

This is the authors version of the article:

Rapid Evaluation of the Particle-Erosion Resistance of Al₂O₃ Ceramics, Composites, and Coatings using a Resonant Acoustic Mixer

by Peter Mechnich and Gözde Alkan

German Aerospace Center (DLR)

Institute of Materials Research

51147 Cologne, Germany

To cite this article: Peter Mechnich & Gözde Alkan (2023): Rapid evaluation of the particle erosion resistance of Al₂O₃ ceramics, composites, and coatings using a resonant acoustic mixer, *Advances in Applied Ceramics*, DOI: 10.1080/17436753.2023.2231230

To link to this article: <https://doi.org/10.1080/17436753.2023.2231230>

Published online: 06 Jul 2023

Rapid Evaluation of the Particle-Erosion Resistance of Al₂O₃ Ceramics, Composites, and Coatings using a Resonant Acoustic Mixer

Peter Mechnich¹, Gözde Alkan¹

¹German Aerospace Center, Institute of Materials Research, 51147 Cologne, Germany

Abstract

The solid particle technology usage in concentrated solar power plants as direct heat absorption and storage medium necessitate well selection of the materials for the components such as transport and sluice systems, which are in direct contact with moving and falling hot particles up to 1500 °C. Beyond mechanical properties, chemical inertness and high temperature stability, abrasion/erosion resistance is one of the key properties, for which, there is no easy-applicable and rapid test method exist enabling controlled lab-scale parametric studies. A novel particle impact test was established using a resonance acoustic mixer, in which ceramic particles are strongly accelerated and collide to the ceramic surface within a closed vessel. After determination of the most representative parameters such as ceramic ball size, vessel diameter and retainment/removal of debris, selected experiments were conducted on three candidate materials aimed to be used as high temperature transport/port systems; dense C 799 Al₂O₃, porous water-plasma sprayed Plascera-type Al₂O₃ and WHIPOX-type Al₂O₃/Al₂O₃ ceramic matrix composites with porous matrix; with and without porous protective Al₂O₃ coating. The distinct mass loss behavior of candidate materials highlighted the viability of the test method and the relevance of microstructures of porous Al₂O₃ materials on abrasion resistance.

1. Introduction

The utilization of solid particle technology in concentrated solar power plants offers higher storage densities and efficiencies over existing state-of-the-art technologies [1-4]. Moving ceramic particles with typical size of one mm are heated in particle receivers through point-focused solar radiation and then transported to storage containers before used to convert heat in steam generators or undergo redox-reaction used for thermochemical fuel generation such as H₂ from H₂O splitting over the CeO₂/Ce₂O₃ redox pair [5,6]. As up to 1500°C hot ceramic particles are brought in direct contact to walls of absorbers, guides,

reactors, and containers, these components must be manufactured from heat- and thermal shock resistant, low thermally conductive, and chemically inert ceramic materials. Moreover, moving ceramic particles require components with mechanical endurance, in particular good surface stability. In the particular case, particle impact, i.e. falling particles as well as moving or 'fluidized' particle beds create both erosion/abrasions loads on surfaces. In the Hotport project, German Aerospace Center (DLR) is developing a new concept comprising a rotating SiC tube as particle receiver and a high temperature transport and port system for hot ceramic particles. For this, different type of Al₂O₃-based ceramics were screened as candidate wall materials for particle guides and storage containers. Thermal, mechanical, and chemical properties of different Al₂O₃ types can be easily assessed with standard characterization methods, However, a method for screening their behavior in a fast and reproducible laboratory approach mimicking expected abrasion/erosion loads was not available. State-of-the-art erosion tests uses impinging small, sharp edged hard particles (~ 50 µm) at high particle velocities up to hundreds of ms⁻¹ mostly through compressed air [7-9]. On the other hand, impact of larger particles, commonly referred to as foreign object damage (FOD), is tested by surface hitting of single, high-impulse spheres with 1.59 mm diameter typically made from steel [10-13]. As both standard methods were not considered to duplicate well the erosion/abrasion scenario expected in the Hotport environment, a new approach using mm-sized hard ceramic balls as erosion/abrasion media and a closed vessel set-up using a high-energy resonant acoustic mixer. The influence of experimental parameters such as ball size was investigated. To assess the viability of the novel test method, three different Al₂O₃ ceramic materials with different microstructures were tested in terms of particle impact resistance. The potential of this novel approach as a fast and reproducible method for screening and ranking of materials' erosion/abrasion stability is discussed.

2. Experimental Procedure

Different commercial and in-house alumina-based materials were evaluated in terms of their resistance versus particle impact. All materials were tested in the form of plates with typical dimensions of 50x50x3 mm. Dense, standard C-799 Al₂O₃ (Degussit® Al-23; Friatec, Mannheim, Germany) and water-plasma sprayed Al₂O₃ (Plascera; LWK PlasmaCeramic, Wiehl, Germany) were tested as representatives for monolithic microstructures with widely

different grain morphology and porosity. LWK Plascera was sintered at 1200°C and surface ground by the manufacturer in order to stabilize the microstructure and reduce the surface roughness of as-sprayed plasma ceramics. In addition to the monolithic materials, a WHIPOX® Al₂O₃/Al₂O₃ ceramic matrix composite (CMC) was tested in order to evaluate the influence of fiber-reinforcement on impact resistance. WHIPOX-CMC were fabricated in-house by a filament winding utilizing Nextel™610 3000 DEN continuous Al₂O₃ fibers and a proprietary, porous Al₂O₃ matrix. After winding, plate formation, and drying, CMC were sintered at 1300°C for 2 h in air. CMC were cut and ground in order to obtain smooth surfaces and exhibited a typical fiber volume content of about 40 %. Moreover, WHIPOX® CMC with additional coatings consisting of porous, reaction-bonded alumina (RBAO) were tested. RBAO coatings were fabricated by dip-coating or painting ground CMC substrates with dispersed micron-sized Al/ Al₂O₃ powder mixtures and subsequent sintering in air (1300°C, 1h). During heat-treatment Al is oxidizing and in-situ forming Al₂O₃ which creates strong bonding between coating particles as well as to CMC substrate. Details of the RBAO process can be found in reference [14, 15].

A resonance acoustic mixer 'RAM' (LabRAM-II, Resodyn, Butte, MT, USA) was utilized to test particle impact resistance of samples, which enables the assesment of various particle sizes using the same set-up. The ceramic substrates are placed on top of a vessel containing a fixed milling ball mass of 2 g and fastened tightly in the RAM sample holder A schematic shown of experimental set-up is presented in Figure 1. Two vessels with same height but different internal diameters were examined; 27 mm and 54 mm; respectively. 100 g RAM acceleration for 100 minutes was used in each test to induce particles collision to the sample surface resulting in abrasion. After each 10 minutes, test was interrupted to weigh the samples after removing the sticking dust by pressurized air. The difference in weight was divided by the cross-sectional area of plastic vessels which determines the interaction surface between balls and ceramic plate. In order to minimize possible interference on results, we employed a quasi-wear-free experimental set-up with HD-polyethylene vessels and yttria-stabilized ZrO₂ milling balls with 0.5 mm, 1 mm, 2 mm, and 5 mm diameter (YTZ®, Tosoh Co., Japan) as test media, respectively.

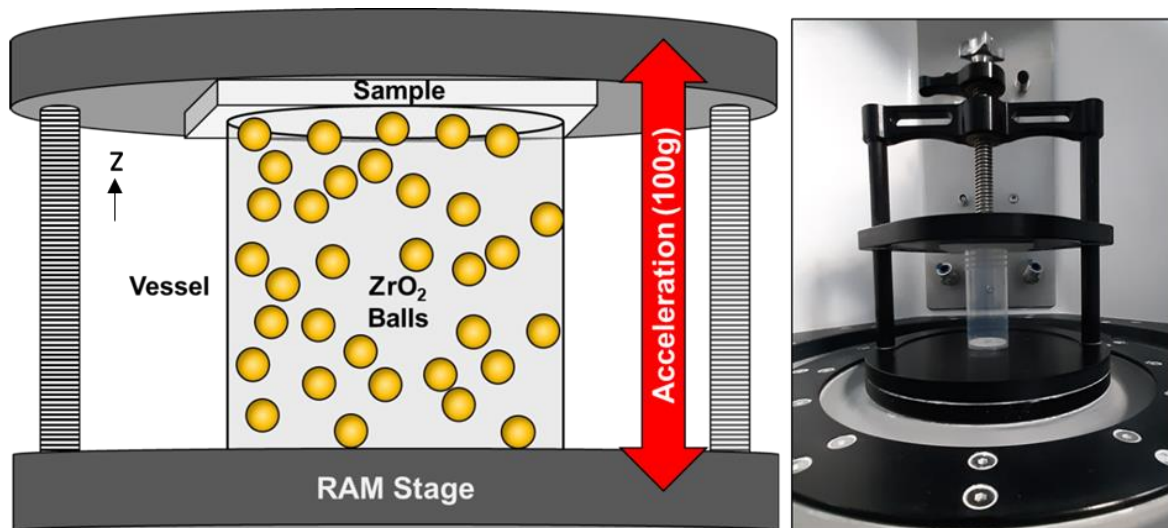


Figure 1. Evaluation of the particle impact resistance of ceramic plates in a resonant acoustic mixer (RAM). Left: schematic; right: Resodyn LabRAM-II with mounted sample

The room temperature hardness was measured by Vickers indentation (VT 2000, Clemex Technology Inc., Quebec, Canada). Detailed surface investigation of the ceramic in as-received state and after abrasion tests were performed by SEM in low vacuum condition (SU 3800, Hitachi High-Tech Europe, Krefeld, Germany).

3. Results and Discussion

3.1. Experimental set-up and parameter assessment

For the novel RAM test method, a series of experiments was performed to gain understanding on key parameters and deduce viable conditions for the envisaged rapid and straightforward assessment of different Al₂O₃ materials. As described in the experimental section, for the determination of experimental parameters, ball mass, time increment and RAM acceleration were kept constant and WHIPOX CMC were used as a test material. In a first experimental set-up, the influence of debris on abrasion/erosion dynamics was studied. For this purpose, two series were performed: (1) debris was kept in the system by retaining the 27 mm vessel and 1 mm balls (2) debris was removed from the system after each cycle by renewing vessel and balls, as represented in Figure 2.

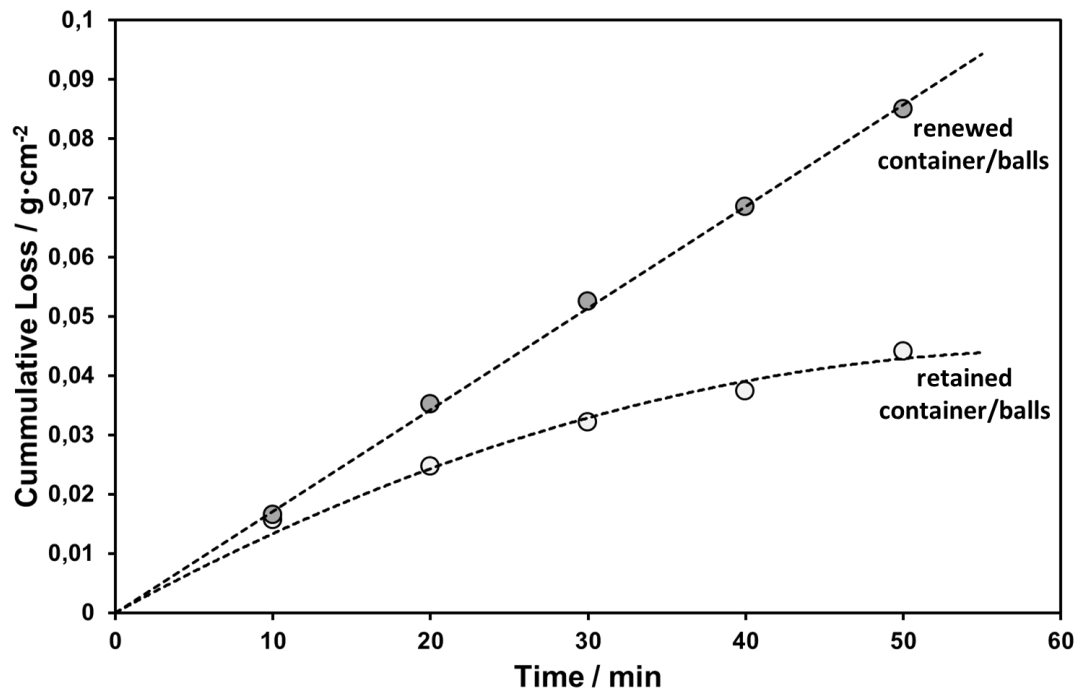


Figure 2. Effect of vessel and balls renewal on mass loss of CMC material

Figure 2 highlights the significant mitigation effect of accumulated debris on the mass loss kinetics especially after 20 minutes, evidently caused by ball energy dissipation by the newly formed powder or ‘aerosol’ inside the vessel. Whereas ‘mitigation’ by accumulated debris may be a more realistic scenario in application, the ‘unmitigated’ approach was used for all consecutive experiments. Since this study is for the exploration of parameter space but not quantitative material analysis, the unmitigated approach eases the interpretation of results and direct comparison of various parameters.

A second series of experiments was performed to study the influence of vessel size, which defines the interaction area between YTZ balls and ceramic substrate, on the mass loss kinetics. Total mass of balls was kept constant while varying the vessel size.

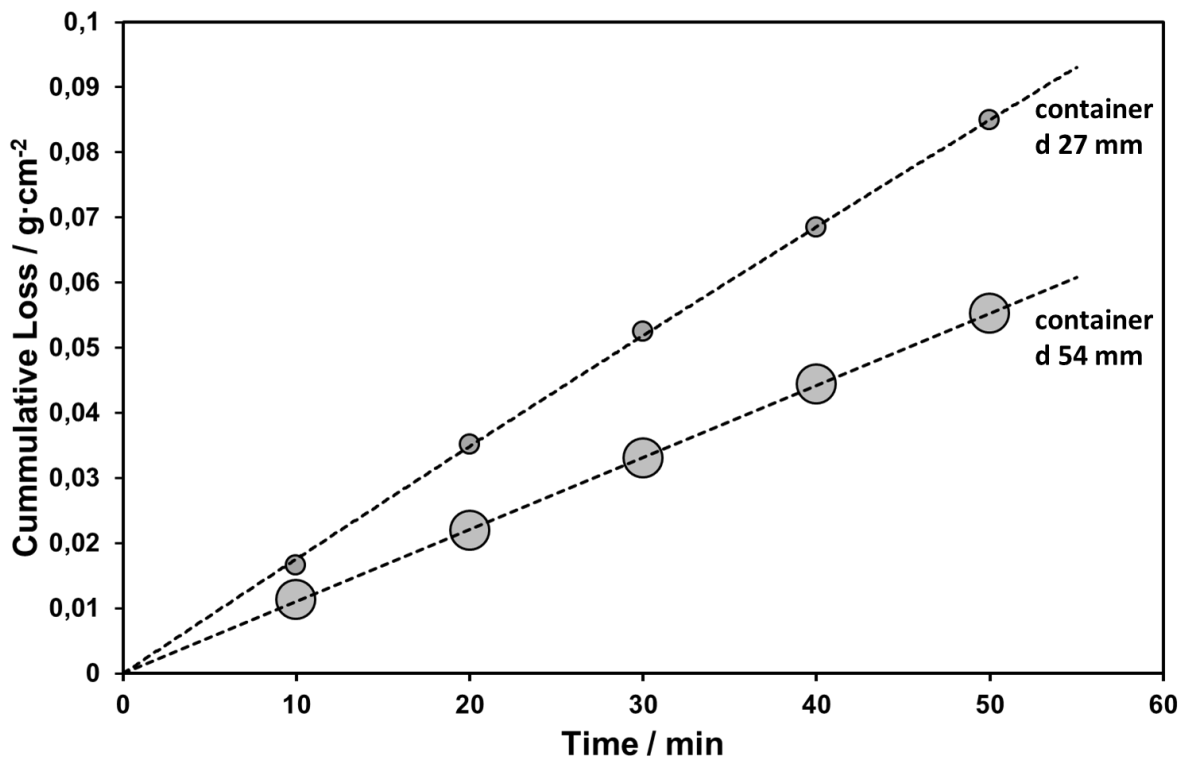


Figure 3. Effect of vessel diameter on cumulative mass loss of CMC material in RAM testing

Figure 3 shows the relative higher mass loss observed for the 27 mm vessel which was not expected at the beginning. As both vessels have the identical height of 70 mm, a similar acceleration of balls is assumed. The vessel diameters of 27 and 54 mm, however, lead to much different sample exposure areas and vessel volumes. It seems plausible that larger 54 mm vessels increase the surface impact probability and reduces the probability of ball-ball collisions. On the other hand, ball-wall collisions resulting in ball trajectories and impact angles far away from the 'z'-direction are more likely in 54 mm vessels and are plausible reasons for diminished ball kinetic energy. Combined effects may explain informally the observed behavior. Again, it was concluded that the 27 mm vessels are the best choice for further experiments. The effect of another key experimental parameter, the YTZ ball size, on abrasion related mass loss was investigated using four different ball diameters: 0.5 mm, 1 mm, 2 mm, and 5 mm, respectively.

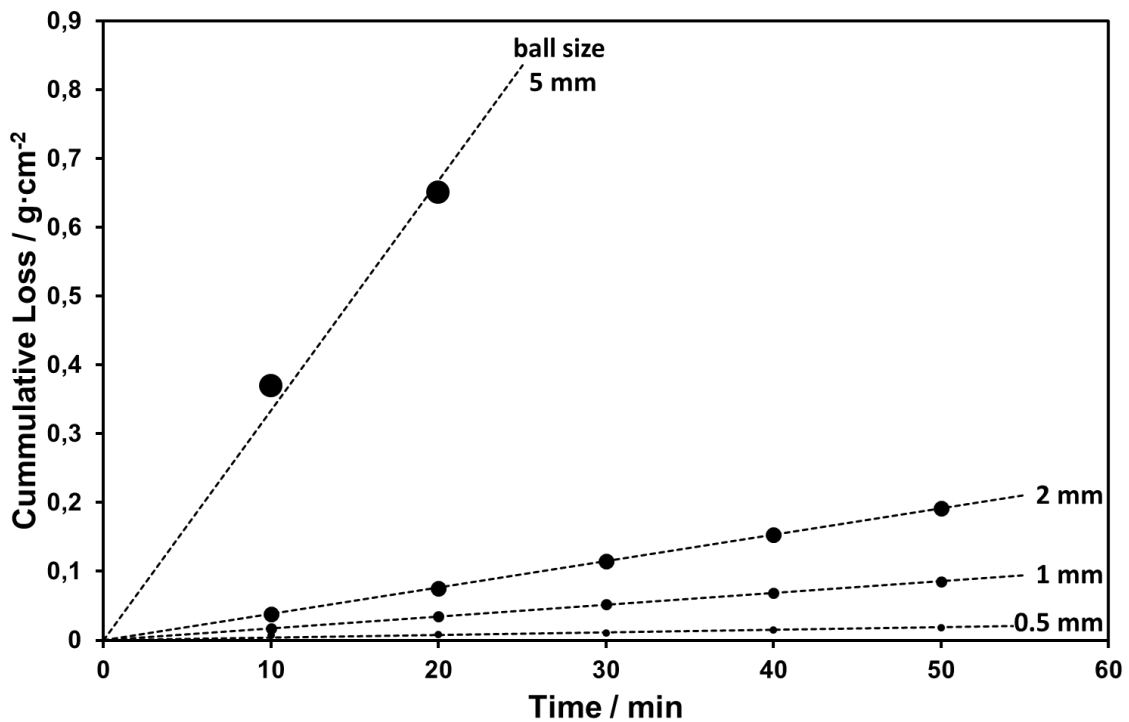


Figure 4. Abrasion related mass loss of CMC material depending on sizes of employed YTZ balls. The 5 mm sample was fully perforated and had to be stopped after two cycles

In order to provide similar kinetic energy for each experiment, YTZ ball mass was set arbitrarily to 2 grams, respectively, which leads to much different ball counts: For example, 2 g corresponds to either five 5 mm balls (individual mass $\sim 4 \cdot 10^{-1}$ g) or about 5000 0.5 mm balls (individual mass $\sim 4 \cdot 10^{-3}$ g). The strong influence of ball size on cumulative mass losses of CMC materials can be seen in Figure 4. The smallest zirconia balls with a diameter of 0.5 mm produce only minor mass loss of CMC material. As ball size increases, significantly higher mass losses are observed. In the case of 5 mm YTZ balls, interaction was so severe that the CMC sample was fully perforated and broke after only two consecutive runs. As ball volume, thus kinetic energy, is related to the third power of the ball diameter ($V \sim \pi d^3$) the destructive behavior of 5 mm YTZ balls is not surprising. Moreover, low ball count results in strongly decreasing probability for ball-ball and ball-wall collisions, resulting in very high kinetic impact on the CMC surfaces. On the contrary, experiments using small 0.5 mm YTZ balls are characterized by low individual kinetic energies and high collision-related energy losses. After assessment of four different variants, both 1 mm or 2 mm YTZ balls appeared to be suitable for the current test campaign. The 1 mm variant was selected as the most representative for subsequent

comparative material tests. Nonetheless, it is easy to determine suitable ball diameter/mass combinations for other test materials. Moreover; it is also worth to mention here that 1 mm particle size was selected for the zirconia balls. In the case of another ball material, depending on the material specific weight, the particle impact related mass losses are expected to be different. The data points of mass loss curves suggest a linear relationship. However, this could be misleading because the substrate surfaces show progressive pitting and eventually will be perforated (as happened in case of the 5mm balls). Consequently the substrate surface area exposed to hitting balls is not constant over time. Therefore, we did not perform a linear regression analysis for deriving any kind of mass loss 'rates'.

3.2 Screening the particle impact resistance of Al₂O₃ materials with different microstructures

After determination of suitable experimental parameters with Al₂O₃/Al₂O₃ CMC, two more monolithic Al₂O₃ materials with different microstructure were selected to elucidate the importance of grain morphology and porosity on their respective abrasion/erosion behavior. The cumulative mass loss of Whipox-type CMC, dense C 799 Al₂O₃, and water plasma sprayed Plascera-type Al₂O₃ were tested in a comparative manner using 27 mm vessels and 1 mm YTZ balls as displayed in Figure 5.

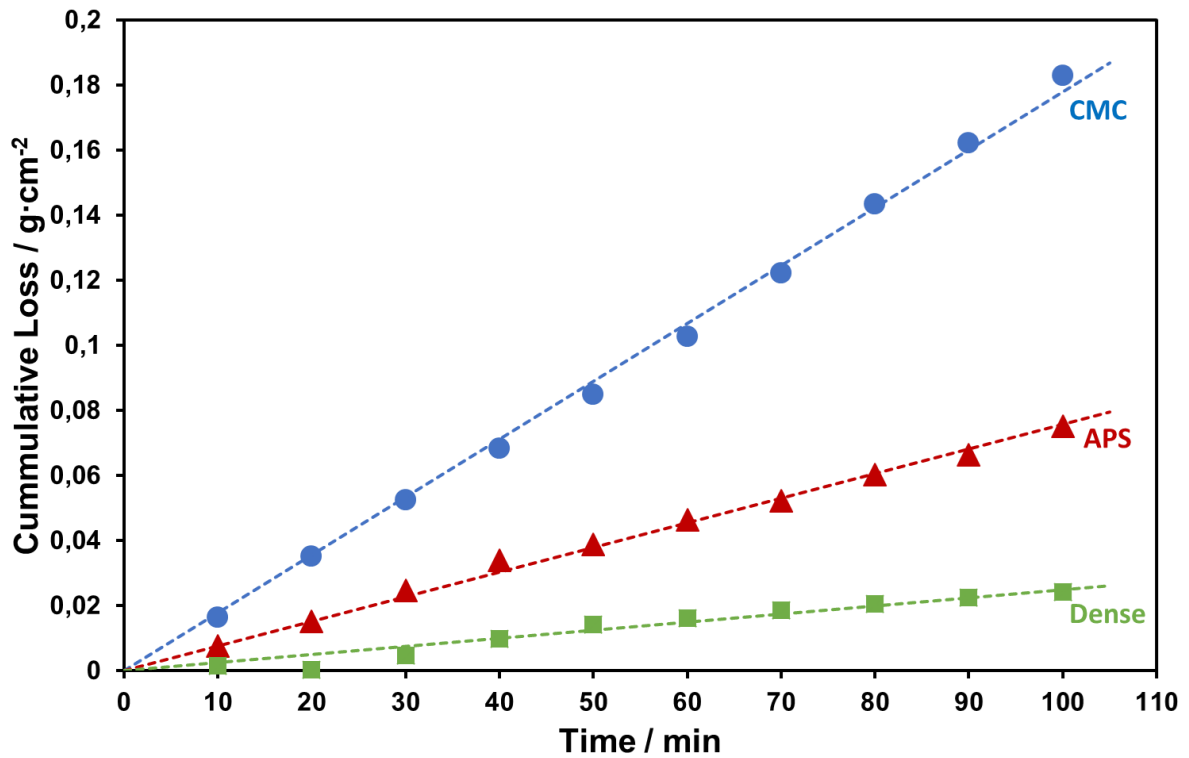


Figure 5. RAM particle impact behavior of Al₂O₃ types: dense C 799 (squares), water plasma-sprayed Plascera (triangles) and filament wound WHIPOX-type CMC (circles)

Table 1. Hardness and density values of Al₂O₃ types: dense C 799, water plasma-sprayed (APS) and filament wound WHIPOX-type CMC

	Dense	APS	CMC
Hardness (GPa)	17	2.1	2.4 (Matrix)
Density (gcm⁻³)	3.9	3.2	3.0
Density (% of theoretical)	100	82	77

At each stage of the experiment, the highest cumulative mass loss is observed for WHIPOX-type CMC. The highest impact resistance against YTZ balls was observed for dense C 799 ceramics, while Plascera-type Al₂O₃ exhibited an intermediate stability. Whereas the much higher stability of C 799-type Al₂O₃ is easily explained by its high density of 3.9 gcm⁻³ and hardness of 17 GPa (supplier data), mass loss behavior of both porous variants must be regarded in view of their distinct properties as listed in Table 1: Plascera Al₂O₃ has a slightly

higher bulk density (3.2 gcm^{-3}) than WHIPOX (3.0 gcm^{-3}). However, WHIPOX contains a significant volume fraction of dense Al_2O_3 fibers. Assuming a fiber volume content of 40 %, the matrix density is calculated to 2.4 gcm^{-3} only. In terms of hardness, Plascera (2.1 GPa, supplier data) [16] is lower than the average value measured for the CMC matrix (2.4 GPa). It must be emphasized, however, that Vickers indents were positioned only in homogeneous matrix areas, i.e. areas with large pores were excluded, resulting in an overestimated hardness value. Thus, there is no simple correlation between Vickers hardness of CMC matrix and CMC impact resistance due to the complex microstructure of fibers, matrix, and pores.

All ceramic substrate surfaces were investigated after cumulated 100 minutes of RAM impact tests by SEM. Each image in Figure 6. displays the unaffected zone (upper left part) along with the impacted areas (lower right part), highlighting the different grades of initial surface quality and post-experiment damaging of Al_2O_3 -types: while C 799 exhibits only superficial roughening, Plascera and especially WHIPOX exhibit formation of deep craters with rough surfaces, as consistent with cumulative mass loss curves in Figure 5.

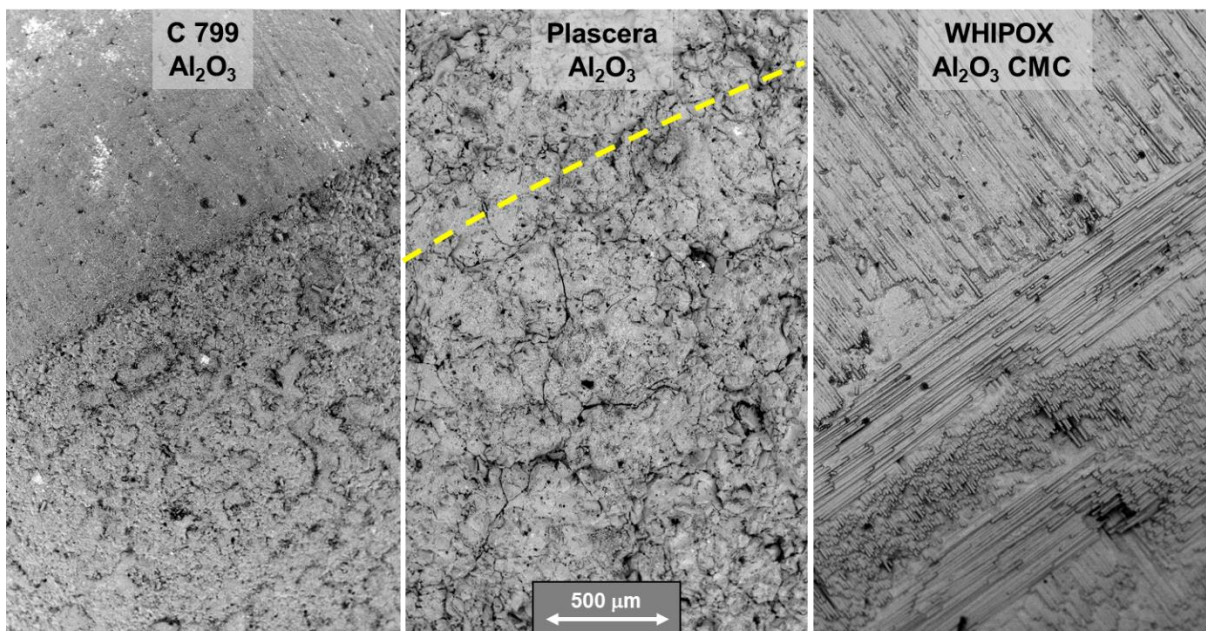


Figure 6. Surfaces of dense C 799, plasma-sprayed Plascera and WHIPOX-type CMC after 100 min RAM particle impact tests. SEM top views show boundary (dashed line) between initial (upper left parts) and post-experimental appearance (lower left parts)

The relatively poor impact performance of WHIPOX-CMC was the motivation for testing of identical material with additional reaction-bonded Al_2O_3 coatings (RBAO). Similar RBAO

coatings were used solely as top-coats well as surface-stabilizing bond-coats for plasma-sprayed Y_2O_3 coatings for WHIPOX-type CMC. Figure 7. shows the RAM impact tests of uncoated and RBAO-coated CMC. Evidently, the cumulative mass loss of the latter is significantly lower in early stages up to 40 minutes test time. Between 40 and 50 min, a significantly increasing slope of the mass loss curve indicates loss of the protective effect of the RBAO coating. In the later stages, the slope of the mass loss curve of the RBAO sample remains still lower, this may be explained by additional matrix sintering induced by RBAO processing at $1300^\circ C$.

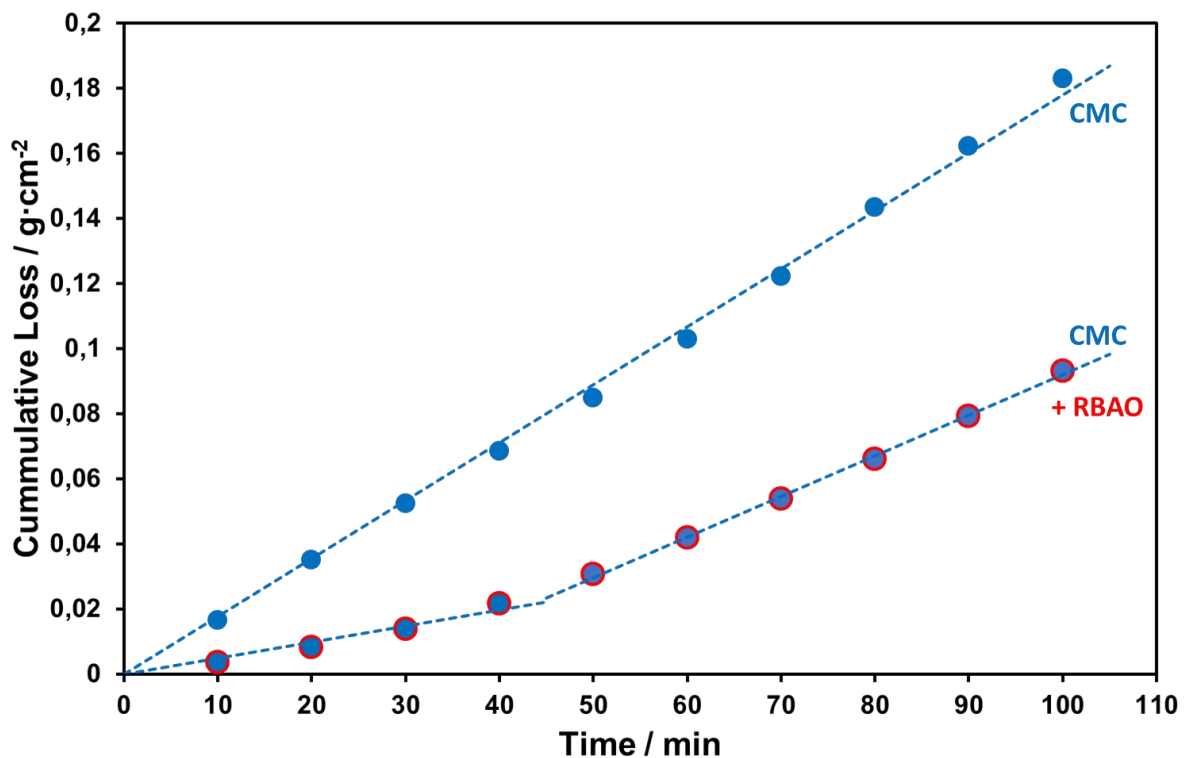


Figure 7. Effect of reaction-bonded alumina coating on cumulative mass loss of CMC. Increasing slope of the mass loss curve after 40 min indicates coating depletion

The SEM images in Figure 8 show the porous, but well bonded microstructure of RBAO and its surface erosion behavior in RAM testing. In general, it can be stated that additional RBAO coatings can raise the impact resistance of WHPOX -CMC to the level of plasma-sprayed Al_2O_3 . Results are also emphasizing the viability of the presented RAM test method for rapid materials screening.

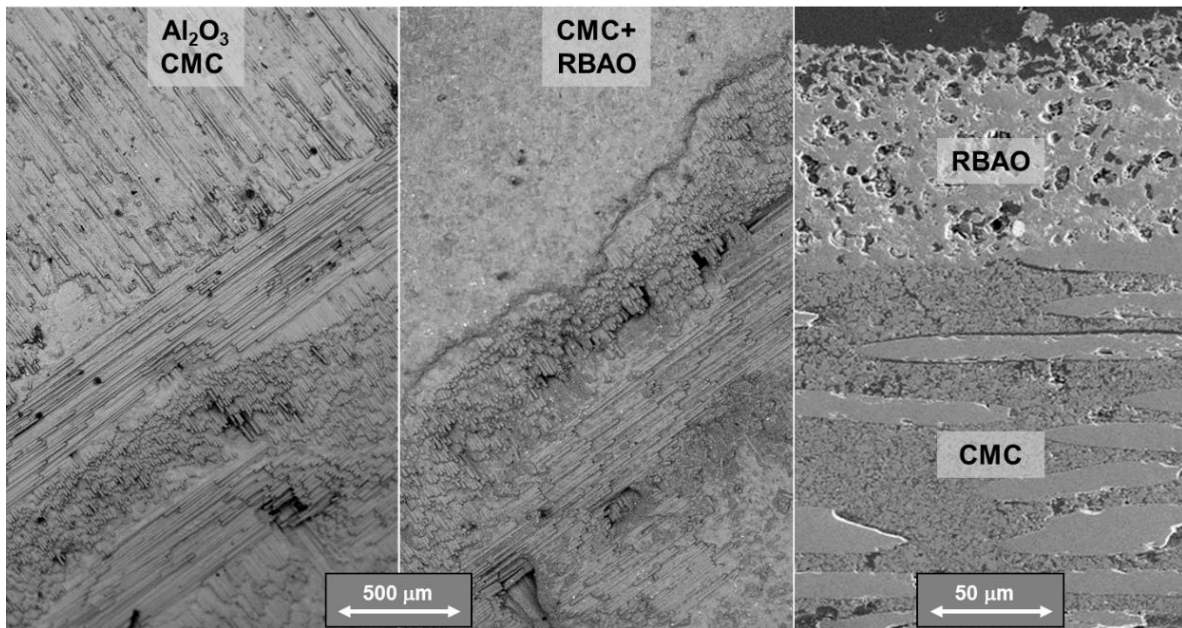


Figure 8. Surfaces of WHIPOX-type CMC without and with RBAO coating after 100 min RAM particle impact test, detailed view of the porous RBAO and CMC cross-section microstructure

4. Conclusions

A resonance acoustic mixer (RAM) has been employed as a new and straightforward particle impact test method for rapid screening of Al₂O₃ materials with significantly different microstructures. In the present campaign, suitable experimental parameters were elaborated using 27 mm diameter closed polyethylene vessels and quasi wear-free 1-2 mm sized Y-stabilized ZrO₂ balls as abrasive media. The closed vessel set-up is particularly suitable to study impact degradation when vessel and balls are renewed after subsequent impact experiments to minimize debris damping effects. The RAM test method can easily be adapted to other test materials by use of alternative abrasive media having different specific weights, such as Si₃N₄ or steel balls, providing a wide range of impact energy. A considerable advantage of the closed vessel set-up is the possibility to capture or recover debris, which can facilitate testing of materials containing critical or hazardous constituents.

As a case study, three materials aimed to be used for high temperature particle transport components for solar receivers were examined: dense C 799 Al₂O₃, water plasma sprayed Plascera-type Al₂O₃, and WHIPOX-type Al₂O₃/Al₂O₃ ceramic matrix composites (CMC). Cumulative mass losses after up to 100 minutes of RAM particle impact testing and

following microstructural analysis by SEM revealed the strong influence of microstructure on abrasion behavior. The highest impact resistance was observed by dense Al₂O₃, followed by porous, water-plasma sprayed Al₂O₃. The highest mass loss due to the poor resistance is observed in the case of WHIPOX-type CMC. Distinct impact resistance of the two monolithic Al₂O₃ variants is easily explained considering their density and hardness values, where increasing density and hardness enhance the impact resistance. On the other hand, the average properties of the porous Al₂O₃ matrix of the CMC seems to be mainly governing the observed poor impact resistance. The application of a protective RBAO-type coating can significantly increase the impact resistance of Al₂O₃/Al₂O₃ CMC materials, which makes them promising candidates for the envisaged application.

Acknowledgment

This work was funded by the EFRE-Project Hotport No. 0801589

Disclosure statement

The authors report there are no competing interests to declare.

5. References

[1] Palacios, A.; Barreneche, C.; Navarro, M.E.; Ding, Y. Thermal Energy Storage Technologies for Concentrated Solar Power- A Review from a Materials Perspective. *Renew. Energy* 2020, 156, 1244–1265. <https://doi.org/10.1016/j.renene.2019.10.127>.

[2] Sarbu, I.; Sebarchievici, C. A Comprehensive Review of Thermal Energy Storage. *Sustainability* 2018, 10, 191. <https://doi.org/10.3390/su10010191>

[3] Calderón, A.; Barreneche, C.; Palacios, A.; Segarra, M.; Prietto, C.; Rodriguez-Sanchez, A.; Fernandez, A.I. Review of solid particle materials for heat transfer fluid and thermal energy storage in solar thermal power plants. *Energy Storage* 2019, 1, e63. <https://doi.org/10.1002/est2.63>.

- [4] Siegel, N.; Gross, M.; Ho., C.; Phan, T.; Yuan, J. Physical Properties of Solid Particle Thermal Energy Storage Media for Concentrating Solar Power Applications. *Energy Procedia* 2014, 49, 1015–1023. <https://doi.org/10.1016/j.egypro.2014.03.109>.
- [5] Knoblauch, N ; Mechnich, P. A Novel Method for the Preparation of Fibrous CeO₂–ZrO₂–Y₂O₃ Compacts for Thermochemical Cycles. *Crystals* 2021, 11(8), 885. <https://doi.org/10.3390/cryst11080885>
- [6] Lee, K.; Knoblauch, N.; Agrafiotis, C.; Pein, M.; Roeb, M. Oxidation kinetics of La and Yb incorporated Zr.doped ceria for solar thermochemical fuel production in the context of dopant ionic radius and valence. *Open Ceramics* 2022, 10 (1000269) <https://doi.org.10.1016/j.oceram.2022.100269>
- [7] Pathak, M.S.; Kumar, V. P.; Bonu; V.; Latha, S.; Mishnaevsky L., Lakshmi, R. V.; Bera, P.; Barshilia, H.C. Solid particle erosion studies of ceramic oxides reinforced water-based PU nanocomposite coatings for wind turbine blade protection. *Ceramics International* 2022, 48 35788–35798. <https://doi.org/10.1016/j.ceramint.2022.07.143>
- [8] Suh, M-S.; Hinoki, T.; Kohyama, A. Erosive Wear Mechanism of New SiC/SiC Composites by Solid Particles. *Tribol Lett* 2011, 41, 503–513. <https://doi.org/10.1007/s11249-010-9658-5>
- [9] Berndt, C.; Herman, H. Solid particle erosion of plasma sprayed ceramic coatings. *Materials Research* 2004, 7(1), 147-153
- [10] Presby, M. J.; Harder, B. J. Solid Particle Erosion of a Plasma Spray – Physical Vapor Deposition Environmental Barrier Coating in a Combustion Environment. *Ceramics International* 2021, 47(5) <https://doi.org/10.1016/j.ceramint.2021.05.154>
- [11] Kedir,N.; Faucett, D.; Sanchez, L.; Choi. R.S. Foreign Object Damage in a SiC Fibrous Composite. *Ceramic Transactions* 2018, 261, 33-44 <https://doi.org./10.1002/9781119423829.ch4>
- [12] Faucett, D. C.; Wright, J.; Ayre, M.; Choi, S. R. Foreign Object Damage (FOD) in Thermal Barrier Coatings. *Ceramic Transactions* 2014, 234 (244-255) <https://doi.org/10.1002/9781118491867.ch25>

[13] Kedir, N; Garcia, E.; Kirk, C.; Guo, Z.; Gao, J.; Zhai, X.; Sun, T.; Fezzaa, K.; Sampath, S.; Chen, W. W. In situ characterization of foreign object damage (FOD) in environmental-barrier-coated silicon carbide (SiC) ceramic. *Journal of the American Ceramic Society* 2020, 103(5). <https://doi.org/10.1111/jace.17165>

[14] Mechnich, P.; Braue, W.; Schneider, H.; Multifunctional Reaction-Bonded Alumina Coatings for Porous Continuous Fiber-Reinforced Oxide Composites. *Int. J. Appl. Ceram. Technol.* 2004, 1(4) 343-550 <https://doi.org/10.1111/j.1744-7402.2004.tb00185.x>

[15] Mechnich, P.; Braue, W. Air Plasma-Sprayed Y₂O₃ Coatings for Al₂O₃/Al₂O₃ Ceramic Matrix Composites. *J. Eur. Ceram Soc.* 2013, 2645-2653. <https://doi.org/10.1016/j.jeurceramsoc.2013.03.034>

[16] Lutz, H. E. Microstructure and Properties of Plasma Ceramics. *Journal of the American Ceramic Society* 1994, 77(5)