Acoustical and thermo physical properties of metal-ceramics composites in dependence on few volume concentration of metal

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Abstract Metal-ceramics composites (cermets) are modern construction material used in different industry branches. Their strength and heat resistance depend on elastic and thermos physical properties. In this work cermets based on corundum and stainless steel (sintered in high vacuum at temperatures 1500 – 1600°C) are investigated. The volume steel concentration in the samples varies up 2 to 20 vol %. The elastic modules were measured by ultrasonic method at room temperature, measuring of thermo conductivity coefficient were carried out at temperatures 100, 200°C by method of continued heating in adiabatic calorimeter. We founded appearance of two extremes on dependences of elastic modules (E, G) on stainless steel concentrations, nature of which is unknown, modules values change in range: E = 110 - 310, G = 60 - 130GPa (for different temperatures of sintering). Similar dependence is observed for thermo conductivity coefficient which values varies up 10 to 40 W/(m.K). There is presented also discussion of results based on structure cermet model as multiphase micro heterogeneous media with isotropic physical properties in the work.

1. Introduction
Composite materials on the base of ceramics and metal are known in different industry branches. These materials were created for industrial application to produce constructions possessing a high strength, thermal stability and resistance to aggressive media. Investigation results of physical technical properties of the materials are published in periodical issues and monographs [1, 2]. It should be noted that thermo-shock resistance composites have to possess high thermo conductivity and mechanical stability which are determined by elastic modules. The measurement of mechanical stability requires special preparation of many samples to provide reliable results, whereas for measurement of dynamic elastic modules one sample is required. Now there is no uniform understanding about connection of elastic and thermo physical properties for sintered dispersed materials. In this work we explore this connection for composite corundum-metal.

2. Experimental technique
We investigated samples of cermets based on α-Al₂O₃ in combination with the commercial quality stainless steel (18 - Cr, 9 - Ni, 1 - Ti, 72 wt% Fe). To fabricate a cermet, the initial fine grained mixture was prepared by milling α-Al₂O₃ powder in a ball mill in the presence of balls 1–2 cm in diameter made from the stainless steel. The milling was terminated when the steel content in the α-Al₂O₃ powder became equal to 2.2, 4.0, 5.5, 11.0, 21.0vol %. Then, the mixture obtained was doped
with a plasticizer and subjected to dry compaction under a pressure of 100MPa and then sintered in a vacuum at 1500, 1600°C. Finally, the samples were cooled in the furnace at an average rate 100°C/h and no further treatment was made. The cerments thus prepared ranged in volume porosity from 3 to 7% and had the steel content from 2.2 to 21.0 vol%. The cermet samples (two series for two temperatures of sintering) were then cut into smaller parts of the required dimensions (10 mm – diameter, 8-15 mm - length), which were further ground and polished, depending on the measuring method. To investigate the elastic properties of the cerments, we measured their density by the hydrostatic method and the velocity of longitudinal and transverse ultrasound waves of a frequency of 2MHz. We used the pulsed phase-interferometer method [3], which made it possible to determine the velocity of ultrasound within an accuracy of 0.1–0.2% and 5% for the elastic modules. With the data obtained, we calculated Young’s modulus E and the shear modulus G from the well-known formulas of the elasticity theory for an isotropic medium [3]. The thermo conductivity coefficient λ was measured by adiabatic calorimeter NT-λ-400 (Thermometer company). The measuring λ was carried out by method of comparison with the standard sample. The measurement accuracy of λ is 2-3% for all samples. Examination of the surface structure (cleaved facet) of the cermet samples was made with a JSM-840 scanning electron microscope (the Jeol company).

3. Results and discussion

In fig.1 typical microstructure of studied cerments obtained by scanning electron microscopy (SEM) are presented. It is seen there are three phases in samples researched: corundum grains, metal drops and pores. The metal drops with in size 2-3 μm are disposed usually on joint of corundum grains, more small are disposed on the grain faces. It should be noted that inter phase and inter grain boundaries are not observed and size of corundum grains don’t change after sintering.

Figure 1. Microstructure of the cermet sample sintered in vacuum at 1500°C by SEM method

Fig.2 shows concentration dependences of elastic moduli E and G for two series of samples (5 pieces in each) sintered at 1500 and 1600°C temperatures. We researched the samples with concentration up 2.2 to 21.0 vol%. (at less values of metal volume share sintering is not possible). As it is seen from fig.2 these curves have complicated view for both serious of samples: moduli E and G decrease and increase at concentration change, (moreover, besides) having maxima at 11.0 vol. %. Minima are observed for curves 1, 1’, 2’ at concentration 2.2-5.5 vol%.
Figure 2. Concentration dependences of elastic modules E, G for cermets, sintered at various temperatures: 1500°C – curves 1, 1'; 1600°C – curves 2, 2'.

Figure 3. Concentration dependences of thermo-conductivity coefficient (arbitrary units) in cermets, sintered at temperatures: 1 – 1500, 2 – 1600°C (λ = 1 corresponds to the standard sample value).

In work [1] the similar concentration dependences of strength for composites metal-ceramic are presented. They have two maxima at 15 and 85vol %. Authors [1] suggest the qualitative explanation of effect based on influence new inter grain phase appeared in sintering process. This phase as result thermo chemical reaction possesses new elastic properties. In the estimation authors [4] the volume share of new phase can average tens percents. Authors [1] consider formation of this phase as important factor for produce composite with optimal properties. If no components interaction exists the composite will be unusable “mechanical mixture”. However this mixture is used as simple model for estimation composite properties. In [5] this model allowed to determine elastic modules of inter grain phase for no porosity Cu powder and ceramic MgO sintered in vacuum. It should be noted that SEM and XRES methods rarely determine presence of inter grain phase (for example, she absent in fig.1). In studied cermets the above phase can exist as one of kind spinels [1], one of them we in [6] and authors [7] founded in boundary medium and identified as FeAl_2O_4. Therefore it is of interest to compare the results of studying elastic and thermo physical properties observing the formation the inter grain phase. The decision of this problem is the aim of this work.

Fig.3 shows concentration dependences of thermo conductivity coefficient λ in arbitrary units for the same samples. We see that the values λ for both series of samples decrease at 2.2 - 5.5vol% and increase for second series at 5.5 - 21vol% concentrations as for first series values λ decrease in this range. Thus minimum elastic moduli (fig.2) correspond to minimum thermo conductivity coefficient (fig.3). This effect is valid for both series samples sintered at various temperatures and in our view may be connect with the formation the inter grain phase described in [1, 2, 5, 7]. Indeed at small metal concentration (~1-2 vol%) the boundaries of inter grain phase can’t be formed, the layer is crumbly therefore the composite has low elastic moduli and thermo conductivity coefficient (fig.2,3). At concentration ~5.5vol% formation of the boundaries are completing, the inter grain layer possesses perfect structure, in this connection the composite gains high properties. If the metal concentration increases to 11vol% elastic moduli value decrease because E_{ceramics} > E_{steel} therefore value of thermo conductivity coefficient increases because λ_{steel} > λ_{ceramics}. Further increasing concentration to 21vol % lead to noticeable decrease in values E,G for all samples and value λ of one of them (Fig.3). The increase in porosity of cermets from 3 to 7% may be reason of above effect. By our opinion the gain of thermo conductivity coefficient for second series will be continued at further increasing volume part of steel because λ_{steel} > λ_{ceramics}. At steel concentration 20 – 30vol % the composite structure approximates to the model “mechanical mixture” [1].
The reason of above considered model of inter grain phase formation may be phonon theory heat transmission in crystals. The theory has been used to explain experimental results of “heat pulses” method in sintered composites [6, 7]. This method studies the time dependence of propagation of no equilibrium phonons (~10^{12} Hz) at liquid-helium temperatures in ceramics [6, 7]. In our work [6] the “heat pulses” method first have been used for cermets. It has been found that appearance of inter grain phase leads: a) to decrease of phonons diffusion coefficient (~10^3 times) as compared with value for the basic corundum ceramic, b) to decrease of composite thermo conductivity at the few metal concentrations. In addition in the work [7] phonons diffusion coefficient in nano disperse corundum-iron composites was investigated at low temperatures. It is shown that noticeable electron-phonon effect is observed if size of metal particles is compared with phonon wave length therefore “phonon capture” takes place. This effect decreases the composite thermo conductivity.

4. Conclusions
Thus in the work: 1) at few volume steel concentration the decrees effects of elastic moduli and thermo conductivity coefficient are establish; 2) complex concentration dependences of this parameters at all researched range are founded; 3) the value of optimal steel concentration for making industrial composite are determined. The employment of acoustical and thermo physical methods and also the of phonons spectroscopy results obtained earlier make possible to propose quality model of inter grain phase formation and to understand her influence on strength and thermo shock protection.

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