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Acoustic Studies of Metal–Ceramic Composites with Meso- and Nanosize Particles

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Abstract—The elasticity moduli and absorption coefficients of longitudinal ultrasonic waves with a frequency of 2–50 MHz depending on the steel concentration and sintering temperature of 1400–1700°C in vacuum were studied in samples of cermet-type composites on the basis of corundum and stainless steel. The results were discussed from the point of view of the elastic waves propagating in the fine-disperse two-phase medium at the presence of the intergrain and interphase boundaries, noticeably affecting the physical properties of the composite.

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INTRODUCTION

New composite materials based of ceramics and metal (cermets) have been under development since the 1960s. This has led to the fabrication of compounds with unique properties combining the advantages of both components: strength, heat resistance, thermal stability, wear resistance, reliable service, chemical resistance, etc. [1, 2]. At present cermets are already used in machine building (cutting instruments, high-temperature elements of gas turbines, roller bearings, precision lathes), in the medical and food industry (details of pumps and devices for transferring aggressive liquids), and in other fields. The difficulties related to the development of the new types of cermets, forecasting and study of their properties are obvious. These are complexity of the high-temperature synthesis, often requiring vacuum and high pressures, and not completely studied mechanisms of the formation of the intergrain and interphase boundaries and their effect on the strength and other physical properties of cermets, as well as self-descriptiveness and reliability of methods of their study. It is well known also that the acoustic and thermal properties of cermets are sensitive to different factors. The main factors are the porosity of the material and the structure of intergrain and interphase boundaries, which is determined by both the synthesis method of the initial material and their final thermal treatment [1–4]. Thus, acoustic studies of such composites are of high scientific interest, since the object is a two-phase system consisting of the alternating sintered dielectric grains (corundum) and metal (stainless steel) [5, 6]. In such a system, as in any sintered disperse system, the elastic, electric, and thermal properties, as follows from the literature data, strongly depend on the state and structure of the intergrain and interphase boundaries.

Cermets on the basis of α-Al_2O_3 and industrial stainless steel 12X18H9T were synthesized and studied in order to: (a) elaborate a comparatively simple and cheap technology and (b) achieve relatively high strength parameters with simultaneously small density of the ready samples. In addition, we attempted to find and reveal the possible effect of the intergrain and interphase boundaries on the elastic properties of such composites.

EXPERIMENTAL TECHNIQUES AND SAMPLE PREPARATION

We synthesized cermet samples on the basis of corundum powders (α-Al_2O_3) and industrial stainless steel 12X18H9T obtained by mutual grinding in a vibromill, then by "semidry" pressing and sintering in vacuum (the technology was described in [3] in detail). As a result, nine types of samples were obtained with three different iron concentrations in the range of 17–53 mole % sintered at 1400, 1500, and 1600°C in vacuum for 1 h. Temperatures, concentrations, and sintering duration were chosen in the preliminary experiments on obtaining mechanically strong and less porous samples. We were not aiming at fabricating a composite with optimal strengthening and other physical properties. The cylindrical samples were 10–15 mm in diameter and 10–20 mm high. The density ρ of the obtained samples was 2.7–4.3 g/cm^3, while the porosity was 1–15%, depending on the composition and the sintering temperature.

The sample composition was determined by X-ray emission spectroscopy (XRES), and their structure was determined by scanning electron spectroscopy

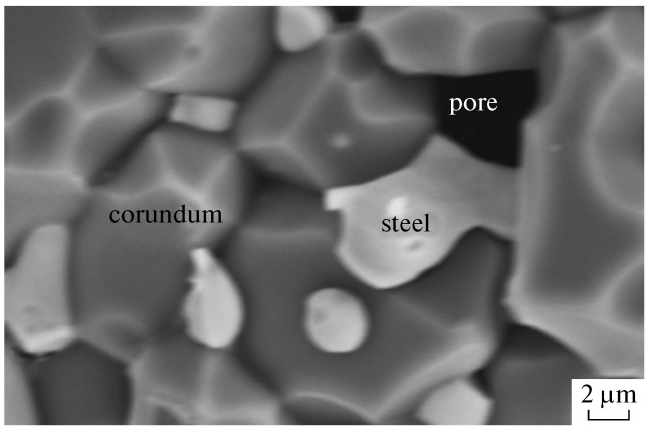

Fig. 1. SEM microstructure of the cermet sample sintered in vacuum at 1500°C. Iron concentration, 35 mole %.

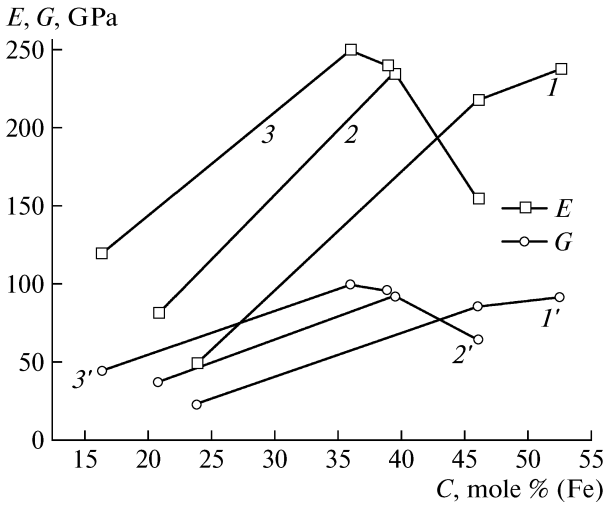

Fig. 2. Concentration dependences of the elasticity moduli E and G of cermets obtained at different sintering temperatures in vacuum: 1400°C—curves *1*, *1'*; 1500°C—curves *2*, *2'*; 1600°C—curves *3*, *3'*. The shift modulus G is marked with an apostrophe.

(SEM). The dynamic elastic Young's modulus E and shift modulus G were measured at the frequency of 2 MHz by a standard ultrasonic method [7]. The velocities of the longitudinal v_l and transverse v_s ultrasonic waves (USWs) were measured, and then the moduli were determined under the assumption of their isotropy according to the well-known relationships:

$$E = \frac{\rho v_s^2(3v_l^2 - 4v_s^2)}{v_l^2 - v_s^2}, \quad G = \rho v_s^2. \qquad (1)$$

The absorption coefficient α of the longitudinal USWs was measured by observing the echo-pulses [7]. This was possible only for low-porosity samples (about 1–10%) due to the high wave decay. Both the USW frequency (10–50 MHz) and the sample thickness were measured. All acoustic studies were performed at room temperature, and their accuracy was ~5% for the elastic moduli; ~3% for the density and ~20% for the absorption coefficient.

EXPERIMENTAL STUDIES AND DISCUSSION

Figure 1 shows an SEM image of a typical microstructure of the cermet samples. It is seen that the corundum grains with the characteristic 120° cut are densely packed with fitting along their natural faces and smaller steel particles are homogeneously distributed over the sample volume and are located either on their surfaces or occupy junctions of three to four grains. The average size of the corundum grains is about 5–8 μm, the size of metal particles is about 0.5–2 μm, and the pore size is about 2–3 μm. It should be noted that the composition and structure of the intergrain and interphase boundaries, according to the literature data, are little studied due to low sensitivity of the known physical methods, although their volumetric portion in the highly disperse solid composites can be considerable [4]. One can assume that an intermediate diffuse layer with a certain composition is formed between metal and corundum during sintering [1–4], which, however, cannot be determined from Fig. 1. In [3, 4] the thickness of this layer was indirectly estimated, being in units of nanometers. Nevertheless, XRES allowed us to find alumospinel $FeAl_2O_4$ in some samples sintered at a comparatively low temperature of 1400°C, which is probably a component of the intergrain or interphase boundary.

Figure 2 shows the concentration dependences of the elasticity moduli E and G for samples with different sintering temperatures. It is seen that, for samples with sintering temperatures of 1500 and 1600°C at an iron concentration of 35–40 mole %, the maximal values of both elasticity moduli are observed (curves *2*, *3* and *2'*, *3'*), unlike for the sample sintered at a lower temperature of 1400°C (curves *1*, *1'*). It should be noted that the porosity of samples affects the elasticity moduli [1, 2]. Its value decreases with increase in the sintering temperature. According to our estimates with the use of calculated dependences given in [2] and measured porosity values, the change of the elasticity moduli due to the difference in sample porosity is about 10%, which is much less than the concentration changes given in Fig. 2.

Figure 3 shows the concentration dependences of the density of the same samples. It follows that their density increases monotonously with increase in the iron concentration, except for the sample sintered at the highest temperature of 1600°C.

The dependences of the absorption coefficient of a longitudinal USW of different frequencies are given in Fig. 4. They belong only to samples with sintering temperatures of 1500 and 1600°C due to their low porosity and possibilities of such measurements. These

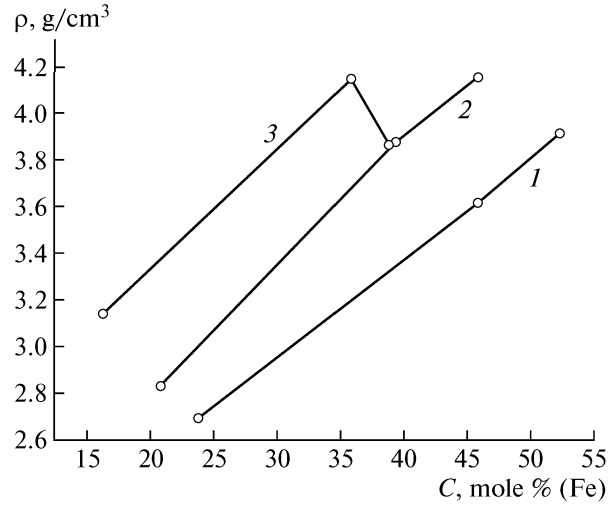

Fig. 3. Concentration dependences of the density ρ of cermets obtained at different sintering temperatures in vacuum: 1400°C—curve *1*; 1500°C—curve *2*; 1600°C—curve *3*.

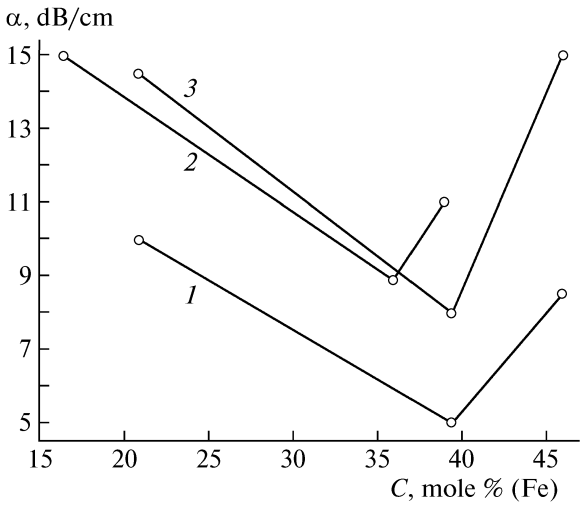

Fig. 4. Concentration dependences of the absorption coefficient α of longitudinal USWs of different frequencies in the cermet samples sintered at different temperatures in vacuum: (*1*) frequency 15 MHz, temperature 1500°C; (*2*) 50 MHz, 1500°C; (*3*) 47 MHz, 1600°C.

curves have a broad absorption minimum for the iron concentration in the range from 25–45 mole %.

The interpretation of the obtained results is not unambiguous, first, due to a great number of factors affecting the elastic properties on the two-phase finedisperse composites in different ways (porosity, grain size and boundary, etc.); secondly, due to the large length of the USW if compared with the grain size. Therefore, only the integral elastic characteristics of the samples are observed in the experiment. Nevertheless, it is possible to draw preliminary conclusions.

According to the classification of [1], the cermets we studied are part of the class of ultracermets, in which the metal phase inclusions are either in the ceramic grains or at the boundary of the grain junction. This does not contradict the structure in Fig. 1. Obviously, the properties of such cermets should depend on the properties of the metal and ceramics, the ratio of their volumes, and the adhesion between them [1]. In cermets it is necessary to establish a strong chemical bond between phases either due to the atoms of one or the other phase. According to [1], it is possible to assume the following possible concentration dependences of strength in cermets (Fig. 5). If purely mechanical adhesion occurs between phases (heterophase mixture), then strength of such a composition is minimal (curve *1*); if the bond strength at the boundary of two phases is the same as that at the grain boundaries of the first or second phase, then linear dependence is assumed (curve *2*); if the bond strength at the boundaries of the heterogeneous phases is lower or higher than the bond strength inside the first and second phases, then dependences *3* and *4*, respectively, are possible. Of most interest to practice is the case in which, at low concentrations (10–15 vol %) of the second phase, the main phase strengthens and high service properties of cermet are achieved (curve *5*). The dependences shown in Fig. 5 (curves *1*–*5*) probably do not exhaust all possible cases, since our results do not fit this picture. In fact, the concentration dependences of the elasticity moduli E and G (Fig. 2) have a more complicated character, even taking into account only data for three concentrations. In addition to the noticeable maximum at the iron concentration of 35–40 mole %, it is possible to have one or two minimum (a) at lower corundum or iron concentrations. The dependence we propose can look like curve *6* in Fig. 5. In fact, the values of the elasticity moduli for "pure" sintered corundum are $E_1 = 380$ and $G_1 = 170$ GPa, and for "pure" stainless steel they are $E_2 = 180$ and $G_2 = 70$ GPa. Therefore, at low concentrations of both components, the cermet moduli probably should be smaller than the maximal values in Fig. 2.

In cermet literature it is noted that the formation of a strong chemical bond between ceramic grains and metal is accompanied by the formation of an intermediate layer, i.e., a new phase the properties of which mainly determine the characteristics of the sintered composite in spite of the estimated layer thickness of several nanometers. In [4] the elastic characteristics of the intergrain boundary were estimated. They can be used when calculating diffusion of thermal phonons performed in [3]. In our samples this layer is reliably recorded by XRES and its composition is determined: alumospinel $FeAl_2O_4$ formed at low (1400°C) sintering temperature. It is possible that a layer of such composition is tens of nanometers thick and has a "friable" structure; therefore, it has low elasticity moduli, which decreases the strength of the composite on the whole and decreases its moduli (curves *1* and *1'* in Fig. 2).

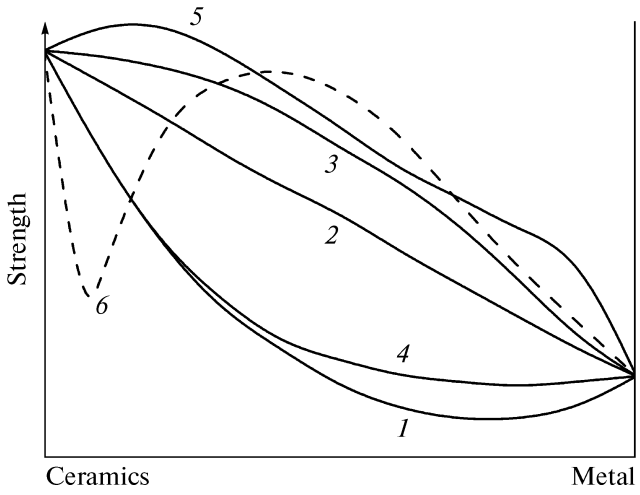

Fig. 5. Possible concentration dependences of cermet strength discussed in [1]: (*1*) mechanical mixture of phases; (*2*) bond strength at the boundaries of the grains of the first or second phase (*3*), (*4*) bond strength at the boundaries of the heterogeneous phases is less or more than the bond strength inside the first and the second phase, respectively; (*5*) cermets with disperse strengthened matrix; (*6*) dependence for the studied cermets proposed in this work.

At higher sintering temperatures, the intergrain boundary can be much thinner and non-XRES-detectable, though it is manifested in increase of the composite elasticity moduli (curves *2*, *2'* and *3*, *3'* in Fig. 2) and decrease in the USW absorption coefficient in Fig. 4. As to decrease of the elasticity moduli when the iron concentration increases, it is likely that its excess leads to a double effect: (a) increase of the volumetric portion of steel with lower E and G than those of corundum and (b) thickening and change of the structure of the intergrain boundary decreasing its elasticity and increasing USW absorption.

Thus, the study of the concentration dependences of the E and G moduli allows one to choose the optimal steel concentration for obtaining a composite with the desired elastic properties. The decrease of the absorption coefficient of the longitudinal USW in the concentration range optimal in elasticity (Fig. 4) may also indicate the perfection of the intergrain boundary in this range. It should be noted that the properties of the intergrain and probably interphase boundaries determine not only the elastic properties of the corundum–metal composites, but also their thermal conductivity important for cermet thermal stability [8]. In this respect, earlier in [3] we comparatively studied the estimate of the diffusion coefficients of the "thermal" phonons (~10^{12} Hz) in the initial corundum ceramics and our cermet samples with one steel concentration by the "ballistic" phonons method. It was established that the diffusion coefficient of the "thermal" phonons in cermet is two orders of magnitude smaller than in ceramics. This is explained by the presence of their strong reflection at the intergrain boundaries and, consequently, worsening of diffusion. As to further studies, it would be useful to perform analogous studies of a series of cermets described in this work for the comparative estimation of the self-descriptiveness of both methods and clarifying the role of intergrain and interphase boundaries.

CONCLUSIONS

Thus, in this work we synthesized and studied by the acoustic method a composite material with high elasticity, strength, and wear resistance. This makes it promising for practical applications. One can state that acoustic studies of sintered composites with meso- and nanosize particles have significant informational content, not only with the aim of obtaining material with optimal elastic and strength properties, but also to indirectly observe the formation of the intergrain and interphase boundaries, and, in combination with other methods, to assess their properties.

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