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Thermal shock behaviour of laminated multilayer refractories for steel casting applications reinforced by residual stresses

Daniel Jakobsen [a], Wei Zhang [b], Nikolay Doynov [b], Anna Böhm [c], Jürgen Malzbender [c], Vesselin Michailov [b], Andreas Roosen [a,*]

[a] University of Erlangen-Nuremberg, Department of Materials Science, Glass and Ceramics, 91058 Erlangen, Germany
[b] Brandenburg University of Technology, Department of Joining and Welding Technology, 03046 Cottbus, Germany
[c] Forschungszentrum Jülich GmbH, Institute of Energy and Climate Research, IEK-2, 52425 Jülich, Germany

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ABSTRACT

Ceramic multilayer structures based on tape cast alumina and zirconia substrates have been manufactured for use as carbon-free refractory materials. The laminates were reinforced via residual stresses due to shrinkage mismatch or differences in thermal expansion in order to achieve an improved thermal cycling capability. Thermal shock tests have been carried out using a plasma test stand. The impact of layer sequence and residual stresses has been demonstrated via measurement of Young's modulus and microstructure images of the layered structures. Hasselman parameters as well as the crack propagation behaviour at interfaces have been analysed via wedge splitting test.

1. Introduction

For decades the ceramic multilayer technology has been an emerging field in the area of functional ceramics, as it offers the possibility to combine layers of beneficial properties generating advanced structures with improved properties [1–4]. These layers which are the base product to build up multilayer structures are typically manufactured via tape casting [5–7]. In contrast, the refractories industry typically uses bricks, which are uniaxially or isostatically pressed. In recent research, there have also been attempts to apply multi-layered structures as planar [8,9] and also rotationally symmetric refractory structures [10–12]. A market analysis, which has been conducted in order to create a roadmap for the refractories industry, predicts potential for such multilayer composites [13].

The most important properties of refractories are corrosion resistance and thermal cycling capability. If the refractory is additionally subjected to mechanical load at higher temperatures for longer periods, plastic deformation via creep also becomes an important property, but for the intended applications in steel casting (e.g. sliding gates), creep effects are less important. The corrosion behaviour of multi-layered refractory structures has been characterised earlier [14]. Depending mainly on composition, porosity and pore size distribution, these alumina, spinel and magnesia tapes exhibited a corrosion behaviour suitable for refractory applications. Similar tapes, but based on the material system calcium-aluminate, were subjected to preliminary thermal shock tests [15]. As a result of the conducted downwards shocks, the critical temperature difference combined with loss of strength could be detected at a temperature difference of over 250 K, which is also suitable for steel casting applications.

During the intended application in steel casting thermal shock is applied only from one side, which causes a temperature gradient through the entire multilayer laminate. Therefore, methods are required, which apply a one-sided thermal shock. A suitable technique is the generation of a high temperature gradient via lamp or laser irradiation [16] or via plasma torch [17]; the latter was applied in this study. In contrast to the established techniques of water and air quenching, this test allows to evaluate the behaviour of a refractory close to its real stress situation during steel casting.

In order to evaluate the thermal shock behaviour quantitatively, Hasselman introduced the so-called R-parameters [18]. They describe the influence of heat conductivity, Young's modulus and other parameters on the damage behaviour in consequence of a thermal shock. The rapid temperature change causes a thermal stress gradient inside a sample due to the limited heat conductivity. This stress gradient may cause cracking, if the strength

* Corresponding author.

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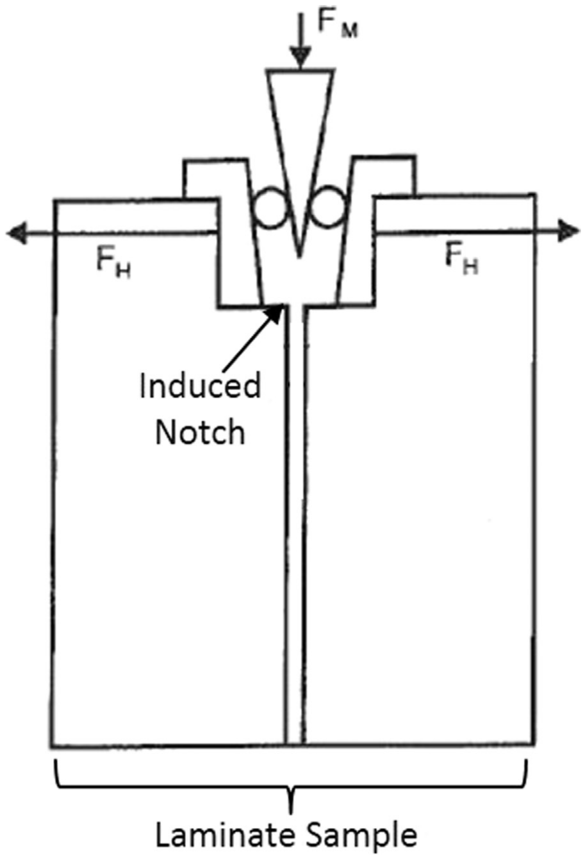

Fig. 1. WST set up schematically [21].

limit of the material which is subjected to a thermal shock is locally exceeded. The most common parameter used to describe the thermal shock behaviour is R'''', which is given by Eq. (1), where γ_{WOF} is the work of fracture, E is the Young's modulus and σ_{max} represents the fracture strength of the material. A high value of R'''' represents a high resistivity against crack propagation.

$$R'''' = \gamma_{WOF} E / \sigma_{max}^2 \tag{1}$$

In order to describe stable crack propagation, the parameter R_{st} (Eq. (2)) is used, where α is the coefficient of thermal expansion.

$$R_{st} = \sqrt{\frac{\gamma_{WOF}}{\alpha^2 E}} \tag{2}$$

The fracture strength is typically determined via four-point or double-ring bending experiments, whereas Young's modulus can be measured non-destructively via ultrasonic impulse reflexion or acoustic methods. A common technique to determine the work of fracture is the wedge splitting test (WST). The WST was introduced by Tschegg [19,20] and has become widely used for refractory materials because it enables stable crack propagation even for large specimen dimensions [21–23]. Fig. 1 illustrates the experimental set-up schematically. A vertical load is applied via a wedge and transformed into horizontal load by a loading transmission device. The influence of friction can be neglected and the horizontal force F_H can be calculated from the vertical load F_V and the angle of the wedge α:

$$F_H = \frac{F_V}{2\tan\left(\frac{\alpha}{2}\right)} \tag{3}$$

The work of fracture can be calculated from the load displacement data if crack propagation is stable during the whole splitting process:

$$\gamma_{WOF} = \frac{1}{2A} \int_0^{\delta_{H,max}} F_H \, d\delta_H \tag{4}$$

where δ is the displacement and A the ligament area; A is multiplied by 2 since two new surfaces are generated.

As the industry demands low-carbon steels in order to ensure good properties and a high ductility of the steels [24,25], the present study forgoes carbon, which is commonly a component in refractories due to its positive effect on the thermal shock behaviour [26]. In order to compensate for this disadvantage, residual stresses were incorporated into the multilayer structure to improve the thermal shock resistance, as the internal stresses counteract a thermal stress gradient in consequence of a thermal shock [27]. Therefore, the multilayer devices were designed in such a way, that the outermost layer is put under compression in order to avoid defects during firing. One way for the generation of residual stresses is the use of layers consisting of materials, which differ in their coefficients of thermal expansion (CTE). After the sintering process during cooling, layers with a higher CTE are put under tension whereas layers exhibiting a lower thermal expansion are set under compression. These stresses are temperature dependent; they will be reduced if the temperature is increased. A second method to generate residual stresses in multilayer structures is constrained sintering, based on the use of tapes exhibiting shrinkage differences. During sintering of such a multilayer system, a high-shrinkage layer will exhibit tensile stresses whereas a low-shrinkage layer will be put under compression. These stresses are permanent and temperature-independent. The shrinkage differences have to be moderate in order to avoid interface delamination or crack formation by stresses which exceed the strength level of the individual layers. An additional possibility to generate residual stresses is based on induced volume changes in the microstructure, e.g. via transformation toughening. A typical material system for this technique is alumina with additions of non-stabilized zirconia [28,29].

In the present study, tape cast alumina and zirconia layers will be used in order to design multilayer composites with residual stresses based on differences in thermal expansion and shrinkage, respectively. Different thermal shocking techniques will be performed to investigate the thermal shock behaviour of such structures. The wedge splitting test will be carried out in order to determine the thermal shock parameters R'''' and R_{st}. Measurements of the Young's modulus as well as microstructure images will be used additionally to discuss the thermal shock behaviour.

2. Experimental procedure

2.1. Tape casting and tape characterization

For the preparation of tape casting slurries, fine- and coarse-grained alumina and yttria-stabilized zirconia powders have been used, which are listed in Table 1. The median-diameters d_{50} of these powders were determined by laser diffraction (Mastersizer 2000, Malvern Instruments Ltd., Malvern, UK). The specific surface area of the powders was measured via BET analysis (ASAP 2000, Micromeritics Instrument Corporation, Norcross, GA, USA) using nitrogen as analysis gas. The density of the powders was measured by He-pycnometry (AccuPyc II 1340, Micromeritics Instrument Corporation, Norcross, GA, USA). All measured properties are listed in Table 2.

All cast tapes were multimodal tapes, i.e. they consisted of two or more powder fractions. Tapes A1 and A2 (Table 3) are bimodal alumina tapes based on a fine and one coarse grain fraction, which differ in their amount. Tapes A3 and A4 are multimodal alumina