

Metal-ceramic composite development based on its modelling results

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Abstract. The modeling (and its experimental verification) of packing and deformation of the composites consisted of aluminum-magnesium alloy AMg6, B₄C powder and W nano-powder has been performed. The powder compositions were determined using discrete element modeling of the composite particles packing based on the particle size distribution functions of real powders. The models of maximum mixture packing densities have been rendered.

1. Introduction

The development of nuclear energy applications, space exploration, and the use of radiation in medicine depends on the solution of various problems of the protection of structural materials, electronic components and products, and biological tissues from their exposure to ionizing radiation. Therefore, the design and development of new materials to improve the effectiveness of anti-radiation protection is an important modern area of research. It includes the reduction of the weight and dimensions of radiation-shielding materials.

Light metal alloys have good mechanical properties, but when subjected to radiation, they are susceptible to swelling and structural changes. These defects can be prevented by creating metal-matrix composites containing particulate fillers with the required radiation-shielding properties. The use of radiation-absorbing nanoparticles of heavy metals and ceramic materials as fillers dispersed within metal-matrix composites improves these composites' absorption properties with respect to the neutron, gamma and X-rays [1, 2].

It is shown in [3] that the use of Al/Mg matrix filled with nano-scale particles of radiation-absorbing ceramic and metallic materials (BN, B₄C, Pb, and W) increases the rate of neutron absorption factor by 50% and the gamma radiation scattering coefficient by 30-40%.

To manufacture this kind of a composite various approaches have been used [4, 5]. In particular, the method of hot extrusion has been applied [3]. The composite included 60-65wt% of aluminum-magnesium alloy (grades AMg6 or V95), 15-20wt% of W nano-powder, and B₄C (or BN) – the rest.

The present work objectives include the modeling (and its experimental verification) of the process of the consolidation of the composites consisted of aluminum-magnesium alloy AMg6 (65 wt%), B₄C powder (15 wt%) and W nano-powder (20 wt%), as well as the optimization of the composite content to achieve higher density.

2. Powder materials and experimental procedures



The characterization of initial powders and their mixtures has been performed by SEM and EDS analysis (JSM-7500F JEOL; LEO EVO 50 Zeiss), XRD analysis (XRD-7000S Shimadzu), particle size distribution analysis using laser diffraction technique (SALD-7101 Shimadzu), and specific surface analysis by BET method (Sorbi META).

The powders were mixed by ball milling in a roller mill using zirconia milling balls. The densification behavior of the dry mixed composite powder has been analyzed using a computer controlled hydraulic precision press (IP-500M-auto ZIPO). The consolidation of the mixed composite powder was conducted using pressureless sintering in a vacuum furnace VHT 8/22GR Nabertherm. The sizes of green samples were 15 mm in diameter and 3-6 mm in height. The mechanical properties of the consolidated specimens were determined by nanoindentation (DUH 211S Shimadzu) of the polished surfaces of sintered samples.

AMg6 powder: the commercial magnesium aluminum alloy powder AMg6 (AA5556 alloy analogue) consists of particles with a shape close to spherical, with a wide spread in particle sizes from 1 μm to 70 μm ($d_{50} \approx 15 \mu\text{m}$). The conducted XRD and EDS analyzes of the AMg6 powder indicate less than 2% impurities. The broadening of the XRD reflexes confirms the presence of submicron fractions in the powder. The degree of agglomeration of the powders has been assessed in terms of the ratio of the mean particle size obtained by laser diffraction method and the average particle size obtained by the BET method. AMg6 powder agglomeration degree is equal to 2 (Table 1), *i.e.*, this powder was weakly agglomerated.

B₄C powder: the commercial boron carbide powder produced by "OKB-BOR" (Russia) has a nearly equiaxed particle shape with a wide spread in particle sizes from 0.1 μm to 10 μm ($d_{50} \approx 1.22 \mu\text{m}$). The amount of fraction of nanoparticles up to 10% has been determined. The conducted XRD and EDS analyses did not reveal any impurities in the powder. B₄C powder agglomeration degree is equal to 4.43 (Table 1).

W powder: tungsten powder was prepared by electric explosion of tungsten wire [12]. The powder particles have a spherical shape and wide bimodal size distribution (from 0.05 to 0.4 μm and from 0.4 to 9 μm). The powder contains 20% nano-sized (80-200 nm) and 20% submicron (200-900 nm) fractions, and therefore the degree of agglomeration of the W powder is 9.3 (see Table 1). No impurities in tungsten powder were detected.

The summary of the dimensional characteristics of the starting powders is provided in Table 1. The analysis of the obtained data indicates that initial raw powders have fairly wide particle size distribution, but their mixture can be optimized with respect to the concentration of the components to form a continuous alloy matrix and to increase the packing density of the particulate filler. Given the morphological characteristics of the powders, the maximum density of packing can be expected when filling the space between tightly packed particles of the matrix alloy by a mixture of boron carbide particles with tungsten particles' agglomerates. Parameter d_m in Table 1 has been determined for spherical particles using specific surface value S_{BET} (BET data) and theoretical density ρ , for respective materials [6]:

$$d_m = \frac{6}{\rho S_{BET}} \quad (1)$$

Table 1. Average particle/agglomerate sizes of the components in the powder mixture

Component	Average size (micron) of structural elements according to different analysis				Agglomeration degree
	XRD (crystallite size)	SEM (agglomerate size)	Laser Diffraction (agglomerate size)	BET (particle size d_m)	
AMg6	0.2	15.4	13.6	6.8	2

B_4C	0.04	1.8	1.22	0.28	4.43
W	0.09	1.1	1.07	0.12	9.3

3. Optimization of powder mixtures

Nearly spherical and equiaxed particle shape of the used powders enables the modeling of their packing and optimization of their mixing ratio in terms of the values of the average number of inter-particle contacts (the coordination number N_c) and of the packing density. For this purpose, the particle size distributions obtained by laser diffraction have been approximated by matching functions. These functions were used to define the relevant classes of particles in the framework of the discrete element model environment of the S3D PorouStructure™ software utilizing the Ichikawa algorithm (with central packing). The total number of particles in a representative model set (Figure 1) varied from 20000 to 72000.

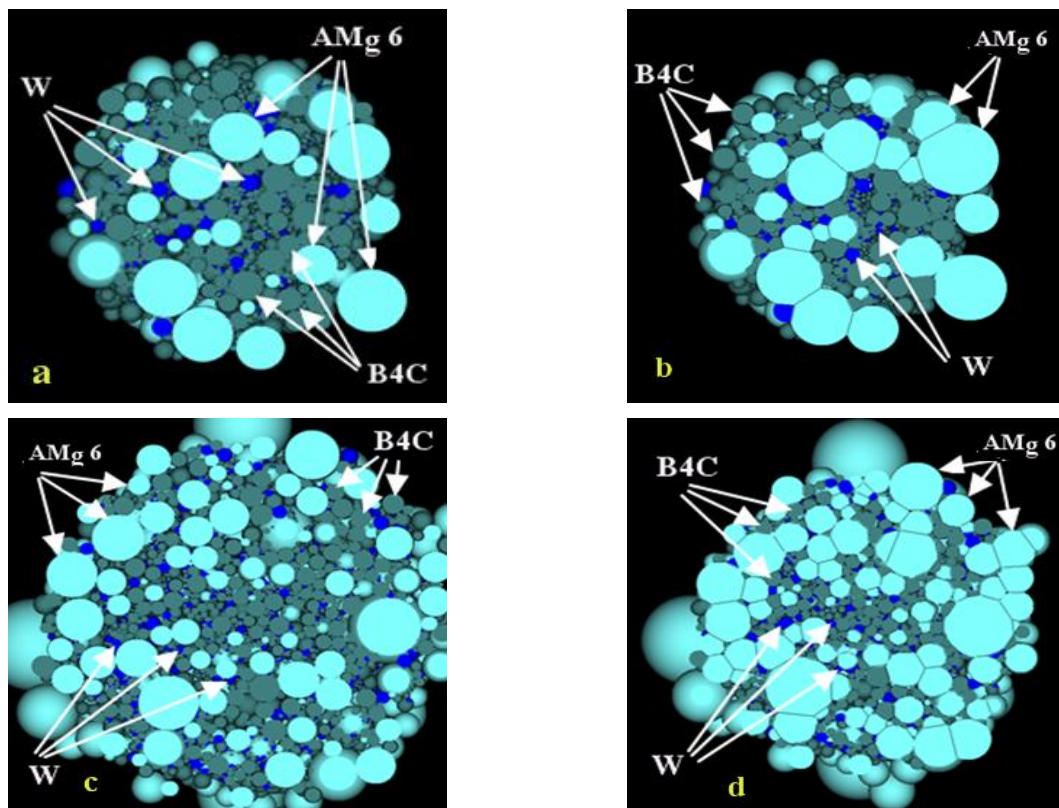


Figure 1. Models of packing for the composites with minimum permissible (a, b) and the optimal density of the matrix alloy and particulate fillers, (c, d) in a free-laying state (a, c) and after plastic deformation of the particles (b, d): AMg6 particles – blue; B4C - gray; W – dark blue.

The comparative analysis of the results of the modeling of the packing of the composite structure AMg6 (65 wt.%) + B₄C (15 wt.%) + W (20 wt.%) was carried out using S3D Evolution™ code. For this simulation, the compaction parameters were associated with the deformation of the particles of the matrix alloy and tungsten, subjected to pressures from 40 MPa to 800 MPa (including at temperatures up to 600 °C). The particles of boron carbide were considered to be non-deformable (Figure 1b, d).

The conducted packing's statistical analysis based on the criteria of the maximum density and of the ensured continuous contact of the aluminum matrix particles showed the following. The minimum

composition forming a contiguous aluminum alloy matrix rendering the optimal dense filling of the dispersed particles of tungsten and boron carbide was found to be 65% AMg6 + 15% B₄C + 20% W (Figure 1a, b). For such a mixture in the initial underformed packing state, the alloy particles' average partial coordination number (the number of contacts between alloy particles only, excluding tungsten and boron carbide particles) is in the range of the minimum allowable values ($1 < N_c < 2$), and the space between them can be filled by the optimal content of tungsten and boron carbide particles with a predetermined size distribution. However, the mixing ratio rendering the maximum packing density of the particles has a different composition: 74% AMg6 + 6% B₄C + 20% W (Figure 1c, d). For this composition, the partial coordination number of the matrix alloy particles ranges from 3 (in a loose powder state) to 4 (after plastic deformation of the particles), and the partial density of the alloy particle packing varies from 43% (Figure 1c) to 67% (Figure. 1d), respectively.

The quantitative results of the modeling for the two above-mentioned mixtures are presented in Table 2. According to the table, the increase in the partial coordination number from 1.78 to 4.29 in a continuous matrix AMg6 can be achieved by increasing the content of the powder alloy by 9 wt.% (from 65% to 74%). In this case, the strength of the composite can be increased by at least 2.5 times, assuming that the matrix material is the only component in the consolidated mixture capable of bearing mechanical load, and the strength of the porous material is directly proportional to its density. An improvement of the strength of such a composite is also possible when using a fine powder of boron carbide which would effectively fill the space between the densely packed particles of the matrix alloy.

Table 2. Parameters of deformed packed structures of composite's particles

Characteristics of composite with optimal particles density packing and minimum possible content of the matrix alloy (after deformation 25%)					
Component (theoretical density, g/cm ³)	Content,%		Partial packing density	Average coordination number for the component particles	Partial coordination number for particles in the powder component
	wt	vol			
AMg6 (2.6)	65	77.9	0.276	95.04	1.78
B ₄ C (2.52)	15	18.8	0.498	6.23	4.42
W (19.25)	20	3.3	0.057	5.17	0.83
Mixture (3.14)	100		0.782	6.49	6.49
AMg6 (2.6)	74	89.2	0.67	124.7	4.29
B ₄ C (2.52)	6	7.5	0.254	6.34	3.28
W (19.25)	20	3.3	0.052	5.23	1.54
Mixture (3.14)	100		0.89	7.23	7.53

4. Analysis of composite material consolidation

The powder compressibility characterization has been performed by plotting the compaction curves $\rho(P)$ obtained for the conditions of uniaxial quasi-static pressing with loading-reloading cycles and using an approximation by a logarithmic equation in a dimensionless form [7, 8]:

$$\rho = b \cdot \ln\left(\frac{P}{P_{cr}}\right) + 1 \quad (2)$$

where ρ is the relative density of the green body; P is the applied compaction pressure; P_{cr} is the critical pressure (pressure enabling compaction up to full density of the powder material); b is a material constant defining densification pressure sensitivity. Due to the limitations on the values of the relative density ($0 < \rho \leq 1$), equation (2) is valid for a limited range of the values of the applied

pressure:

$$\frac{P_{cr}}{\exp(1/b)} < P \leq P_{cr}.$$

The results presented in Figure 2 (with approximation certainty not less than 99.9%) show that, for the conventional quasi-static pressing in a rigid die, the critical pressure value P_{cr} for the tested composite is 5.219 GPa, which exceeds the strength limit of hardened steel – the structural material of the pressing die. While the AMg6 alloy powder without a particulate filler may be densified at low temperatures down to a non-porous state under the critical pressure of 1.262 GPa using a special tungsten carbide die of the collector type [8], the achievement of the critical pressure of 5.219 GPa for pressing bulk components of complex shapes made of the studied composite material appears to be impossible.

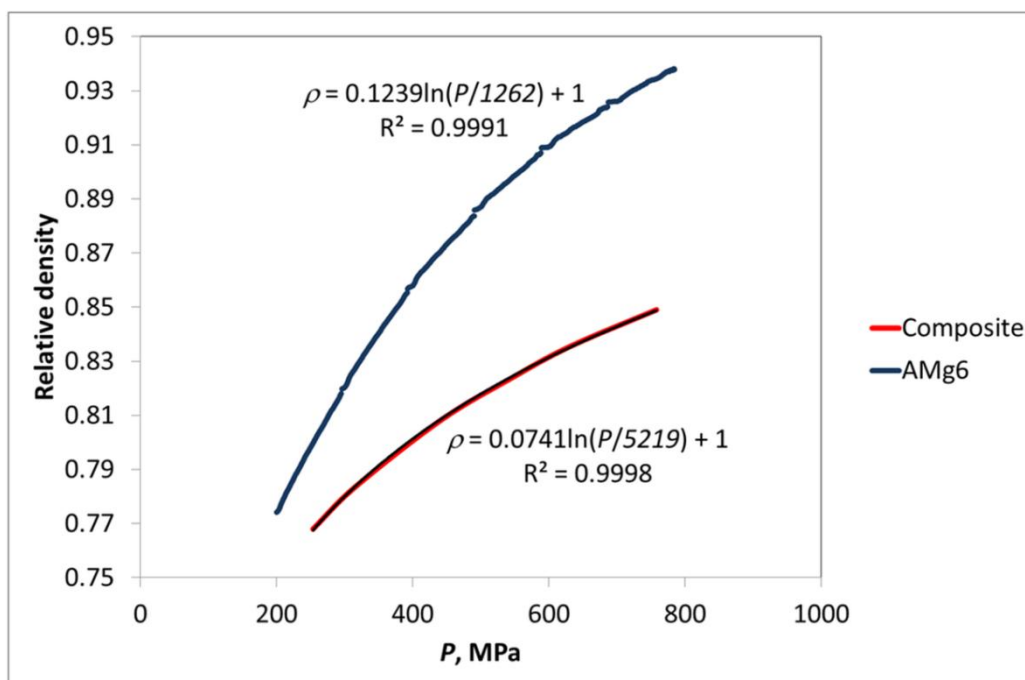


Figure 2. Pressure-density dependence of AMg6 matrix alloy and composite (65%AMg6+15%B4C+20%W) powders compacted by conventional quasi-static pressing in a die

At pressure of 750 MPa the composite powder (65% AMg6 + 15% B₄C + 20% W) samples with a density of 85.3% were obtained by the conventional cold quasi-static pressing. During free sintering of these samples in a vacuum at temperatures of 450, 500, 550 and 600 °C the linear dimensions of the

processed specimens remained unchanged up to the melting point of the aluminum-magnesium matrix. This fact indicates a non-significant contribution of the pressure-less solid state diffusion-based mass transport into the densification of the studied powder composites.

For the powder with an optimized composition (74% AMg6 + 6% B₄C + 20% W) at the same pressure of 750 MPa the density of 91.6% has been achieved, which confirms the results of the simulation-based optimization of powder mixtures (Figure 1,c, d; Table 2). Density after conventional compacting at 800 MPa was equal to 92.6% and 95.1 % with using powerful ultrasonic action (PUA) [8]. After sintering for 2 hours at 590 °C, the density was equal to 93.4% (without PUA) and 96.8 % (with PUA).

It should be noted that for the tested composite the conventional quasi-static pressing may be applied also in the case of the yield strength reduction of the AMg6 alloy even under ambient temperature conditions. One can use a well-known acoustic-plastic effect by utilizing powerful ultrasonication during materials compression, which can significantly increase the concentration and mobility of dislocations of the crystal lattice of the matrix material, thereby reducing its yield strength. In particular, according to [9], the effect of ultrasound on the deformation process of aluminum at an oscillating stress amplitude of 0.2 kg/mm² (~1.9 MPa) and under static loading of 1 kg/mm² (~9.8 MPa) increases the steady creep rate almost 10 fold, and the yield strength decreases by half.

Additional effects can be achieved by using carbide materials for the collector type dies, which enable the achievement of the pressure levels higher than the tensile strength of the material of such dies [8].

The equation (2) will be utilized not only for the description of the plastic deformation of the AMg6 matrix alloy, but also as one of the possible models for the analysis of the consolidation of the studied composite material, containing boron carbide particles, which are essentially non-deformable in the specified pressure range.

5. Conclusions

1. Discrete element modeling of the composite particles' packing based on the particle size distribution functions of real powders describes well the mixing of powder components and enables the determination of the powder compositions rendering maximum mixture packing densities.
2. The optimal content of the studied composite has been determined (74%AMg6+6%B₄C+20%W). This content provides the maximum dense particle packing for all the components and strong alloy matrix with the coordination number greater than 4 and packing density of 0.67. The experimentally obtained density of this composite material subjected to the conventional quasi-static pressing (under pressure of 800 MPa) was achieved to be 92.6% and 95.1% after PUA. After sintering for 2 hours at 590 °C, the density was equal to 93.4% (without PUA) and 96.8% (with PUA).

Acknowledgements

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