Simulation-based Assessment of Refractory Materials

S. Pirkelmann, D. Schweiger, H. Friedrich, G. Seifert

The current trend towards improved environmental and economic efficiency of thermal processing requires the rapid development of improved refractory materials. Detailed characterization of materials through the application of advanced experimental and simulation methods and software tools from the broad field of machine learning can contribute to achieving this goal. The development of future refractory materials with increased performance can benefit significantly from such digital techniques. In this paper, a pertinent approach is presented to understand in detail the effects of the material structure of a refractory (pores and solid inclusions), with the aim of identifying the most critical elements with regard to component reliability. The technique is based on non-destructive analysis using 3D computed tomography, a neural network to automatically identify the structural features and finite element simulations to assess the impact of external thermal and mechanical loads.

1 Introduction

Refractory materials are facing a number of challenges owing to their application at harsh conditions in high temperature environments. While the individually most important performance requirements to a refractory material depend on its intended usage as, e.g., kiln furniture or as furnace linings with or without direct contact to hot solid or liquid material, several properties are almost always relevant. Typically, any refractory material must withstand mechanical and thermal loads, should be tolerant of high thermal stresses or thermal shocks, and have good resistance against corrosion, wear and creep. In addition, the increasing demand for low-carbon and energy-efficient production creates the need to develop refractories further towards minimal heat losses during use. This implies small heat capacity or low thermal conductivity, which stands in clear competition with classical requirements such as thermal shock resistance or mechanical strength. Above all, cost is always an issue.

Traditionally, these requirements are satisfied by experience-based composition of refractories from several raw material components, each of which contributes to certain aspects of the overall material. For example, pores in the structure improve thermal insulation, but are adversely affecting mechanical stability; larger grains of high-temperature stable phases (e. g., alumina) can stop crack propagation, thus increasing the fracture toughness and lifetime of the material, etc. As a consequence, in order to address the most important requirements, many refractory materials are strongly heterogeneous in terms of chemical as well as structural composition including pores at multiple length scales. In this situation, computation-assisted analysis of limitations of existing materials caused by local heterogeneities (pores or discrete grains) can be very helpful to improve the material. More generally, a detailed understanding of the contribution of each component to the overall properties of the refractory prospectively enables the development of improved materials tailored to specific applications by computational materials engineering.

In this paper, with respect to these goals, a novel combined approach for simulation-based analysis of refractory materials is presented based on the complex structure of the material. The method extracts a detailed virtual 3D model of the material structure from computed tomography (CT) images, taking into account the multiphase nature of the material. With this model, various finite element (FE) simulations can be performed for external loading cases replicating realistic conditions during component application. The focus will be on components in the range of some 100 µm like large pores or inclusions, which are prone to lower the material's fracture strength. The outcome of these simulations will serve to estimate the effects of these components on the fracture probability of refractories under different operating conditions. This, finally, can help guiding the conception of novel refractories with improved properties.

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The methodology is demonstrated using a common bauxite refractory material as an example.

2 Structural analysis

Basis for our analysis is a characterization of the refractory material's internal 3D structure. For this purpose, it is resorted to computed tomographic (CT) images of the material with a resolution of 33 µm. Spatial resolution and sample size were adapted to enable a reliable identification of all details relevant for fracture mechanical assessment on the one hand and to allow the detection of a representative sample volume on the other hand. An example of such an image recorded on a cylindrical sample of the refractory material is shown in Fig. 1a. The material exhibits a highly heterogeneous micro- and mesostructure, consisting essentially of three relevant main components, which are evident from the CT images: larger discrete grains (light areas), porosity (dark areas), and a fine-grained matrix in between. The latter has been studied in more detail by electron microscopy, providing evidence that, on the mesoscale considered in this work, it can be treated as a homogeneous phase (more details are given in chapter 3).

An automatized extraction of the three-dimensional structure of the constituents from a large number of originally high-resolution CT images is necessary to obtain sufficient statistical reliability. Freely available machine learning tools facilitate this task considerably. In this work, an advanced image segmentation method based on convolutional neural networks (CNN) was applied: the segmentation framework nnUNet [1]. In an offline training phase, the network is presented with examples of manually labeled images in which the corresponding structure type (discrete grains, pores, matrix) is annotated for each voxel in the image. Using this data, the network learns to independently create segmentations for new, unseen images. Depending on the number and quality of training images provided and the time spent training the network, the method can ultimately achieve a segmentation quality equal to that of human segmentation, with the annotation/classification process lasting a fraction of the time. Compared to classical threshold-based image segmentation algorithms, the approach based on CNNs enables reliable and fast segmentation of large sets of CT images without the need to manually set the threshold parameters for each image. In addition, the method is robust to variations in the input images and does not suffer from typical shortcomings of threshold segmentations such as misclassification due to not globally selectable thresholds, resolution-related smooth transitions at phase boundaries, and a variety of imaging artifacts.

The segmentation image in itself already provides valuable information about the composition of the material at hand, which can be used to detect certain obvious defects such as large pores. Fig. 1b shows the result of the segmentation generated by the nnUNet for the CT image of the refractory sample from Fig. 1a. The resulting segmentation image clearly distinguishes the three selected main components of the material. In addition, it is possible to extract additional data from the segmentation, which enable more sophisticated mechanical investigations, like statistical distributions of pores and grains, their size and shape distribution as well as their location within the structure.

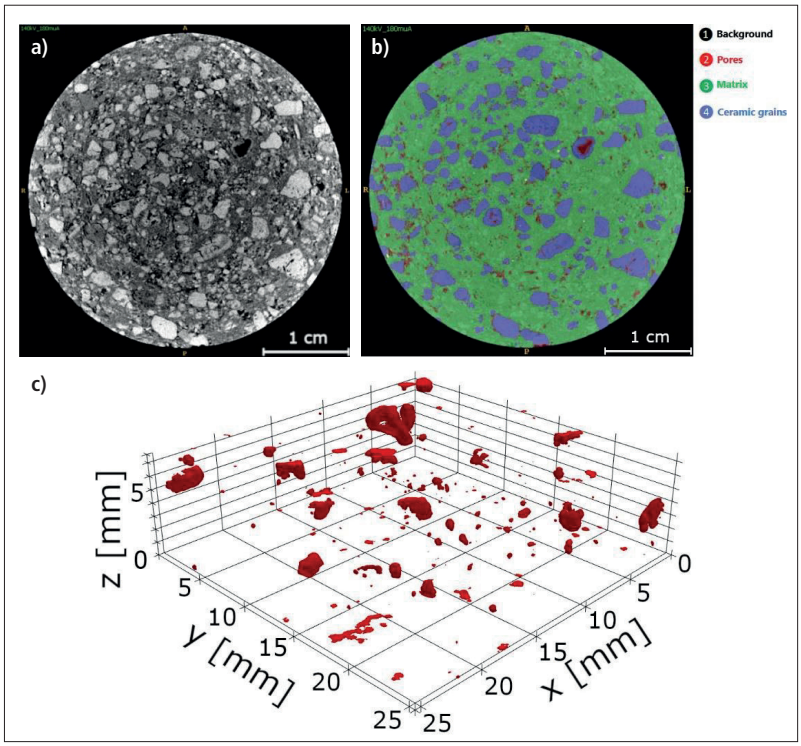

Fig. 1 a) Slice through a computed tomography (CT) scan of a sample of a refractory material. **b)** Segmentation of CT image into four distinct phases generated by a convolutional neural network. **c)** 3D visualization of extracted pores of the sample within a volume of about 25 mm x 25 mm x 7 mm.

3 Creation of 3D structure model

Once the segmentation of a sufficiently large volume of a refractory is finished, the automatically labelled 3D data from CT analysis are converted into three-dimensional models for further finite element (FE) analysis. Goal is to identify the most critical structural elements (pores or discrete grains) with respect to component failure under external loads. The expected benefits are guidelines on which kinds or which geometric characteristics of structural elements (considering size, shape and material) have to be avoided to improve performance and lifetime of a refractory material. FE simulations nowadays enable obtaining this information without the need for costly experiments.

In this paper, the focus in on the effects of large pores. Therefore, a selection of cubic volume elements containing pores with a typical size of 1 mm or larger is extracted from the labeled 3D voxel data. The size was chosen in accordance with the critical crack length estimated from fracture mechanical characterization of the refractory material. From these extracted volume elements, fi-

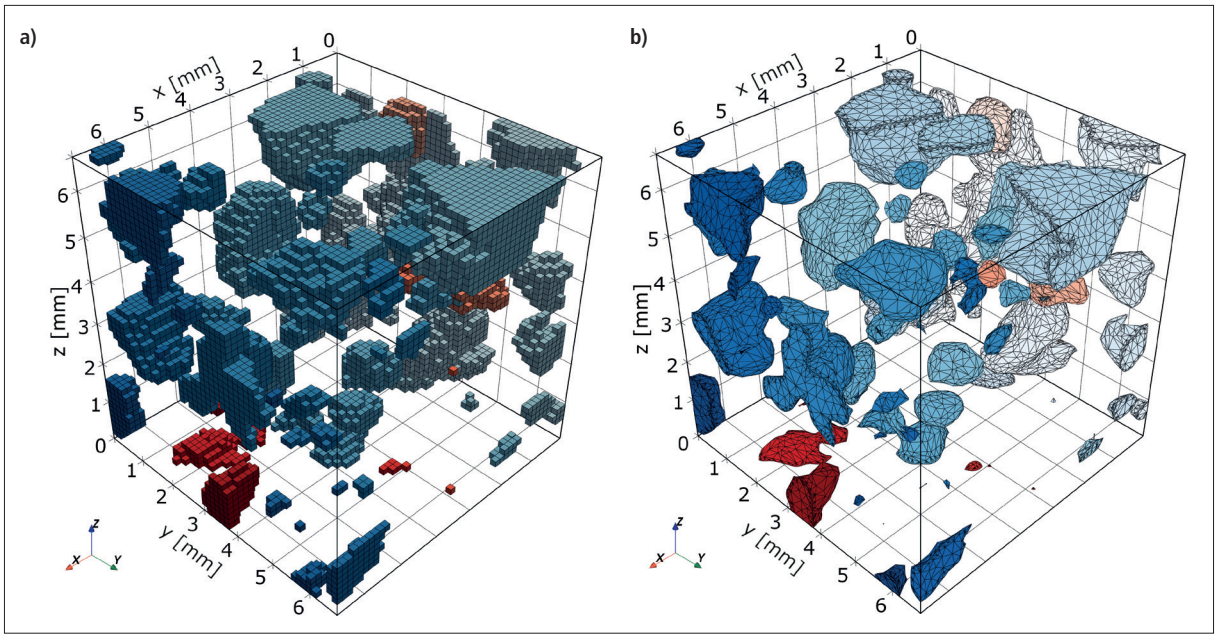

Fig. 2 a) 3D volume element of 50 x 50 x 50 voxels showing discrete grains (blue) and pores (red) extracted from the segmentation of the refractory material. **b)** Surface mesh generated from the voxel structure for use in FE simulations.

nite element meshes were generated using an in-house toolchain for microstructure simulations [2].

Fig. 2 shows, for one exemplary volume element, three-dimensional representations of the extracted voxel structure (Fig. 2a) and the FE mesh generated from it (Fig. 2b). In both graphs distinct ceramic grains are represented in blue, whereas pores are shown in red. The third component, the matrix phase filling the remaining space completely, is hidden for better visualization.

After assigning appropriate material properties to the different phases, FE simulations of specific loading cases can be performed, representing for instance large temperature gradients combined with weight-induced mechanical loads. For a general assessment of the effects of the mesostructure on fracture probability, as intended here, an analysis of tensile stresses around the pores is the most straightforward approach for any ceramic material. Below, the results of simulations under uniaxial tensile stress will be shown and evaluated with respect to stress concentration in the vicinity of the pores and the expected consequences for material strength.

3.1 Material properties

Bricks of a standard, commercially available Bauxite material type B80 have been used to demonstrate the method described above. In addition to the mesoscale analysis by computed tomography (CT) shown above, the microstructure including the chemical composition as well as the respective crystalline phases of the material have been characterized by scanning electron microscopy (SEM) including energy dispersive X-Ray (EDX) spectroscopy, X-ray diffraction (XRD) and X-ray fluorescence spectroscopy (XRF), not detailed here. On a length scale of 10 µm and below, the two solid phases assigned by the segmentation algorithm, i.e., discrete grains and bonding phase, exhibit a substructure and multiphase composition. The grains, represented in blue in Fig. 2, consist of 96,5 %-vol Al_2O_3, 2,0 %-vol Al_2TiO_5 and 1,5 %-vol microporosity. The matrix phase (hidden in Fig. 2) is composed of 25 %-vol Al_2O_3, 45 %-vol mullite and 30 %-vol porosity. The total porosity of the material (microporosity and mesoscale pores) is about 21 %. This complex composition of the material is represented in the FE simulation by using established mixing rules. The following approximate values for the constituents are used: Young's moduli of the matrix phase and the grains were 95 GPa and 320 GPa, respectively. The lower value of the matrix phase mainly reflects its considerable microporosity. For Poisson's ratio a value of 0,22 was used for both phases. For fracture toughness and bending strength of the B80 material, values of K_{Ic} = 1 MPa $\sqrt{m}$ and σ_c = 15 MPa were used. These values were estimated from own experiments, and are compatible with published, experimental data [3, 4].

3.2 FE Simulations

To study the effect of pores on the material strength, selected volume elements have been subjected to finite element analyses using the commercial software ANSYS. The basic idea is to assess the stress concentration around the pores in terms of its relevance for the component strength. This was done by performing the following steps of FE simulation:

- For each of the principal axes separately, a constant tensile stress σ_{ext} was applied on two opposite external faces of the volume element.
- A single step of a static structural FE simulation was carried out to obtain the resulting stress concentration within the material.
- The stress concentration in the surroundings of each individual pore was evaluated: regions were selected, where the first principal stress in the fine-grained ceramic matrix phase is increased by more than 10 % compared to the externally applied tensile stress σ_{ext}. The mean stress σ_{incr} in the identified volume is evaluated and a factor of local stress increase due to the presence of the pore is computed by

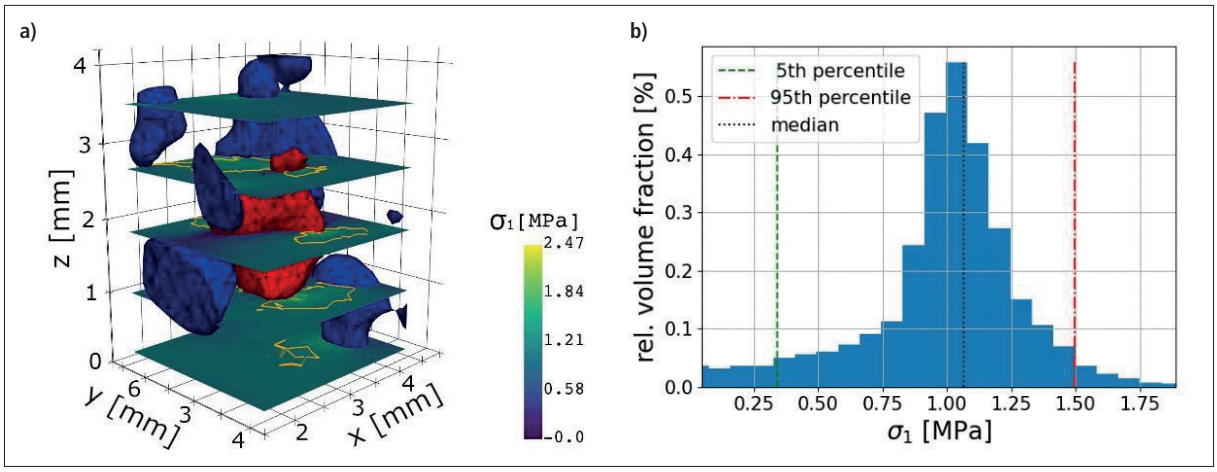

Fig. 3 a) Local principal stresses computed by the FE method in the vicinity of a pore (red) surrounded by several grains (blue) for a load of 1 MPa applied along the x-axis. The results show regions of locally increased stress (outlined in orange) in a halo around the pore orthogonal to the load direction. **b)** Distribution of principal stress inside a small region around the pore (weighted by relative element volume with respect to the whole volume element).

$$f_{incr} = \frac{\sigma_{incr}}{\sigma_{ext}}$$

- Using this factor, a local critical crack length in the vicinity of the pore can be estimated by

$$a_c^{loc} = \frac{1}{\pi}\left(\frac{K_{Ic}}{\sigma_c\, f_{incr}}\right)^2$$

- Finally, the identified volume V_incr of the region of increased stress was quantified and an equivalent diameter was calculated by

$$d = 2\sqrt[3]{\frac{3V_{incr}}{4\pi}}$$

- This quantity is compared to the local critical crack length a_c^{loc} from above to judge whether the current pore can be critical for the material's fracture strength.

The above procedure is illustrated for the refractory material by means of an exemplary chosen volume element of 6,7 mm × 6,7 mm ×6,7 mm and one large pore. An external stress σ_{ext} = 1 MPa is applied. Fig. 3a) shows multiple slices for a tensile stress applied along the x-axis. The resulting stress field leads to stress concentration by a factor f_{incr} = 1,22. Applying this value to the external fracture stress σ_c as specified in chapter 3.1 yields an increased local stress which, together with the fracture toughness, can be used to estimate a local critical crack length of a^c_{loc}=0,95 mm, which exceeds the size of the disturbed region. So the pore is considered relevant for the strength of the material. For a quantitative conclusion, a significant number of similar pores are evaluated and used for a statistical analysis. Details will be described in a forthcoming paper.

4 Conclusions and outlook

The methodology briefly described above is part of a large ongoing study with the goal to establish a quantitative relation between structure and service behavior of refractories [5]. Using FE simulations on the furnace and component scale, local thermal and mechanical loads during operation in thermal processes are extracted. On the other hand, the effect of structure on material properties of refractories is calculated by FE simulations on the micro- and mesoscale. Such material properties are elastic moduli, thermal conductivity, coefficient of thermal expansion and specific heat. Due to the complex structure of refractories, usually multiscale approaches are required, where the effective properties of constituents on the mesoscale are obtained from simulations on the microscale, before the macroscopic material properties are obtained in a second step.

Strength and fracture toughness are complex properties. Strength relies on both: the mesostructure of the refractory and defects introduced during production at the surface and within the interior of the components. Therefore, refractory components have to be analyzed by experimental methods to obtain the relevant structure. Computed tomography was proven to be an efficient tool to measure the structure at an adequate resolution and sample volume. However, it turned out that it has to be supplemented by image analysis methods to extract enough data for statistical analyses. This is done in a fully automatized procedure where a neural network identifies all relevant elements of interest within a sample, and the successive simulation chain determines the material properties. The transfer of the analyzed structures to FE models enables the application of various loads and the assessment of local effects like stress concentrations.

Currently this assessment is based on measured values of fracture toughness. The effect of pore shape and arrangement on crack initiation has to be further investigated. In some simplified models, it was discussed that single pores might stop propagating cracks and so strengthen the material [6], while only pores in close vicinity have a weakening effect [7]. On the other hand, experimental results have shown that larger pores decrease the material strength considerably more than smaller ones at the same degree of porosity [8]. Due to these ambiguities, simulated properties are compared to measured data for strength and reliability of the refractory material within our study. In addition, high temperature properties limiting lifetime like creep and resistance to thermal cycling are measured by special thermooptical measuring devices,

which have been developed recently for that purpose [9].

Altogether, the expected correlation between material structure on one hand, and its thermal and mechanical properties on the other hand, will enable a tailored optimization of refractory materials.

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