Integrated Computational Ceramics Engineering – an Approach to Radically Reduce Time-to-Market

F. Raether, G. Seifert

Integrated Computational Materials Engineering (ICME) combines various simulation tools on different scales to identify adequate composition, structure and process parameters of materials according to the performance requirements of new components. It includes inductive methods of data analysis and pattern recognition. The benefit of ICME is a drastic reduction of development cost and time which meets the needs of highly dynamic markets in a rapidly changing world.

Introduction

The demand for ICME was realised already in the last decade of the 20th century, where G. B. Olson published a number of papers on computational design of hierarchically structured materials [1]. He pointed out that the links between (i) processing and structure, (ii) structure and properties, and (iii) properties and performance are crucial for a systematic design of new materials (Fig. 1). Note that the term "structure" is used in a broad sense including composition. In USA, ICME was rated as a topic of national strategic importance and even as a matter of national security [2]. Consequently substantial funding was allocated. E.g., a Materials Genome Initiative (MGI) was founded in 2011, which coordinates the activities of all relevant governmental and funding organisations in USA [3].

Goal is to provide a R&D infrastructure which allows to discover, to manufacture and to deploy advanced materials at least twice as fast and at much lower cost than today. The scope is a bit wider than with initial ICME since the MGI combines computational tools with advanced experimental tools and digital data handling.

While the focus is on computational methods, experimental tools are intended to fill the gaps until all links (compare Fig. 1) are established. Experiments are considered essential for model validation as well. High throughput screening is considered as a valuable tool for fast data generation. Digital data handling is mainly a topic of data exchange between different research groups addressing data bases, standardisation of data formats and the qualification of data. A comprehensive review on ICME was presented by J. H. Panchal et al. in 2013 [4]. They point out that the top-down approach, where materials composition, structure and processing parameters are derived from the product requirements, is still very ambitious and needs much more research.

Also the relevance of structure is usually underestimated and not sufficiently taken into account in current databases. Therefore, a Microstructure Sensitive Design (MSD) was proposed. Panchal et al. also emphasize the importance of adequate handling of experimental and modeling uncertainties.

A proper balance of the trade-off between higher uncertainty on one hand and higher cost on the other hand requires better knowledge on error propagation and their impact on decision making within the development chain. Verification (the process of ensuring that the model meets the requirements) and validation (the proof that the model works in the right way) are core components of ICME which are still underdeveloped. Model experiments are also considered as a valuable alternative if reliable ab initio simulations are not available [4].

In Europe, there were relatively little activities on ICME. One of them was the ICMEg

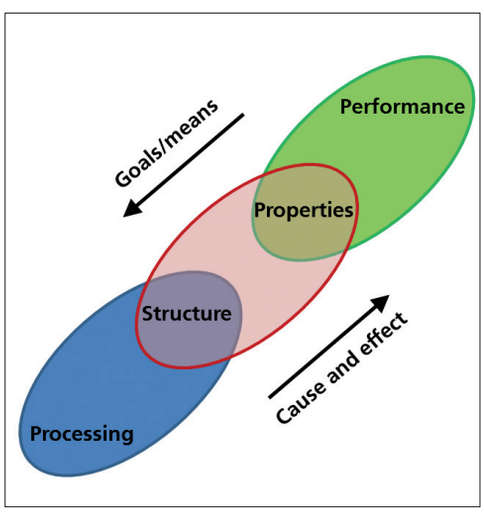

Fig. 1 Three-link chain model according to [1] (links are indicated by coloured ellipses)

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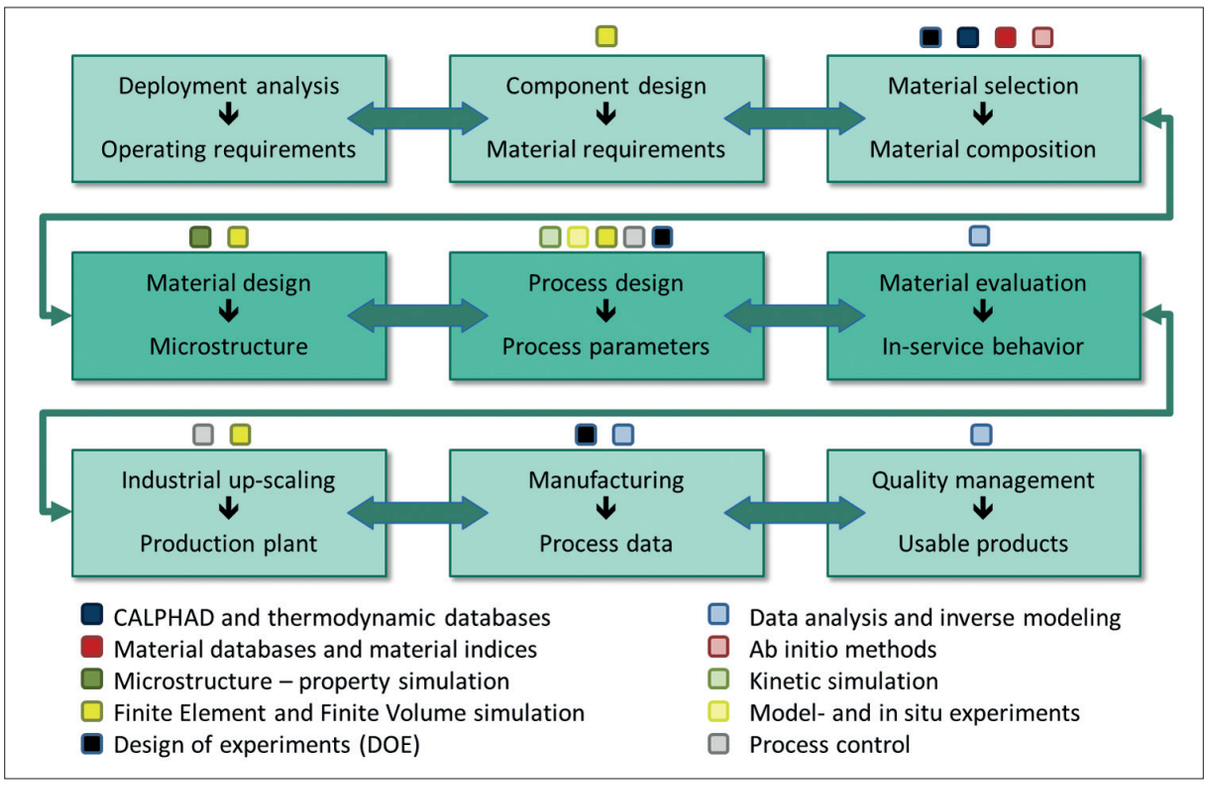

Fig. 2 Development chain for new ceramic products and building blocks for ICCE (building blocks are marked by coloured squares)

project which was funded by the EU within the 7th Framework Program [5]. Most success stories of ICME were on metals where sophisticated computational tools led to several new alloys [2].

Growing interest in ICME is reflected by an increasing number of annual publications. 400 publications focusing on ICME were published in 2017; references to ICME can be found in 4000 papers. However, there are nearly no ICME activities in the development of novel ceramics. Insofar, the title of the present contribution is still more an opportunity than a description of established tools. In the following, the status quo of integrated computational engineering of novel ceramics is outlined and recent examples are presented.

ICME in ceramics engineering

In general, the market volume of new ceramics is much smaller than that of new alloys. Accordingly, development costs have to be lower, and less data are available. This restricts buildings blocks for ICCE (Integrated Computational Ceramics Engineering) to more practical tools close to application. Fig. 2 shows the different steps from first planning to serial production for new ceramic products and attributes available building blocks to the individual steps within the development chain. These building blocks are either computational or experimental methods.

Finite Elemente (FE) and Finite Volume (FV) methods are well established in component design and up-scaling. Moreover, they are used in microstructure – property simulations and process design as will be illustrated in the next chapters. Material selection is a very important step in top-down development of new ceramic components. Benchmarking of existing materials is done using reliable material data bases and special material indices [6]. The stability of phases and concurrent reactions can be determined using thermodynamic databases.

Often, thermodynamic data on ceramic phases are missing. This gap can be filled by CALPHAD (CALculation of PHAse Diagrams) methods which are well established for decades; they have been occasionally used for the design of new ceramic phases, e.g. in the system Si–B–N–C [7]. Ab initio methods are rarely used in ceramics development, but it is expected that their importance will increase especially for functional ceramics and the identification of dopants.

Microstructure – property simulations are particularly important in multiphase ceramics with high material contrast. E.g., pores or grain boundary interphases can considerably affect thermal, mechanical and electrical behaviour of technical ceramics.

Commercial tools for microstructure – property simulations are available, but they often fail in the representation of special ceramic microstructures [8], since they are developed for use in metal and polymer science. Kinetic simulations are occasionally applied in different steps of ceramic process development for forming, drying, debinding and sintering [9].

Among the experimental methods, high throughput screening is not of practical importance in ceramics development. Investment costs are rather high and sample size cannot be reduced to the submillimetre range, since surface effects impair the testing results. Yet, other experimental tools are essential. DOE (Design of Experiments) can be used in different steps of the development to determine heuristically main and interaction effects of parameters and to identify optima with reduced experimental effort. DOE tools are widespread and accepted in ceramics development, but can

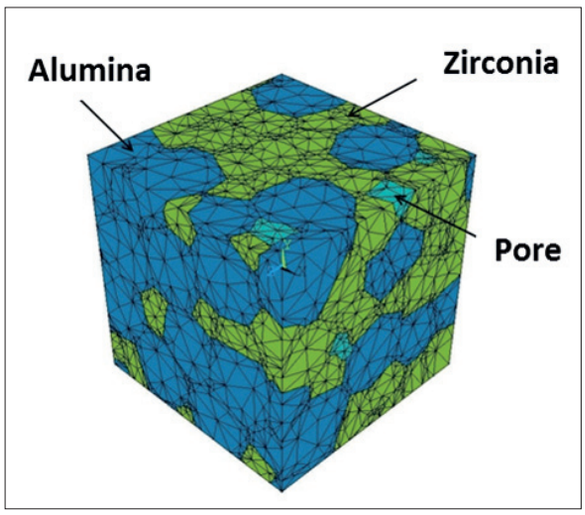

Fig. 3 Representative Volume Element (RVE) for microstructure – property simulation of ZTA ceramics after meshing

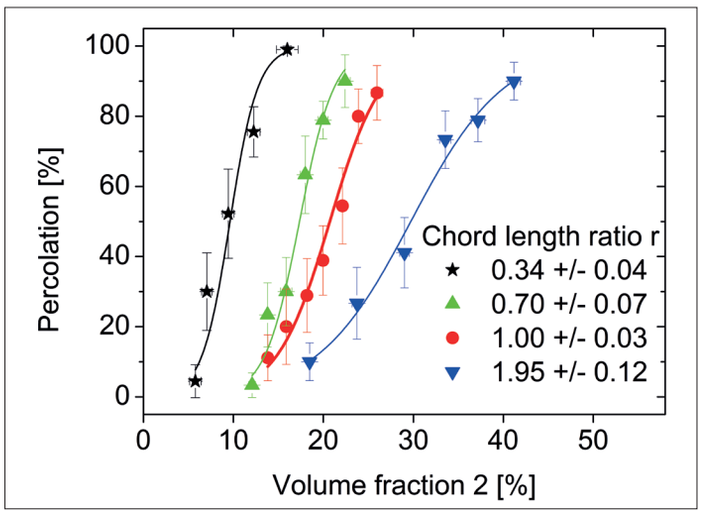

Fig. 4 Effect of particle size – represented by chord length ratio between phase 2 and phase 1 – on the percolation of phase 2 in a random arrangement of particles of both phases (compare [11])

as well be used during serial production to achieve a continuous improvement of processes. Process control is an important tool to guarantee the implementation of ideal processes in practical use. It needs more attention; especially high-performance and robust sensors for industrial high temperature processes are missing. On the other hand in situ measuring methods

are available which provide input data for computational process optimisation using lab experiments [10]. Like ICME in general, model experiments are often undervalued in ceramics development especially with respect to their capability in the validation of computer simulations.

Each of the building blocks shown in Fig. 2 is valuable by itself in order to reduce development cost and time. But the adaptation of these building blocks towards a smooth interaction within an integrated development system creates a considerable added value.

To illustrate the concept of Integrated Computational Ceramics Engineering and to make the interaction between the different building blocks of the engineering chain comprehensible, some simulation tools developed at the Fraunhofer Center for High Temperature Materials and Design HTL are briefly overviewed. Examples will be presented in the following three sections according to the three-link scheme of Olson (compare Fig. 1), which is still the base for ICME. Different from previous publications especially the integration which is achieved by these tools is pointed out.

Structure – properties

Material design can strongly benefit from models which are able to predict macroscopic material properties in dependence of the structural parameters, e.g.: phases, porosity, grain size and shape.

At Fraunhofer Center HTL, such structure-property simulations are done on the basis of a 3D-ceramic structure generator [11]. The in-house software is particularly designed for creating the special geometries of all kinds of ceramic microstructures, which are realised in form of small Representative Volume Elements (RVE) with periodic boundary conditions.

The latter strongly increases the representative character of the simulations and reduces computer time. Starting from RVEs containing typically 50–200 ceramic particles in a representation by 64^3 or 128^3 voxels respectively, structure generation considers features of the real processes like wet forming, sedimentation and agglomeration as well as sintering.

By that, the later identification of process parameters for the production of selected structures shall be simplified. The structure generator is combined with an in-house mesh generator to transform the voxel structure into a smooth FE mesh. Then an interface to commercial FE software provides rapid data transfer and automated calculations of macroscopic properties like thermal conductivity, elastic moduli, thermal expansion and stress concentration using the material properties of the involved phases as input [11].

Multiscale simulations are performed on different levels by initial calculation of material properties on the smaller scale and later use of these homogenised data on the larger scale [8]. Top-down modeling is achieved by setting predefined features, then generating in loops multiple structures until they match the target values. For that, the structure generator was combined with automatic

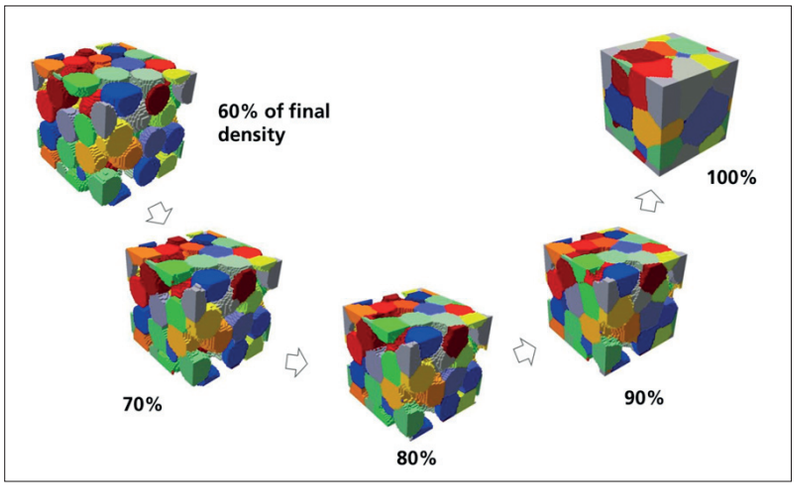

Fig. 5 Periodic RVE of a ceramic in different stages of solid stage sintering calculated by the integrated microstructural sintering model

tools for structure analysis in analogy to experimental methods – e.g. chord length analysis. The comparison of measured and simulated material properties of computer generated and experimental structures with identical structural features enable the validation of the computer models.

Some properties of ceramics are strongly affected by interface layers at the grain boundaries which are thinner than the particle diameters by about three orders of magnitude. This difference requires special methods to bridge scales. A recent example is the design of Zirconia Toughened Alumina (ZTA) ceramics [11] (Fig. 3).

Electrical conductivity at the grain boundaries was considered by introducing special thin interface layers during the meshing operation which were attributed to shell elements during FE simulation. By that, realistic simulations of electrical behaviour were obtained [11]. The requirements to the ZTA composite from the viewpoint of application are high mechanical stability combined with good electrical isolation. While higher zirconia content increases toughness, its electrical conductivity would discourage the use of the material as isolator as soon as percolation of the zirconia phase occurs.

Fig. 4 shows the impact of particle size on the percolation threshold which was calculated by the structure generator [11]. It was concluded that the particle size ratio between alumina and zirconia can be adapted to obtain sufficient zirconia content without percolation. By help of microstructure-property simulations covering mechanical, thermal and electrical material parameters, distinct design criteria have been identified. In the framework of ICCE, microstructure-property simulations establish a unique correlation between microstructural composition and macroscopic properties of ceramic materials, which drastically reduces the number of experiments required for the development of new ceramics.

Processing – structure

Knowing the favoured ceramic microstructure with respect to its desired functionality, the next challenge is to design the manufacturing process in a way that the optimal microstructure is safely reached. Sintering is the most important process step controlling structural development in ceramics. Currently three essential approaches exist to describe different aspects of microstructural development during sintering [12]:

- Ideal sintering which considers effects of interface energies;
- Kinetic Monte Carlo Simulation which considers the stochastic nature of sintering by diffusion processes;
- Discrete Element Modeling which considers interparticle forces and rearrangement during the sintering process.

Up to now, an interaction between the three models was not possible. Recently, at HTL an integrated microstructural sintering model was developed which includes the most important aspects of all three approaches [13]. Fig. 5 shows an example for the calculated structural changes during sintering.

The integrated model will be used to investigate principles of special time temperature – cycles like two-step sintering, pre-coarsening or rate controlled sintering. It can also help to understand microstructural development in processes like hot isostatic pressing and constrained sintering. While these simulations are a very useful tool to derive general guidelines how to design the processing conditions, they are not suited to model the behaviour of real ceramic components in detail, because too many simplifications are necessary in microstructure simulations.

For modeling sintering of real components, a continuum mechanical approach is required. The established way for this kind of simulation is to analyse the deformation of a sintering body, which is considered as a visco-plastic continuum, under a general state of stress during sintering [12]. The basis is a constitutive relationship between strain rates and stresses in the principal coordinate system (principal coordinate directions *i, j, k*). For an isotropic sintering body consisting of a linear viscous material, the relationship can be expressed by

$$\dot{\varepsilon}_i = \dot{\varepsilon}_f + \left(\frac{1}{E_{vis}}\right)\left[\sigma_i - \nu_{vis}(\sigma_j + \sigma_k)\right] \quad \text{(eq. 1)}$$

with free sintering strain rate $\dot{\varepsilon}_f$ external stress σ, uniaxial viscosity E_{vis} and viscous Poisson's ratio ν_{vis}. External stresses can result simply from gravity and friction between component and support material, or occur in cases of constrained sintering [12]. If the constitutive parameters E_{vis}, ν_{vis} and $\dot{\varepsilon}_f$ are known as a function of temperature and sintering state, a prediction of the densification, and shape distortion of a sintering body under a specific temperature-time cycle, with or without external load, can be calculated. Since all three parameters strongly depend on temperature heat transfer properties of the sintering body have to be known as well in order to calculate realistic temperature distributions. I.e., thermal diffusivity α and often emissivity ε and specific heat c_p are required as well.

The reliability of predictions calculated with the continuum sintering approach depends strongly on the quality of these input parameters. There are in situ measuring methods which deliver data on shrinkage kinetics, heat transfer and viscous properties during sintering [10].

However, these data have to be obtained at reasonable cost meaning that the param-

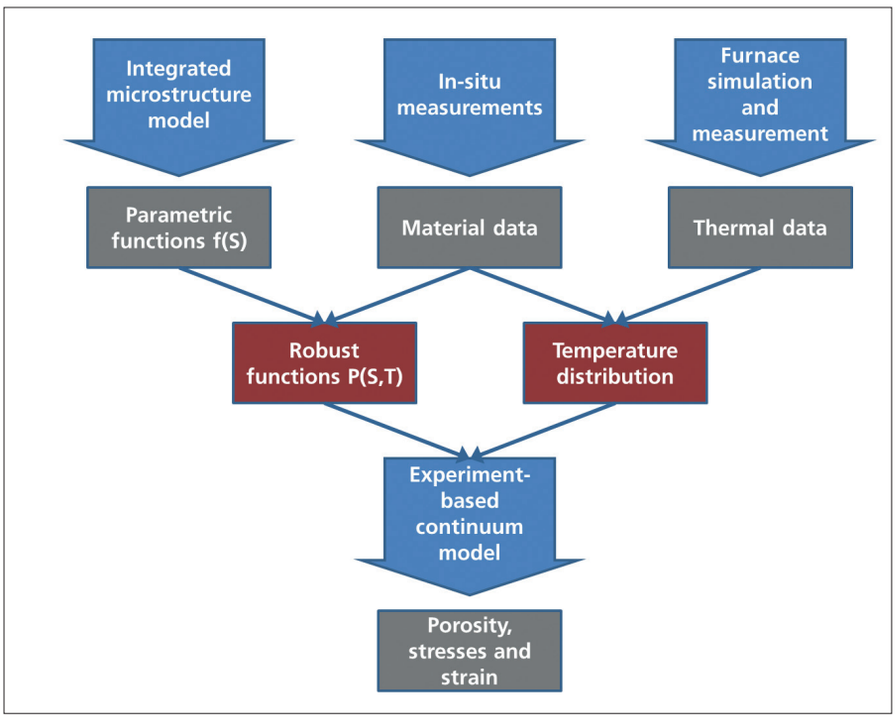

Fig. 6 Interaction of in situ measurements, microstructure and continuum models of sintering for a prediction of sintering distortions

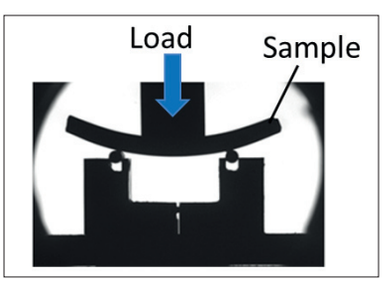

Fig. 7 Model experiment for the validation of the continuous sintering model using a bar shaped green sample of alumina loaded by a constant weight and measuring the deformation during sintering

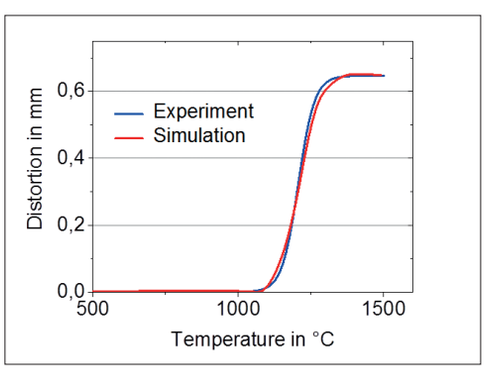

Fig. 8 Comparison of predicted and measured distortion of the alumina sample shown in Fig. 7

eter space can only be tested selectively. In addition, the measuring data have uncertainties. Both require a careful parameterization of the required properties in terms of sintering state *S* and temperature *T*. In general, variables *S* and *T* can be separated:

$$P(S,T) = f(S) \cdot g(T) \qquad \text{(eq. 2)}$$

In eq. 2, *P* denotes a property (E_{vis}, ν_{vis} and $\dot{\varepsilon}_f$ or α). The thermophysical change of the respective property is given by *g(T)*, its structural component by *f(S)*. While adequate functions *g(T)* are available, the functions *f(S)* are usually based on very old and simple sintering models. That is how the integrated microstructure model comes into play: it provides realistic parameterization functions for the input parameters in the continuum sintering model allowing for measuring at grid points and robust interpolation between them. Fig. 6 shows the interaction of models and experiments in the simulation of sintering processes.

In the resulting experiment-based continuum model of sintering processes, the constitutive eq. 1 is coupled with commercial FE solvers for mechanics and heat transfer. Predictions of the shrinkage and shape distortion of sintering components can be calculated for arbitrary temperature-time cycles within the limits of the measured parameter space. The model can be combined with furnace models, allowing design and optimisation of the thermal processing with respect to component quality and energy efficiency.

A model experiment was performed to validate this simulation technique: An alumina green body in the shape of a beam was put on two roller supports and has been loaded with a constant weight in the center of the bar (Fig. 7). The deformation of the beam during sintering was measured in situ by optical dilatometry.

Fig. 8 shows the measured vertical distortion in the center of the bar in comparison to a simulation using the experiment-based continuum model of sintering processes. A very good agreement between model experiment and computer simulation was obtained.

Properties – performance

The third link in the concept of ICME is to correlate the properties of a material with the functionality and performance of the final product. Due to the brittleness of ceramics, one of the most important performance indicators is its reliability, i.e., its safety against failure under operation.

The development time for achieving optimal reliability can be reduced by understanding the role of defects in crack initiation – depending on their shape, size and position. At Fraunhofer Center HTL, an integrated experimental and computational approach is used. Different experimental imaging techniques identify defects and characterise their shape: Computer Tomography (CT) for volume defects; CT, laser scanning or atomic force microscopy (depending on relevant length scale) for surface defects. Appropriate volume elements containing the defect region are selected and converted into FE meshes by in-house software, which are then used to analyse the stress concentration around the defect under different loading situations.

Qualitatively, these FE analyses allow identifying the most critical shapes and positions of defects with respect to crack initiation and component failure. Quantitatively, comparison of the stress distribution over the relevant finite elements enables assessment of the strength reduction due to different kinds of defects. Fig. 9 gives an

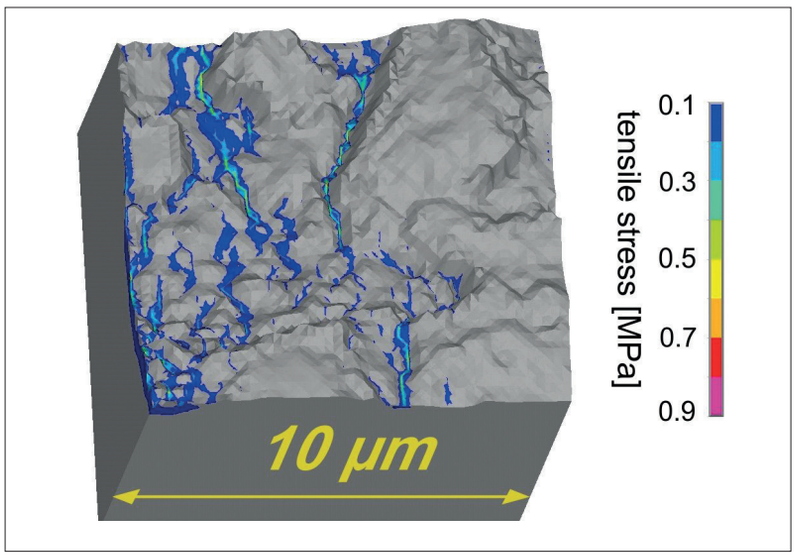

Fig. 9 **Representative volume element generated from experimental surface topography of used alumina sample; local stress under tensile load given by colour code**

example for a surface of an alumina part after heavy wear.

At the macroscopic level, the service life of ceramics under well-defined thermo-mechanical loads is estimated using a combination of Weibull statistics and FE modeling. Fracture tests at different temperatures and different loading rates provide the input data. For materials showing subcritical crack growth, fracture strength decreases towards lower loading rates [14].

Lifetime parameters for established material models can be extracted from the data, enabling a description of the decrease of strength as a function of applied stress, time and temperature. Using these models in an FE analysis allows us to calculate a specific failure probability for any special component and loading case.

Conclusions and outlook

ICCE is still at its very beginning. Although powerful building blocks are available (compare Fig. 2), the integration of methods is a challenge which will need at least one decade. However, the examples presented above show that some progress has already been achieved. Interaction of different models and experiments has to be balanced to combine the strengths of individual building blocks from a functional perspective.

ICCE has to be combined with other methods for the reduction of development time and cost like simultaneous engineering. Including many different fields of expertise, interdisciplinary cooperation and communication is a precondition for successful application of ICCE.

In general, the cost ratio between experiments and computer simulations will shift in favour of the latter. Decreasing computer time will enable many top-down developments (compare Fig. 1) simply by establishing iterative simulations which are controlled by fitting algorithms. Today, an open task is the efficient handling of either experimental or computational data. Since the acquisition of data is expensive, new models for data sharing are required. Besides standardisation of data formats, quality control is a big issue. Moreover, data sharing involves questions of copyright and new business models for selling data as well. The Fraunhofer Material Data space® is an approach to push this topic.

Opportunities for ICCE are new lightweight ceramic components – amongst others available now by Additive Manufacturing methods – and sustainable production routes required by climate change. Also the implementation of digital twins of production units would significantly contribute to a faster up-scaling of processes and a better process control during production – provided that the reliability of the digital twins is sufficient.

Acknowledgement

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