 400 °C at an excitation wavelength of 460 nm.

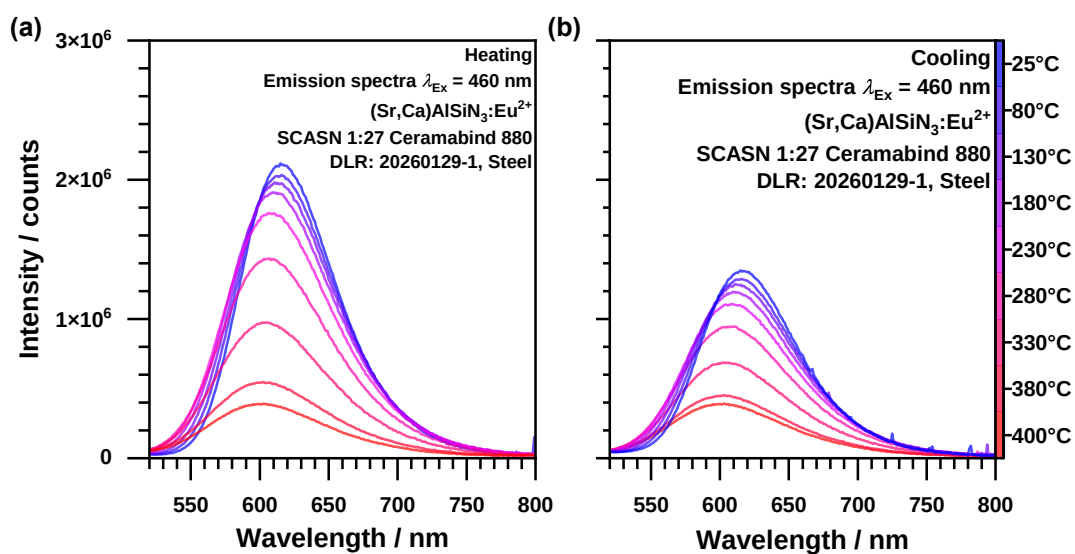


Figure A4: Emission spectra recorded upon heating (a) and cooling (b) of sample 20260129-1 (SCASN 1:27 Ceramabind[®] 880, steel substrate) in the temperature range from 25 to 400 °C at an excitation wavelength of 460 nm.

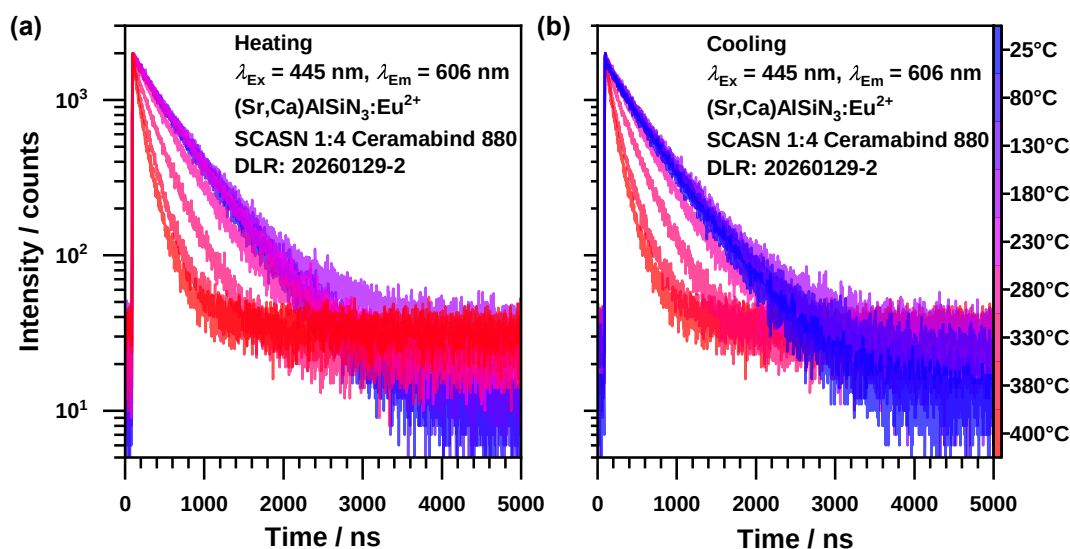


Figure A5: Decay curves recorded upon heating (a) and cooling (b) of sample 20260129-2 (SCASN 1:4 Ceramabind[®] 880) in the temperature range from 25 to 400 °C at an excitation wavelength of 445 nm and a monitored emission wavelength of 606 nm.

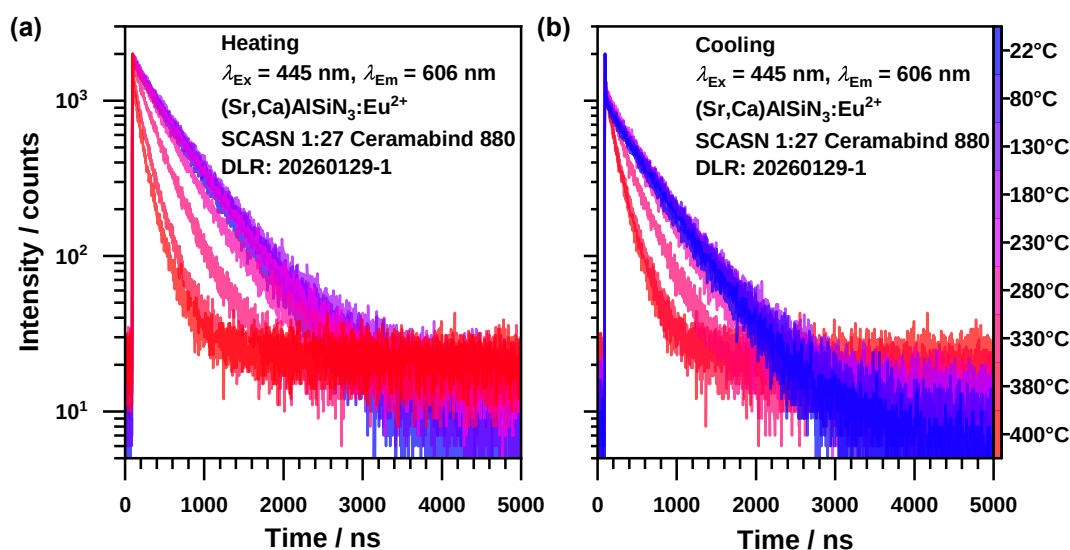


Figure A6: Decay curves recorded upon heating (a) and cooling (b) of sample 20260129-1 (SCASN 1:27 Ceramabind[®] 880) in the temperature range from 22 to 400 °C at an excitation wavelength of 445 nm and a monitored emission wavelength of 606 nm.

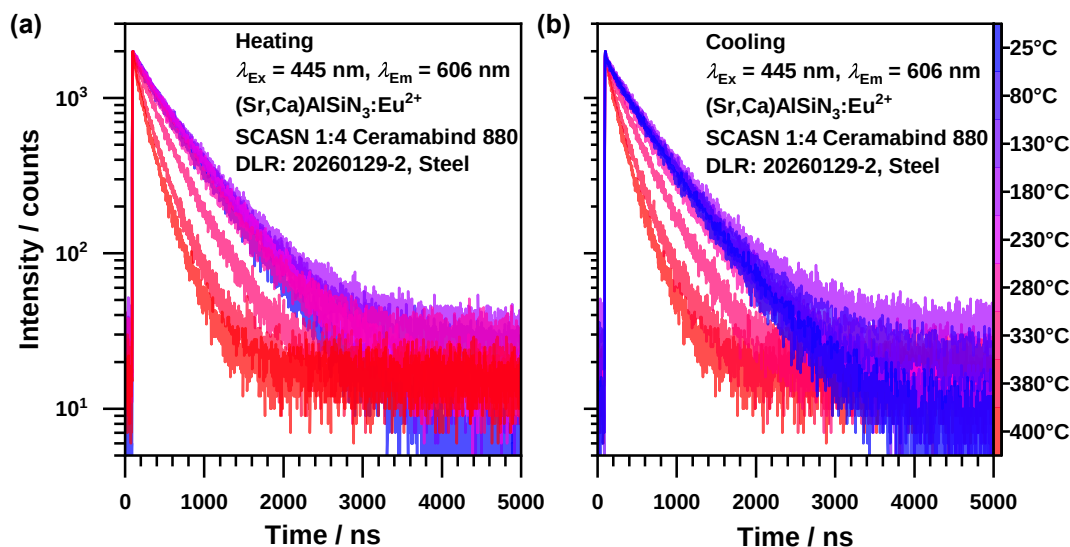


Figure A7: Decay curves recorded upon heating (a) and cooling (b) of sample 20260129-2 (SCASN 1:4 Ceramabind[®] 880, steel substrate) in the temperature range from 25 to 400 °C at an excitation wavelength of 445 nm and a monitored emission wavelength of 606 nm.

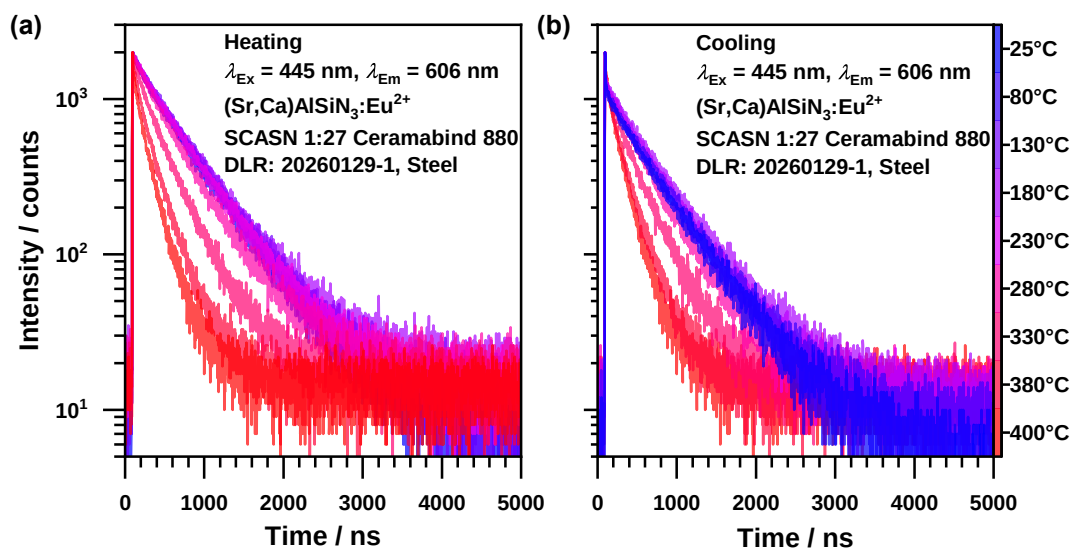


Figure A8: Decay curves recorded upon heating (a) and cooling (b) of sample 20260129-1 (SCASN 1:27 Ceramabind[®] 880, steel substrate) in the temperature range from 25 to 400 °C at an excitation wavelength of 445 nm and a monitored emission wavelength of 606 nm.

A.1.2 Photographs of Coated Samples

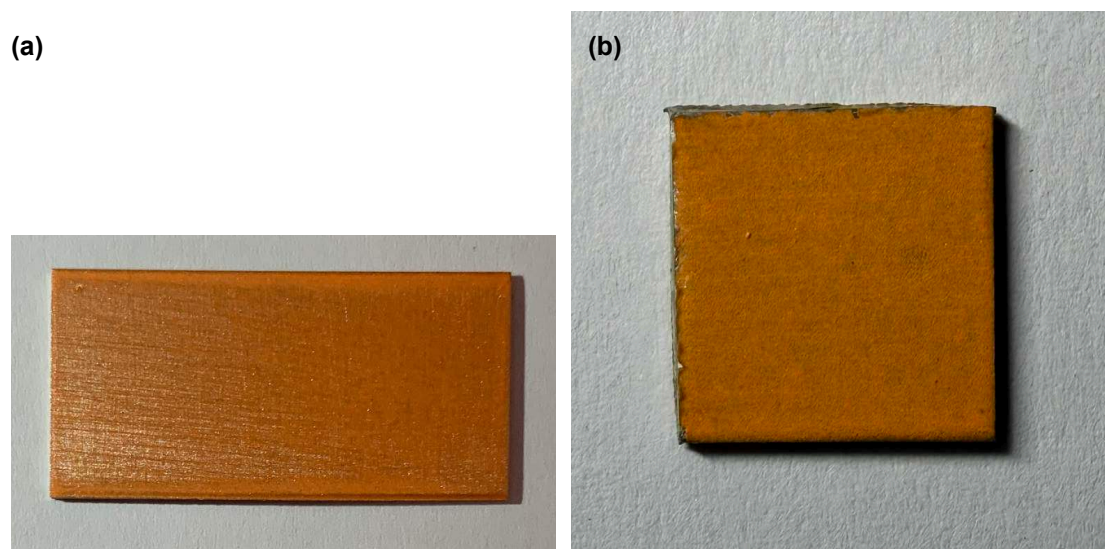


Figure A9: Representative photographs of sample 20250812-15 (SCASN 1:4 Ceramabind[®] 880) before heating (a) and after heating up to 480 °C (b). The photographs shown correspond to different specimens from the same preparation batch.

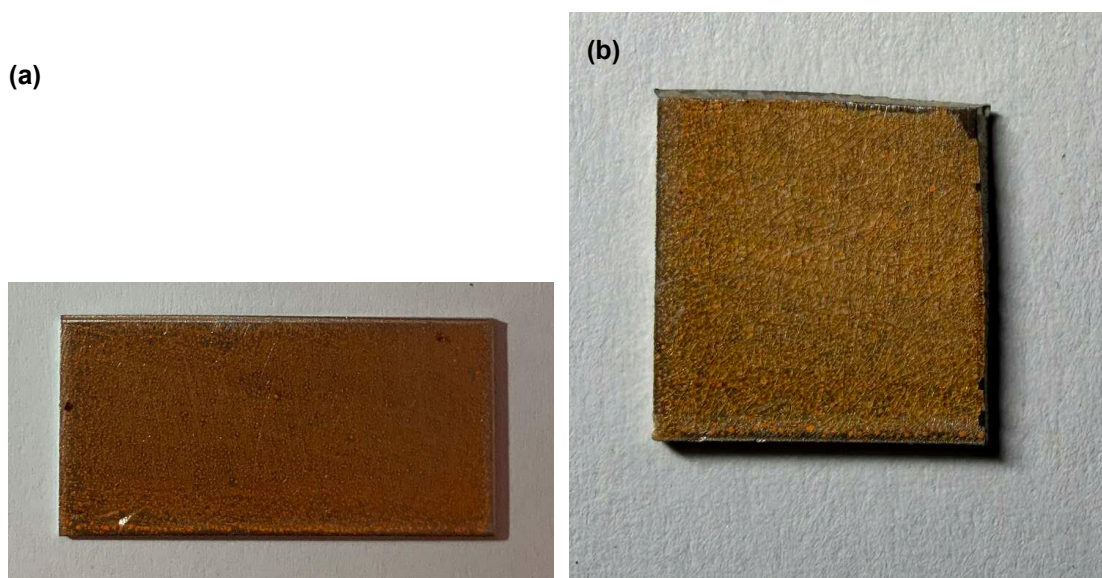


Figure A10: Representative photographs of sample 20250812-16 (SCASN 1:27 Ceramabind[®] 880) before heating (a) and after heating up to 480 °C (b). The photographs shown correspond to different specimens from the same preparation batch.

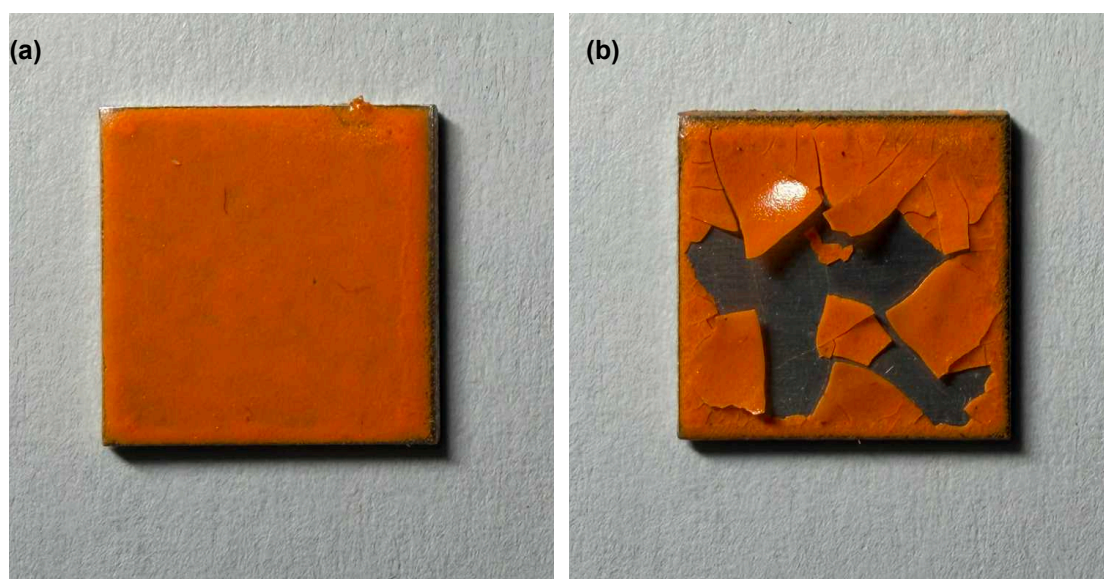


Figure A11: Representative photographs of sample 20260129-2 (SCASN 1:4 Ceramabind[®] 880) before heating (a) and after heating up to 430 °C (b). After heating strong cracking and severe delamination is visible. The photographs shown correspond to different specimens from the same preparation batch.

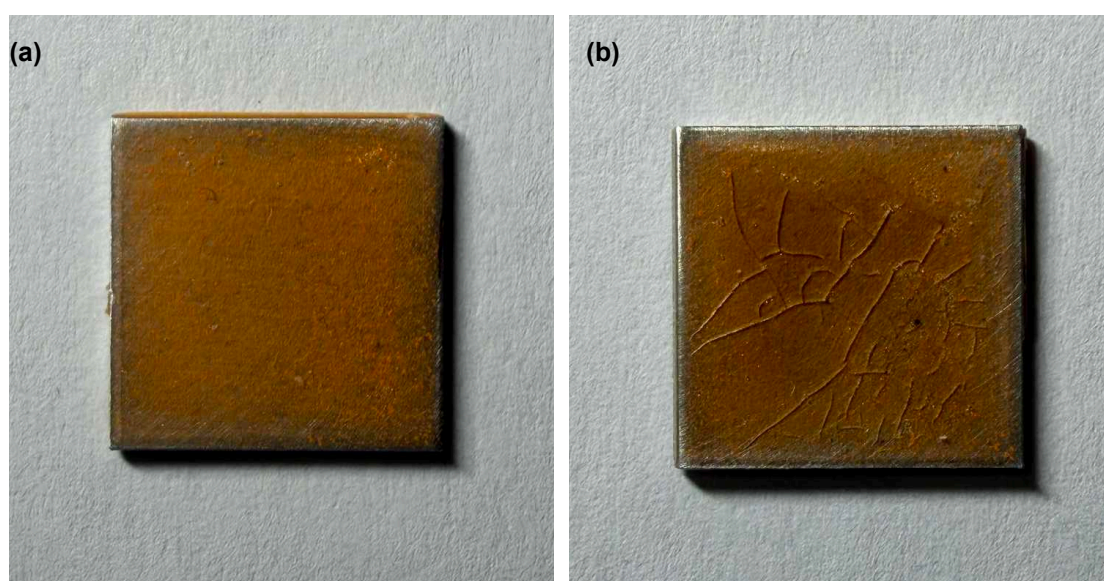


Figure A12: Representative photographs of sample 20260129-1 (SCASN 1:27 Ceramabind[®] 880) before heating (a) and after heating up to 400 °C (b). After heating strong microcracking and slight delamination is visible. The photographs shown correspond to different specimens from the same preparation batch.

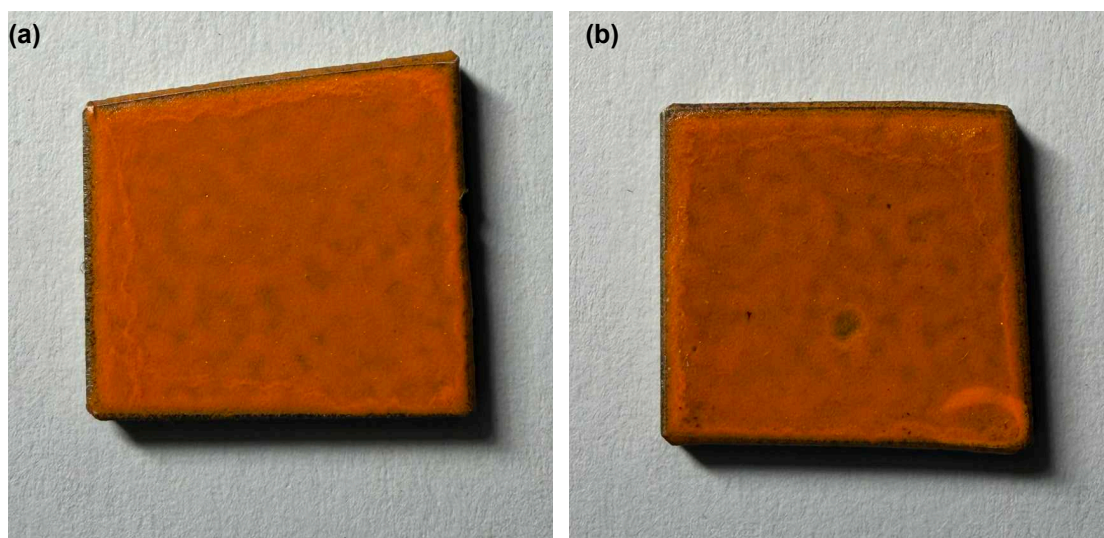


Figure A13: Representative photographs of sample 20260129-2 on steel substrate (SCASN 1:4 Ceramabind® 880) before heating (a) and after heating up to 400 °C (b). After heating only slight microcracking is visible. The photographs shown correspond to different specimens from the same preparation batch.

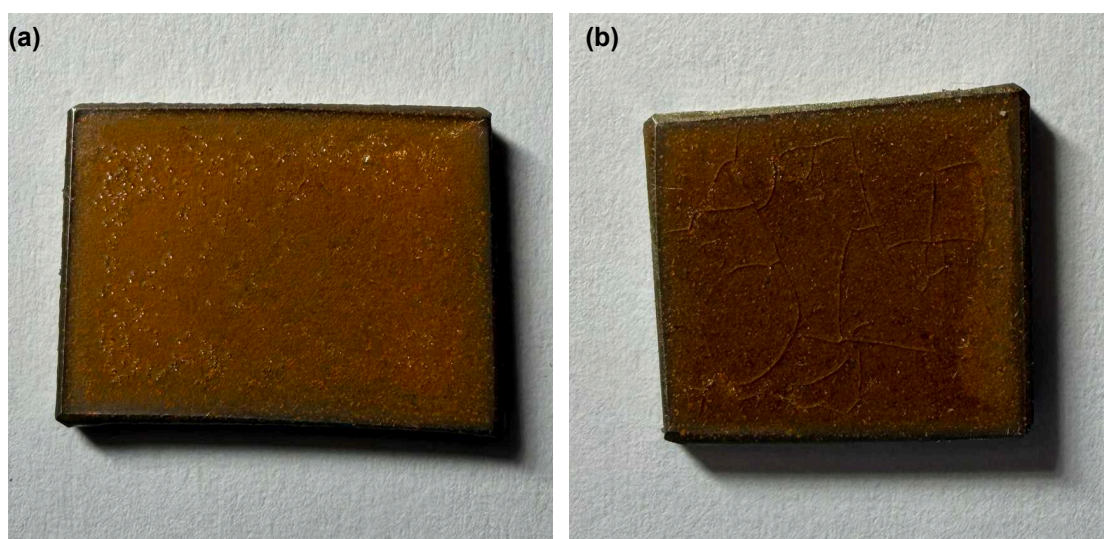


Figure A14: Representative photographs of sample 20260129-1 on steel substrate (SCASN 1:27 Ceramabind® 880) before heating (a) and after heating up to 400 °C (b). After heating strong microcracking and slight delamination is visible. The photographs shown correspond to different specimens from the same preparation batch.



Figure A15: Representative photograph of sample 20251119-1 (KLPE 1:4 Ceramabind[®] 880), showing the uneven and inhomogeneous coating surface.



Figure A16: Representative photograph of sample 20260129-3 (KLPE 1:27 Ceramabind[®] 880), showing the uneven and inhomogeneous coating surface.

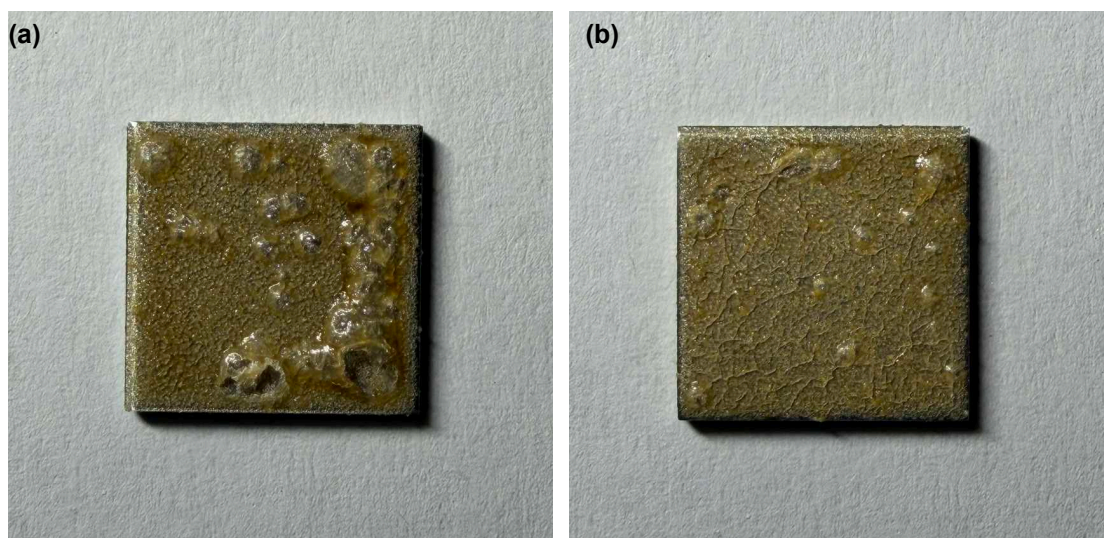


Figure A17: Representative photographs of sample 20260129-4 (KLPE 1:4 Ceramabind[®] 880) before heating (a) and after heating up to 180 °C (b). Before and after heating the uneven and inhomogeneous surface is visible. After heating significant microcracking and slight delamination is visible. The photographs shown correspond to different specimens from the same preparation batch.

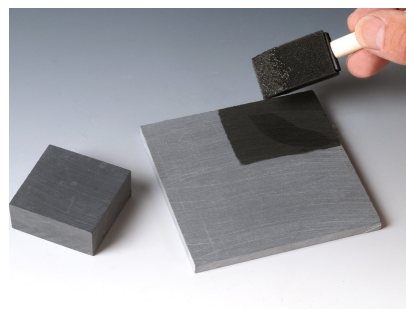
A.2 Ceramabind® 880 Datasheet and Application Guidelines



HIGH TEMPERATURE INORGANIC BINDERS

Technical Bulletin A11

Aremco's Ceramabind™ materials are unique inorganic, water-based binder systems used in the formulation of specialty adhesives, coatings, sealants and putties for applications to 3200 °F. The versatility of Aremco's Ceramabind™ products enables users to blend formulations using most ceramic, glass and metal-oxide powders. Specific properties such as coefficient of thermal expansion, thermal conductivity, dielectric strength, and chemical and moisture resistance can be optimized.



Ceramabind™ 542 seals porosity in ceramic plate.

PRODUCT HIGHLIGHTS

- 542** An acidic, etching solution which is ideal for use in adhesive systems for bonding non-porous ceramics and glass. Stable when mixed with copper. Reacts with bases such as carbonates, oxides and hydroxides of alkali metals.
- 642** A basic solution which is highly compatible with most ceramic and metal powders. Good wettability and tack, and excellent acid resistance after curing. Extremely moisture resistant after a high temperature cure. Sets up in thick cross-sections when properly formulated.
- 643-1** A basic solution compatible with most ceramic and metal powders. Excellent binder for producing high temperature protective coatings and refractory and chemically resistant adhesives and patching materials. Fully cures at low temperatures and sets up in thick cross-sections when properly formulated.
- 643-2** Similar to 643-1. Excellent for formulating thin coatings that set at room temperature and can be raised rapidly to high temperatures.
- 644-A** An acidic, colloidal alumina binder developed for mixing with sized refractory flours and grains to produce high temperature refractory coatings for ceramic fiber boards. Used as a superior standalone system to rigidize refractory fiber shapes.
- 644-S** A colloidal silica aqueous solution which produces high adhesive strength. Ideal for blending with all types of granular and fibrous ceramics. Excellent resistance to temperature, moisture and mechanical shock.
- 830** A basic solution compatible with most oxide and metal powders. Ideal for formulating high pigment-to-binder ratios to produce dense adhesives and coatings. Sets at room temperature to a moisture resistant film and does not require a heat cure. Use only for thin coating systems less than 1 mil thick.
- 875** An acidic powdered binder system used to formulate high strength, hydraulic-setting cements for electrical potting or molding applications. A powder blend is typically formulated by adding one part binder to four parts filler by weight. Water is then added in a ratio of 15–20 parts to 100 parts powder blend by weight.
- 880** High temperature, water-dispersible silicone resin for producing corrosion and moisture resistant coatings and sealing porous ceramics.

TYPICAL PRODUCT SPECIFICATIONS

Product	542	642	642A	643-1	643-2	644-A	644-S	830	875	880
pH	2.5	10.7	10.7	11.0	11.5	4.0	9.0	11.4	2.8	6.5
Specific Gravity, g/cc	1.47	1.41	1.25	1.26	1.27	1.23	1.40	1.20	1.36	1.04
Viscosity, cP	50	370	200	60	30	7	35	10	NA	480
Solids Content, % by weight	40	40	25	30	30	30	40	25	100	50
Temperature Resistance, °F (°C)	3200 (1760)	3000 (1650)	3000 (1650)	3000 (1650)	3000 (1650)	3000 (1650)	3200 (1760)	2000 (1093)	3000 (1650)	1200 (650)

APPLICATION GUIDELINES

Mixing & Application

1. Liquid binder-to-powder weight ratios of 4:1 to 1:1 are recommended when formulating adhesives, coatings and pastes. Powder binder (875) to filler ratio of 1:4 is recommended.
2. Blend powder slowly into binder until desired viscosity is achieved. Vacuum degas as required to reduce entrapped air.
3. Apply mixture to clean surfaces. Extremely smooth surfaces are difficult to wet and should be sandblasted, etched, or slightly oxidized wherever possible. Porous substrates tend to absorb and separate the binder from the powder; these substrates should be pre-coated with the binder only prior to applying the mixture.

Curing

Ceramabind™ 542

1. Air dry at room temperature for 1–2 hours.
2. Heat cure at 200 °F for 1–2 hours.
3. Heat cure at 500 °F for 1–2 hours.
4. Final cure at 700 °F for 1 hour for maximum adhesive strength and moisture resistance.

Ceramabind™ 642, 642A, 643-1, 643-2

1. Air dry at room temperature for 1–2 hours.
2. Heat cure at 200 °F for 2–4 hours.
3. Heat cure at 350 °F for 1–2 hours.
4. Final cure at 500 °F for 1 hour.

Ceramabind™ 644-A, 644-S

1. Air dry at room temperature for 2–4 hours.
2. No heat cure is required if substrate is ramped slowly at ~200 °F per hour to the operating temperature.

Ceramabind™ 830

1. Air dry at room temperature for 1–2 hours.
2. No heat cure is required.

Ceramabind™ 875

1. Air dry at room temperature for 1–2 hours.
2. Heat cure at 200 °F for 2–4 hours.
3. Final cure at 250 °F for 2–4 hours.
4. Note: This binder can also be set at room temperature in 16–24 hours without heat curing.

Ceramabind™ 880

1. Air dry at room temperature for 1–2 hours.
2. Final cure at 450 °F (≈ 230 °C) for 1 hour or 480 °F (≈ 250 °C) for 45 minutes.

Storage

Unopened containers have a six-month shelf life when stored at room temperature. Make sure opened containers are capped securely to prevent evaporation. Place a plastic film in between the cap and container to prevent air leakage. Store containers between 45 °F and 95 °F.

CERAMABIND™ COMPATIBILITY CHART

Product	542	642 / 643-1 / 642A 643-2				644-S	644-A	830	875	880
Aluminum	R	S	R	S	R	S	R	S	R	S
Aluminum Oxide	S	S	S	S	S	S	S	S	S	S
Aluminum Nitride	R	S	S	S	S	S	S	S	S	S
Boron Nitride	S	S	S	R	S	S	S	S	S	S
Brass	S	S	S	S	S	S	S	R	S	S
Bronze	S	S	S	S	S	S	S	R	S	S
Chromium	R	S	S	S	S	S	S	R	S	S
Cobalt	R	S	S	S	S	S	S	R	S	S
Copper	S	R	S	S	S	S	S	R	S	S
Dolomite	S	S	S	S	S	S	S	S	S	S
Inconel	S	S	S	S	S	S	S	S	S	S
Indium	S	S	S	S	S	S	S	R	S	S
Indium Oxide	S	S	S	S	S	S	S	R	S	S
Invar	S	S	S	S	S	S	S	S	S	S
Iron	R	S	S	S	S	S	S	R	S	S
Iron Oxide	R	S	S	S	S	S	S	S	S	S
Magnesium Oxide	R	S	S	R	S	S	S	R	S	S
Manganese Dioxide	S	S	S	S	S	S	S	R	S	S
Mica	S	S	S	S	S	S	S	S	S	S
Molybdenum	R	S	S	S	S	S	S	S	S	S
Mullite	S	S	S	S	S	S	S	S	S	S
Neodymium Oxide	R	S	S	S	S	S	S	R	S	S
Nickel	R	S	S	S	R	S	S	R	S	S
Nichrome	S	S	S	S	S	S	S	R	S	S
Silicon Dioxide	S	S	S	S	S	S	S	S	S	S
Silicon	S	S	S	S	S	S	S	R	S	S
Silicon Carbide	R	S	S	R	S	S	S	S	S	S
Stainless Steel	R	S	S	R	S	S	S	S	S	S
Tantalum	R	S	S	R	S	S	S	S	S	S
Titanium	R	S	S	R	S	S	S	S	S	S
Titanium Diboride	R	R	R	S	S	S	S	R	S	S
Titanium Dioxide	S	S	S	R	S	S	S	S	S	S
Zinc	S	S	S	R	S	S	S	R	S	S
Zirconium Carbide	R	S	S	S	S	S	S	S	S	S
Zirconium Diboride	R	S	S	S	S	S	S	S	S	S
Zirconium Oxide	S	S	S	S	S	S	S	S	S	S
Zirconium Silicate	S	S	S	S	S	S	S	S	S	S

Key

S = Stable R = Reacts

Safety

Read Material Safety Data Sheet carefully prior to use. All Ceramabind™ products are water-based materials which can be washed from the skin, in the uncured state, with mild soap and warm water. Prolonged skin contact should be avoided to prevent irritation. If any material contacts the eyes, flush continuously with water or neutralizing solutions, then consult a physician immediately.

Refer to Price List for complete order information.

Aremco Products makes no warranty express or implied concerning the use of this product.

The user assumes all risk of use or handling whether or not in accordance with directions or suggestions, or used singly or in combination with other products.