PRODUCING METAL MATRIX COMPOSITES WITH NON-AGGLOMERATED NANODIAMOND REINFORCEMENTS BY CONSOLIDATION OF MECHANICALLY ALLOYED GRANULES WITH LIQUID PHASE FORMATION

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Abstract

This work deals with the use of liquid forging for consolidation of granules from a composite with a high content of nanodiamond particles. The developed method includes: 1) the treatment of nanodiamond powders and metal particles in a planetary mill, i.e., mechanical alloying, to produce composite granules with non-agglomerated uniformly distributed nanodiamond particles; 2) arrangement of granules in a mould; 3) heating of the mould to a temperature close to the melting temperature; 4) compaction of heated granules into bulk material; 5) cooling under a load. During the compaction, granules are totally or partially melted to enable elimination of the boundaries between granules. The absence of the boundaries leads to increase the strength of the composite.

1. Introduction

Industries constantly require materials with enhanced operational characteristics. Properties unattainable in common materials occur in composites. Nanocomposites can potentially combine all the advantages of nanomaterials and composites and demonstrate higher levels of operational characteristics; for this reason, research into nanocomposites has been lately given increased attention [1-5]. As reinforcing nanoparticles, particulate composites often make use of carbon materials, which occupy a special place in the nanoworld owing to their great diversity. The main and well-investigated crystalline modifications of carbon are graphite and diamond. Diamond, due to its unique physico-chemical characteristics, is widely used in technology; for this reason, interest in production of artificial diamond crystals, in particular, in the detonation transformation of explosives, has revealed itself many years ago. In 1963, nanodiamonds (ultradispersed diamonds) were discovered. Nanodiamond (ND) particles formed in detonation synthesis are, mainly, 4–6 nm in size; herewith, 10- to 20-nm particles are also observed. ND particles have a cubic crystal lattice with the lattice parameter α = 0.3575 nm (in natural diamond, α = 0.3566–0.3567 nm) [6, 7]. Nanodiamond powders have a complex multilevel structure. Primary nanodiamond particles (d ~ 4 nm) are combined into strong cluster aggregates 40 to 400 nm in size. In turn, primary aggregates are combined into secondary aggregates and agglomerates 0.4 μm in size. Remnants of non-diamond carbon forms not eliminated in oxidation are distributed predominantly on the surface of crystallites inside aggregates. Diamond particles were found to have on their surfaces various oxygen-containing hydroxyl, carboxyl etc. groups and, to a lower extent, methyl and nitrile groupings.

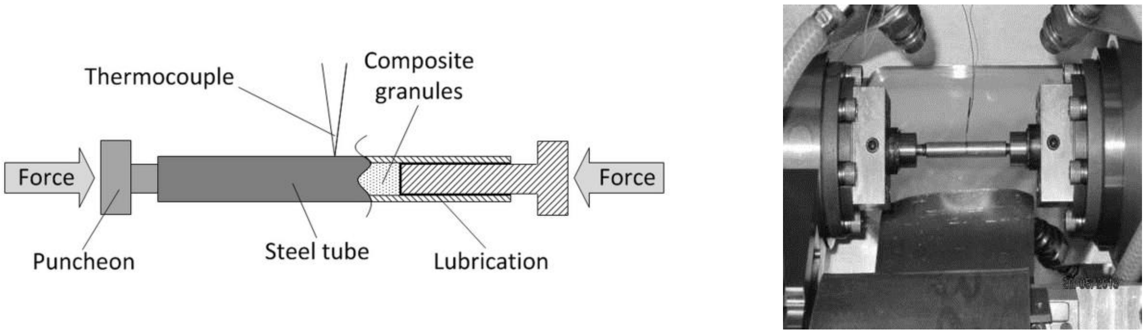
Altogether, the whole body of experimental facts suggests that ND can be assigned to fractal systems, and the essential stage of the ND structure formation mechanism is fractal formation by the cluster–particle and cluster–cluster type to form multilevel macrostructures.

Composite materials with nanodiamond reinforcing particles are produced by various methods: liquid metal infiltration [8, 9], spark plasma sintering [10], electrochemical methods [11]. Mechanical alloying, however, can be recognized to be the most promising. This technique makes it possible to break down agglomerates of nanoparticles and produce composite granules with a uniform distribution of non-agglomerated nanodiamonds in the metal matrix. Consolidation of these granules leads to produce a compact composite with non-agglomerated reinforcing particles.

The process flow diagram of consolidating fabricated items by powder metallurgy methods consists of the following major operations: production of powders with required properties (in our case, mechanical alloying to produce composite granules); preparation and compaction of powders; sintering of produced blanks; subsequent pressure shaping, thermal or chemical thermal treatment. An efficient way is also to combine compaction and sintering, i.e., warm pressing. However, at a high content of nanodiamond particles in the composite, compaction of granules according to this protocol is made difficult because granules with high nanodiamond content possess an increased strength and hardness. Compact material may have porosity and break down by the boundaries of granules. To eliminate these drawbacks, a novel technique of producing composites with a high content of reinforcing nanoparticles was worked out. The developed technique includes: 1) the treatment of nanodiamond powders and metal particles in a planetary mill, i.e., mechanical alloying, to produce composite granules with a uniform distribution of non-agglomerated nanodiamond particles; 2) arrangement of granules in a mould; 3) heating of the mould to a temperature close to the melting temperature; 4) compaction of heated granules into bulk material; 5) cooling under a load. During the compaction, granules are completely or partially melted for a short time; this makes it possible to eliminate boundaries between granules. The absence of boundaries leads to increase the strength of the composite. Consolidated material should be cooled under a load to eliminate the formation of cavities that may form because at high temperatures the geometric size of articles is much larger than in cold state (the thermal expansion phenomenon). This technique can be attributed to the variant of liquid forging or squeeze casting.

2. Materials and methods

Composite material “aluminium + nanodiamond reinforcing particles” was chosen for studies. The nanodiamond volume fraction was 10, 20 and 30%. Nanodiamonds produced by Kombinat “Elektrokhimpribor” were used. Primary nanodiamond particles 4–6 nm in size were combined into agglomerates whose size reached 50–100 μm. Mechanical alloying was carried out in a Retsch PM400 planetary mill in an argon atmosphere, without using surfactants, in sealed steel grinding jar containers of 500-ml nominal volume. The technological milling tool was chromium steel balls 12 mm in diameter. The following technological conditions were chosen: (i) the ratio of the weight of the balls to that of the treated mixture was 8:1; (ii) rotation velocity of the grinding jar containers around the common axis (the rotation rate of the carrier) was 300 rpm; (iii) the grinding jar containers were air-cooled during the operation; (iv) to prevent strong overheating, the mill was stopped for 5 min each 5 min of operation; the treatment time (without time for stops) was varied from 1 up to 3 hours. Compaction of composite material granules chosen for consolidation was performed on a Gleeble System 3800 for physical simulation of thermal mechanical processes in a steel attachment consisting of a tube (inner diameter, 6 mm; length, 50 mm) and two male die parts. The scheme of work and general view of the equipment are given in Fig. 1. Powder was preliminarily compacted at room temperature at a pressure of 70 MPa. Then it was heated by the direct passage of current in a low vacuum medium (10^{-1} mm Hg) at a rate of 30 degrees/min with pressure maintained at 50 MPa. Temperature was controlled by a chromel–alumel thermocouple welded to the middle of the steel tube. Compacted granules were held at temperatures of 600, 660 and 680°C (three specimens, each at a different temperature) and pressure of 50 MPa for 2 min and then cooled at a rate of 50 degrees/min. The obtained cylinders were cut in transverse and longitudinal directions; sections were prepared on a LaboForce polishing machine. The structure study was done on JEOL JSM-6700F field emission scanning electron microscope.



a

b

Figure 1. Scheme of work (a) and general view of the equipment (b)

3. Results and discussion

The morphology study of the granule surface showed nanodiamond particles to embed into the aluminium matrix and to have a firm defect-free contact with it, which should lead to strengthen the material.

The percentage of nanodiamonds and time of treatment in a planetary mill have a significant effect on the uniform distribution of reinforcing particles. Increased contents of reinforcing particles require greater treatment times. Even a 3-hour treatment was not sufficient for the uniform distribution of nanodiamond particles in the aluminium matrix for a composite with 30 vol.% nanodiamond particles; a considerable amount of nanodiamond particles was observed on the surface in an agglomerated state.

It was also found that in some cases significant amounts of particles of oxides occurred on the surface of granules. Analysis showed the presence of aluminium oxide. In the case of the complete breakdown of agglomerates and the uniform distribution of nanoparticles in the matrix, no increased content of oxides was observed on the surface of granules.

Granules with the nanodiamond content of 20 vol.% after the treatment for 3 hours were chosen for compaction by the developed technique. The appearance of such granules is shown in Fig. 2. It is clearly seen that agglomerates are completely split, i.e., nanoparticles are in a non-agglomerated state and are uniformly distributed in the aluminium matrix.

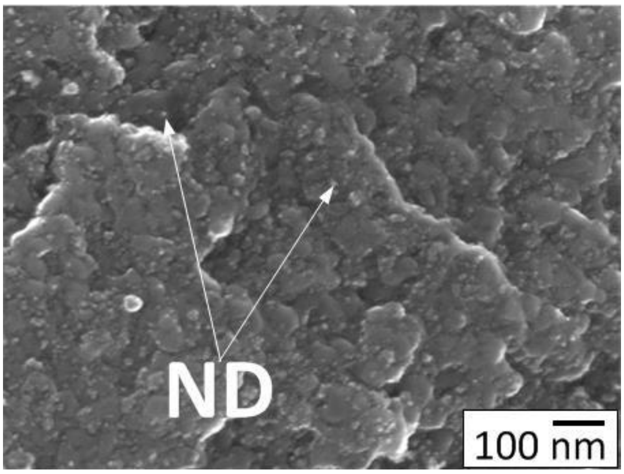
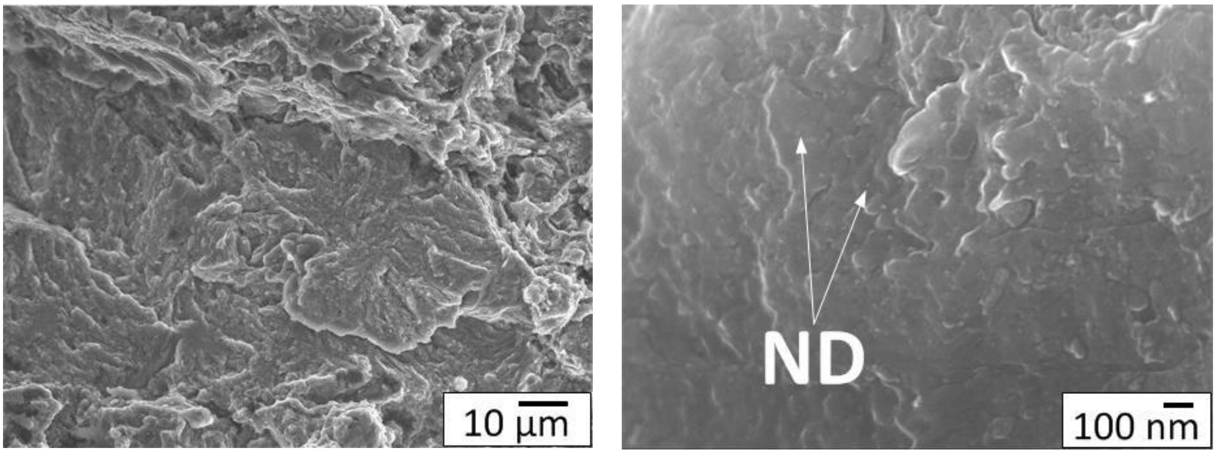


Figure 2. General view of composite “Al+ND” granule surface

The structure study of consolidated specimens on transverse and longitudinal sections by methods of scanning electron microscopy showed that the developed compaction technique totally eliminated the boundaries of granules if the matrix was completely or partially melted. Non-agglomerated nanodiamond particles were uniformly distributed in the matrix. Figure 3 demonstrates the formed structure.



a b

Figure 3. Fracture of compacted specimens of composite “Al+ND” under small (a) and big (b) magnification

There was no complete consolidation of material at temperatures deliberately lower than the melting temperature. It is evident that a higher pressure is required in that case. An increase of temperature resulted in the melted part of the matrix flowing out to gaps of the mould and hardening; this did not enable application of a load to the produced compact during its cooling. This led to microdiscontinuities in material; it can be said that the shrink hole was spread in the entire volume of the specimen. The choice of optimal process regimes (heating temperature, 660°C; pressure, 50 MPa) enabled the formation of a defect-free composite structure with non-agglomerated uniformly distributed nanodiamond reinforcing particles.

4.Conclusion

A technique of consolidating composite granules produced by mechanical alloying was developed. The technique includes the arrangement of granules in a mould; heating of the mould to a temperature close to the melting temperature; compaction of heated granules into bulk material; cooling under a load. The studies have shown that the developed technique of consolidating composite granules makes it possible to produce compacts with a defect-free structure and a uniform distribution of nanodiamond reinforcing particles in the aluminium matrix.

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