Computer simulation of low-temperature composites sintering processes for additive technologies

A O Tovpinets, V N Leytsin and M A Dmitrieva
Immanuel Kant Baltic Federal University, Kaliningrad, 236016, Russia

E-mail: tovpinets_a@mail.ru

Abstract. This is impact research of mixture raw components characteristics on the low-temperature composites structure formation during the sintering process. The obtained results showed that the structure determination of initial compacts obtained after thermal destruction of the polymer binder lets quantify the concentrations of main components and the refractory crystalline product of thermal destruction. Accounting for the distribution of thermal destruction refractory product allows us to refine the forecast of thermal stresses in the matrix of sintered composite. The presented results can be considered as a basis for optimization of initial compositions of multilayer low-temperature composites obtained by additive technologies.

1. Introduction
A relevant task is to research technological methods of sintering of multilayered low-temperature ceramic. Additive manufacturing technologies using these materials are distinguished by layer-by-layer material growth and subsequent sintering. The heterogeneity of particle sizes and the density of their packing during the formation of initial compacts are technological parameters of layers packing. And the formation of interlayer interfaces should be taken into account. The under study model structure of the initial compact should reflect the heterogeneity of concentrations and sizes of original powder components and pores in the consolidated layers and interlayer interfaces.

The computer modeling approach is developed to study the determining factors of low-temperature composites structure formation with considering the hierarchy of rheological processes in powder bodies and modification of hierarchically organized structures. The physical mesomechanic approach was taken in this research. It is combining the ideology and tools of solid state physics and mediums micromechanic with a structure. [1]. Local processes and materials characteristics are simulated based on the classical results.

The initial compact preparation regimes, low-temperature ceramics sintering, and the effect of structural-mechanical characteristics on residual strains in the sintered material are examined.

The initial compact of low-temperature ceramics is formed layer by layer from a mixture of components with certain porosity. Material matrix is forming during the composite sintering by the low-melting mixture raw components in a temperature lower than its melting point. The effective characteristics of the sintered ceramics are determined by the macroscopic structure of the initial compact, components and pores distribution and concentrations inhomogeneity of the phase and fractional initial components composition.

2. Methods of modelling
The raw polydisperse mixture material of the initial compact refractory elements is presented by a set of particle sizes and volume fractions of each set in a mixture of refractory components. Coarse and microheterogeneous particles of the initial powder refractory components are different in sizes and referred as "large" particles because of ability to form the interacting particles power frame. Ultra-microheterogeneous refractory particles are attached to the dispersion phase forming and modifying its characteristics. The prediction of forming the refractory particles framework on macro-, meso-, or micro levels determines possibility to forecast the minimal sintered body porosity in accordance with the conditions for achieving mechanical contact of the fraction particles. It depends on the type of discrete distribution. The forming possibility of refractory components mixture frame at any structural level decreases with a local refractory components proportion decrease (and with increasing low-melting component proportion). The forming possibility of refractory components frame at any indenture level makes a key contribution to the pore structure of the sintered sample formation [2].

Gas-phase decomposition products and a crystalline residue are forming during the binder’s thermal destruction. The initial porosity of the green compact is increasing after removing the gas-phase.

The crystalline residue of thermal destruction is an extra refractory phase in the sintered samples matrix. This leads to the appearance of thermal residual stresses in the matrix caused by the difference in the thermal expansion coefficients of the disperse compositions and the matrix. The following expression can be used from the position of thermoelasticity to estimate the residual thermal stresses in the sintered ceramic matrix:

\[
\sigma_T = \int_{T_0}^{T_p} K_{glass}(T) \left[ \frac{\beta_{glass}(T) C_{glass} - \beta_{ceramics}(T) C_{ceramics}}{C_{glass} + C_{ceramics}} + \frac{\beta_{glass}(T) C_{glass} - \beta_{SiO_2}(T) C_{SiO_2}}{C_{glass} + C_{SiO_2}} \right] dT, \tag{1}
\]

where \(K_{glass}(T)\) is the temperature dependence of the elastic bulk modulus of glass; \(\beta_{glass}(T), \beta_{ceramics}(T), \beta_{SiO_2}(T)\) are the temperature coefficient dependences of thermal matrix and dispersed refractory components expansion; \(C_{glass}, C_{ceramics}, C_{SiO_2}\) are the volume concentrations of the matrix and dispersed refractory components; \(T_p\) is the temperature point of transition to an elastic stage, \(T_0\) is the initial temperature.

Calculation of residual thermal stresses should take into account the stage of viscoplastic stress relaxation when the sintered ceramics are cooled to laboratory temperature. In [3], it is proposed to introduce the temperature transition point of the material into the elastic state \(T_p\) in order to estimate the residual thermal stresses. The assumption is made of an abrupt transition to an elastic state, with allowance for which it can be assumed that in the temperature range from the sintering temperature to the temperature point of the transition, the temperature stresses relax to a negligible level. It is assumed that the transition to an elastic state stops the relaxation of residual stresses, and the evaluation of thermal stresses is possible from the position of thermoelasticity. For borosilicate glass we can use the temperature value \(T_p = 900\) K, indicated in Figure 1 as the point of transition to the elastic state.

The type of the residual stresses (tensile or compressive) is determined by numerator signs in integrand in the temperature range \(T_p - T_0\). It is assumed that for temperature values \(T \geq T_p\) the values of temperature stresses \(\sigma_T \approx 0\).

3. Results and discussion

Numeric evaluations were made using the component’s material characteristics in [4-7]. The sintered ceramic sample model structure forming conditions were examined. The initial compact is formed by three successively applied layers. Two compositions of the initial mixture, differing in the mass content of components are considered to study the factors determining thermal stresses in the matrix of sintered composite.
Figure 1. Temperature dependence of the viscosity of borosilicate glass.

Figure 2 shows the distributions of the volume concentrations of mixture components by the thickness of the model sintered sample at different mass compositions of the components in the initial compact:

- a) $\alpha\text{Al}_2\text{O}_3$ – 40 mass.%, borosilicate glass – 40 mass.%, polyorganosiloxane (the binder) – 20 mass.%,
- b) $\alpha\text{Al}_2\text{O}_3$ – 30 mass.%, borosilicate glass – 50 mass.%, polyorganosiloxane (the binder) – 20 mass.%.

The porosity and the standard deviation of the components volume concentrations in the initial compact are equal (=10%) in both cases.

Areas bounded by dashed lines represent the interlayer interfaces of the simulated layers. They are characterized by high content of refractory component $\alpha\text{Al}_2\text{O}_3$ and low content of sintered composite matrix which is borosilicate glass. A - B intervals are the external boundaries of the simulated periodicity cell, which is developing in each phase of additive initial compact preparing process. The model periodicity cell consolidates over layer and the interlayer interface.

Figure 2. Distribution of volume concentrations of sintered ceramics components by the thickness of the modeled layer.

The prediction of thermal stresses in the matrix of simulated samples is made using the relation (1) at the next step of computational experiments.
Figure 3 shows the residual thermal stress distributions for compositions (a) and (b) by the thickness of the model layer.

Figure 3. Distribution of residual thermal stresses in the matrix of sintered ceramics by the thickness of the modeled layer. Mass content of components in the initial compact: a) $\alpha$Al$_2$O$_3$ – 40 mass. %, borosilicate glass – 40 mass. %, polyorganosiloxane (the binder) – 20 mass. %, b) $\alpha$Al$_2$O$_3$ – 30 mass. %, borosilicate glass – 50 mass. %, polyorganosiloxane (the binder) – 20 mass. %.

The local tensile stresses are observed in interlayer interfaces (areas bounded by thin lines). The presence of significant tensile stresses exceeding the strength limit can cause defects and cracks in model composite matrix. As follows from the results shown in Figure 2, it is possible to change residual stresses ranges and to accomplish the strength criteria by varying the mass fraction balance. The tensile stresses for composition (b) in the interlayer interface are 2 times less than for the composition in (a). In addition, the significant local compressive thermal stresses in the ceramics sintered from composition (b) are predicted in the deposited (additive) layer, which prevent the development of defects.

4. Conclusion
The computer simulation of binder thermal destruction, which is the initial stage of composite sintering process, makes it possible to obtain a prediction of the sintered samples structure and residual thermal stresses.

Analysis of the results shows the possible appearance of both tensile and compressive quantities of residual stresses in the matrix of sintered ceramics. This can lead to destructive or hardening effects in the resulting products. Changing of value and residual stresses sign in the sintered material is possible by optimizing the concentration composition of initial compact components.

References
[1] Leitsin V N, Dmitrieva M A and Kolmakova T V 2008 Governing Factors of Physical and Chemical Behavior of Reactive Powder Materials Powder Metallurgy Research Trends (New York: Nova Science Publishers)
[2] Leitsin V N, Dmitrieva M A, Tovpinets A O, Ivonin I V and Ponomarev S V 2016 Analysis of the low temperature ceramics structure with consideration for polydispersity of initial refractory components Advanced materials with hierarchical structure for new technologies and reliable structures: Proceedings of the International Conference on Advanced Materials with Hierarchical Structure for New Technologies and Reliable Structures (Tomsk) pp 146-147
[3] Chernyshev G N, Popov A L, Kozintsev V M and Ponomarev I I 1996 Residual stresses in deformable solids *Nauka* (Moscow) p 240

[4] Imanaka Y 2005 Multilayered low temperature cofired ceramics (LTCC) technology *Springer science business media* p 205

[5] Saito K 1981 Use of Organic Materials for Ceramic-Modeling Process-Binder, Deflocculant, Plasticizer, Lubricant, Solvent, Protective Colloid *J. of Adhesion Society of Japan* 17 3 104-113

[6] Kamehara N, Imanaka Y, and Niwa K 1987 Multilayer Ceramic Circuit Board with Copper Conductor (Denshi Tokyo) 26 143-148

[7] Tummala R.R. et al. 1992 High performance glass-ceramicl copper multilayer substrate with thin-film redistribution *IBM J. Res. Develop.* 36 5 889-904