Analysis of the low temperature ceramics structure with consideration for polydispersity of initial refractory components

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Analysis of the Low Temperature Ceramics Structure with Consideration for Polydispersity of Initial Refractory Components

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Abstract. The results of computer simulation of the structure and physical properties of sintered low-temperature ceramics specimens with different volume fractions of different components of refractory components are presented. Properties of sintered ceramics, residual porosity, and shrinkage anisotropy are determined by features of packing of various fractions of refractory particles. The results indicate the determining factor of the presence of particles of the coarse fraction of refractory components capable of forming a internal skeleton of interacting particles.

INTRODUCTION

In a microporous sintered body, the heterogeneity of the particle size and density of their placement is considered as the determining factor of the behavior of disperse systems [1]. At the same time, there are microvolumes in the sintered body in which the local level of Laplace capillary pressure is markedly different from the average one, which means that the study of a sintering process [2], irrespective of the mechanism of substance flow, should be carried out with consideration for structural and geometrical factors.

The investigation of technological synthesis conditions of low-temperature ceramics formed during sintering of layers formed from mixtures of different material powders requires the development of computer-aided approaches to material designing based on the hierarchy of rheological processes in powder bodies and on modifications of hierarchically organized structures. At the same time, local characteristics of materials and processes are modeled on the base of the classical results. It is proposed to use the approach of physical mesomechanics, which unites the ideology and tools of the solid state physics and micromechanics of structured media [3].

Initial low-temperature ceramic compacts under additive technologies are formed of layers of the material. The heterogeneity of the particle size and density of their placement is a certain technological parameter of the layer formation process after the removal of the binder. The heterogeneity of joining of different layers and the formation of interlayer interfaces should be added to this heterogeneity. An adequate model of the original compact structure should reflect the overall heterogeneity of concentrations and sizes of powdered components and pores in initial layers and interlayer interfaces. From the standpoint of micromechanics media with the structure of heterogeneous materials can be associated with an effective medium with a periodic macrostructure under natural limitations of statistical averaging and the principle of translational symmetry [4].

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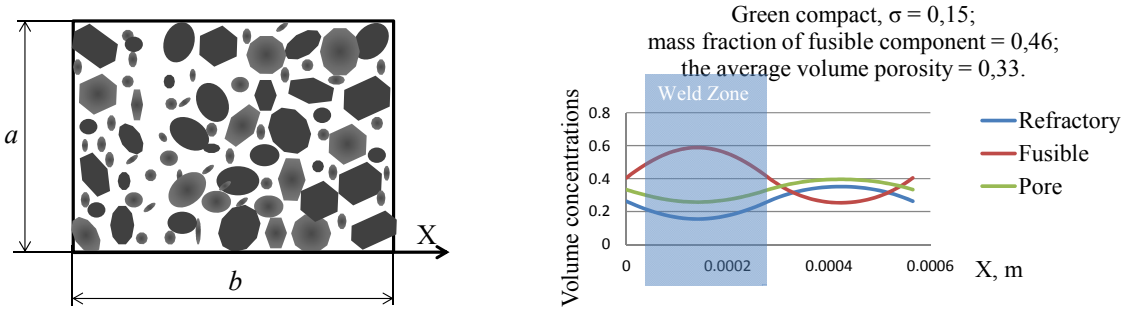

(a) (b)

FIGURE 1. Powder body before sintering

COMPUTATIONAL EXPERIMENT

The approach and adapted computer simulation scheme detailed previously [1] for mechanochemical reacting powder systems are used for a computational experiment. In microvolumes of a model body formed during discretization of the computational domain, all characteristics of the medium are considered to be effective in view of local concentrations and state parameters. In each time point, these characteristics are specified iteratively.

Figure 1a is an example of the structure of refractory components of the cross-section of a representative volume of the initial dispersion formed of individual layers. A representative volume of the original compact includes representative parts of individual layers and interlayer interfaces. The model structure of the original compact is submitted by the periodic system of selected volumes. A representative volume, which is shown in Fig. 1a, is characterized by a given cross-section $a \times a$ and length b corresponding to geometric properties of the original powder body.

In this model, local microvolumes of the sintered compact are represented by the $a \times a \times dx$ cross-section of the representative volume. Here a is the cross-sectional size of the representative volume and dx is the microlayer thickness, which is formed by division of the representative volume into equal N elements over the layer thickness during the implementation of a numerical scheme for computer simulation.

The standard deviation σ is convenient to consider as the degree of heterogeneity of component concentrations. For the considered model of a porous powder body, the one-dimensional distribution of volume component concentrations over the layer thickness are investigated: $C_{\text{refractory}}$, C_{fusible}, and C_{pore} are refractory and low-melting (fusible) components and pores, respectively, as it is shown in Fig. 1b.

Sintering of the powder compact is initiated by heating to the sintering temperature, which is usually lower than the melting temperature of the fusible component.

The model structure of the polydisperse mixture of refractory components can be represented by the particle size of individual fractions: d_{max}, d_{midi}, d_{mini}, d_{nano} and volume fraction of each component D_{max}, D_{midi}, D_{mini}, D_{nano}.

Three coarser and microheterogeneous fractions of refractory components of the initial powder vary in size $d_{\text{max}} > d_{\text{midi}} > d_{\text{mini}}$ and are classed as coarse particles due to their ability to form a power structure of interacting particles. In the initial mixture, ultramicroheterogeneous refractory components of the d_{nano} fraction actually join the dispersion medium, thus forming and modifying its characteristics.

The structure and properties of low-temperature ceramics can be compared by the example of compacts characterized by equal values of the standard deviation of volume concentrations of the components along the length of the representative volume, average pore volume, and other structural characteristics, except for the initial distribution of refractory components of different fractions. Three variants of original mixtures of refractory components are shown in Table 1.

Depending on the model discrete distribution (a), (b) or (c), the prediction of the possibility of formation of the skeleton of refractory particles at the macro-, meso- and microlevels determines the prediction of the minimum porosity of the sintered body in accordance with the condition of achievement of the mechanical contact of considered fraction particles. The possibility of the formation of the skeleton of refractory particles at any structural level decreases with a decrease in the local fraction of the considered refractory component (with an increase in the proportion of the fusible component).

TABLE 1

Model composition	Fraction size d_{max}, µm	Volume fraction D_{max}	Fraction size d_{midi}, µm	Volume fraction D_{midi}	Fraction size d_{mini}, µm	Volume fraction D_{mini}	Fraction size d_{nano}, µm	Volume fraction D_{nano}
(a)	16.0	0.88	4.0	0.03	1.0	0.02	0.01	0.01
(b)	16.0	0.03	4.0	0.88	1.0	0.02	0.01	0.01
(c)	16.0	0.02	4.0	0.03	1.0	0.88	0.01	0.01

As model compositions (a), (b), and (c), we consider initial powder compacts characterized by the mass fraction of the fusible component 0.46, the standard deviation of volume concentrations of the components 0.15, and the average relative pore volume (after the removal of the organic binder) 0.33. For these three cases, the concentration distribution of initial components over the length of the representative volume of the original mixture will be uniform, as shown in Fig. 1b. The prediction of the minimum porosity of sintered materials, which is defined by the difference between initial porosity and porosity necessary to achieve dense packing, is shown in Fig. 2a by lines P_{min} (a), P_{min} (b), and P_{min} (c). The prediction of the distribution of the true concentration of the components of low-temperature sintered ceramics is shown in Figs. 2b, 2c, and 2d for the three model compositions with different fractions of refractory components. An opportunity of anisotropic shrinkage caused by the requirement of the translation symmetry of the strain state of the representative volume to ensure the continuity of the model medium is taken into account.

As we can see from the simulation results of the structure of low-temperature sintered ceramics in Fig. 3, the possibility of the formation of the skeleton of refractory components located at different hierarchical levels of the structure makes a decisive contribution to the development of the pore structure. The contribution of the microheterogeneous fraction d_{mini} of refractory particles capable to form a sintered ceramic skeleton in the presence of residual porosity is most significant.

The use of a mixture composed of refractory ceramic components with the multimodal particle size distribution as the green compact allows us to obtain low-temperature low-porosity ceramics.

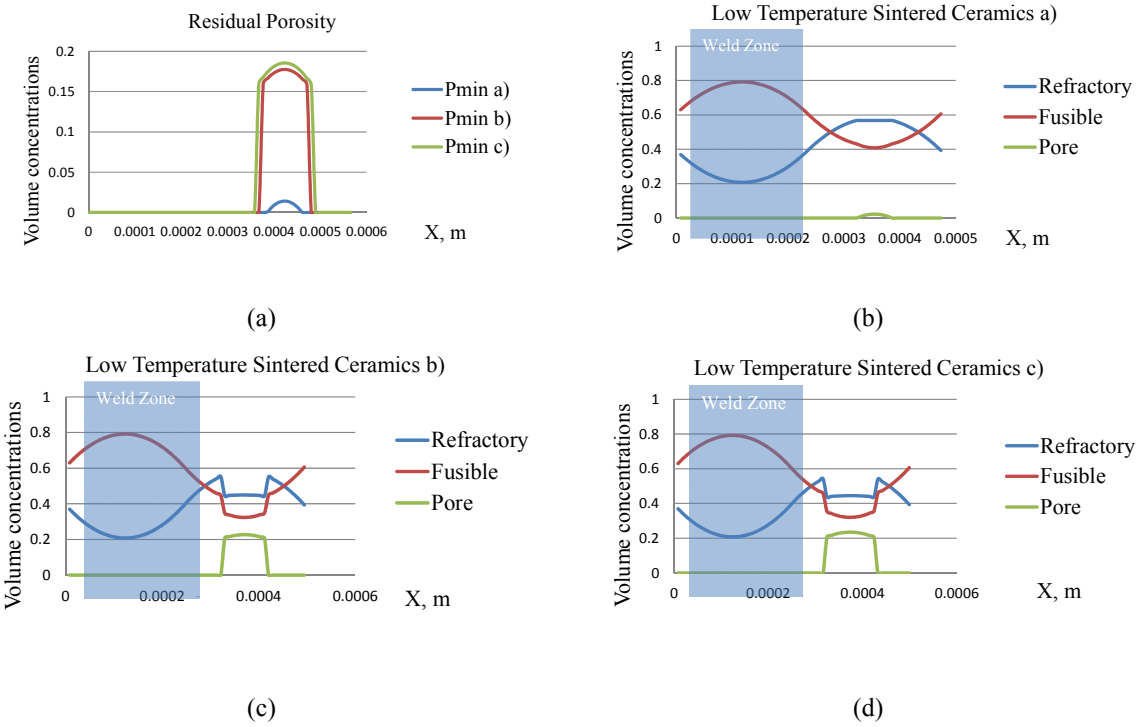

FIGURE 2. Structures of low-temperature sintered ceramics

TABLE 2

Parameters	Model composition (a)	Model composition (b)	Model composition (c)
Density, kg/m³	2929	2806	2779
Coefficient of thermal conductivity, W/(m×K)	1.45	1.234	1.171
Dielectric constant, F/m	6.814	6.117	5.969
Coefficient of thermal expansion, 1/deg	6.95×10^{-6}	6.11×10^{-6}	5.93×10^{-6}
Bending strength, MPa	167	159	157.6
Shrinkage of ceramics in the layer plane	0.10	0.10	0.10
Shrinkage of ceramics in the layer thickness	0.16	0.13	0.12

CALCULATION OF CHARACTERISTICS OF SPECIMENS OF LOW-TEMPERATURE CERAMICS

A theoretical estimate of physical and mechanical characteristics of experimental specimens of low-temperature ceramics using mathematical modeling is carried out by the prediction of the model structure of the sintered body. We consider α-Al_2O_3 as the refractory component of the initial mixture and borosilicate glass as the fusible component. Physical characteristics of the mixture components, which are known from the literature, are used in the computational experiment.

The results of numerical experiments of estimation of effective physical characteristics of low-temperature sintered ceramics are shown in Table 2.

CONCLUSION

The results of estimation of effective physical characteristics confirm an expected fact that the possibility of formation of the skeleton of refractory components located at different hierarchical levels of the structure makes a decisive contribution to the formation of low-temperature ceramics parameters. Moreover, the microheterogeneous fraction of refractory particles capable of forming a sintered ceramic skeleton makes the largest contribution.

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