Liquid Phase Production Technologies of Metal Matrix Composites (Review)

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ABSTRACT

Liquid phase production technologies of metal matrix micro- and nanostructural composites are considered. Complex using magnetohydrodynamic stirring, ultrasonic treatment of melt during liquid and liquid-solid states and thixocasting advantages are proposed.

Keywords: metal matrix composites; liquidphase technologies; magnetohydrodynamic stirring; ultrasonic treatment; thixocasting

1. Introduction

The research and development of metal matrix composites (MMCs) are given a significant consideration practically in all economically developed countries due to the complex of mechanical and service properties that could be obtained in this class of structural materials and that are unattainable in the traditional materials, produced using the traditional technologies. Metal matrix provides a number of advantages if compared to other (polymer, carbon, ceramic) matrices, in particular, higher hardness, strength, electric and heat conductivity, crack resistance, melting temperature, the possibility of the use of the liquid phase technologies. The use of the liquid phase technologies means that in the process of MMCs production at least one of the components is in the liquid phase (casting technologies, liquid forging, laser and plasma spraying, sintering with liquid phase, etc.)

Depending on the method of their production, the two types of composites are distinguished: artificial and natural. In artificial composites the strengthening disperse phase is either introduced artificially from the outside or is formed when matrix melt interacts with artificially introduced agents. Natural composites include the alloys, in which the disperse phases are formed under the natural processes of primary, in particular – oriented crystallization. Typical natural composites are graphitized cast irons. The properties of natural composites can also be improved using the technologies typical to artificial composites. However, artificial composites are regarded as the prospective materials with unique properties.

Composites, reinforced with fibers or whisker crystals, and layered composites are widely used in industry. The technologies of their production are relatively simple, the scientific bases of their development, analysis and prognostication of their behavior while using the articles are thoroughly studied. Excluding the production of prepregs, that serve as semi-finished articles for the production of the constructions, the composites of this type and the constructions are produced simultaneously, allowing of considering the specifics of the use of these constructions and purposely orient. These composites obtain a number of useful properties (high specific strength, hardness, wear resistance, fatigue resistance, etc.), at the same time possessing substantial defects (anisotropy of properties, high cost, low maintenance workability, etc.), that result in a narrower range of their application, specific technology and engineering properties taken into consideration.

Dispersion-reinforced composites with disperse particles as reinforcers are more general-purpose structure materials in the field of their application and manufacturing capability. This type of composites includes MMCs with casting alloys and wrought alloys as a basis and disperse particles as reinforcers, artificially incorporated (ex-situ-processes) or initiated as a result of the occurring (in-situ) processes. Generally, refractory high-strength, high-modulus particles of oxides, carbides, borides, nitrides (more frequently SiC, Al₂O₃, B₄C, TiC) are used as micrometric size reinforcers. Chemical reactions in-situ, occurring in the melt in the process of incorporation of the reactive metals, gases or chemical compounds, form thermodynamically stable, wetted by the melt due to the coherent boundary formation, thermostable at high temperature of maintenance reinforcing phases [1].

Processes of composite production, where the processes of spontaneous high-temperature synthesis are used to produce reinforcing particles in the metal melts, pertain, in fact, to in-situ processes. This technology draws attention not only as a method of the finished article production, but also as the possibility to produce composites with high quantity of reinforcing disperse particles of various phase content, that serve as addition alloys in the cast MMCs production.

Comparatively high cost of the refractory particles, necessary to incorporate into the melt, technological problems connected, in particular, with providing the wetting of the incorporated refractory particles by the melt and with preserving sedimentation stability of the produced MMCs – all these factors in some of cases offer advantages to in-situ processes. However, the main criteria for choosing the matrix melt composition, size, quantity, nature of the reinforcing phase and method of its implantation into the melt, method of shaping and conditions of obtaining properties of the blanks are the requirements to the finished article properties, stability of the structure and the properties of these articles in the process of their use. This requires diversity and complexity of the technological solutions for the MMCs article production.

Three basic processes of MMCs production are used:

- incorporation of particles into the melt, intensively mixing with impeller or magnetohydrodynamic (MHD) mixer;
- impregnation of disperse particles or preforms by matrix melt;
- powder technology.

Over 60 foreign companies with various production volume specialize on the production of composites (mostly based on aluminium alloys), and also on reinforcers, mainly SiC and Al₂O₃. A number of foreign companies, in particular Advanced Reflectory Technologies (ART), DWA AL Composites (DWA), Metal Matrix Cast Composites (MMC) (USA), Aerospace Metal Composites (AMC), AMETEK Specialty Metal Products (Great

Britain), widely use the composites, reinforced with refractory particles and fibrous preforms, for the articles of aerospace and military equipment.

Excluding the cases when powder technology is implemented by the compaction of the original matrix alloy powders and reinforcers in the solid state, the all three processes are connected with using liquid phase technologies to produce MMCs.

Considering the problem of the prospective development and implementation of MMCs, it is necessary to take into account that the objectives of the development of the composites with the given level of properties is only a part of the objective concerning the finished article production out of these composites. Therefore, in the end, high level of the certain composite properties, obtained at their evaluation by the results of the standard sample test, may not always serve as a structural strength indication of the given material. As a rule, it is necessary to be guided by the optimum value of the property or a group of interconnected properties, that determine the operating capacity of the finished product, with limitations imposed on the other, less important properties. New engineering solutions may be possibly used for the articles, in which composites substitute the traditional materials.

Despite the abundance of works dedicated to the research and production of MMCs, their implementation is still in the semi-industrial production stage. One of the reasons of such situation is limited possibilities of the MMCs use for the production of the cast shapes of varied weight and dimensions with complex internal cavities. Due to their general-purpose properties, the traditional foundry technologies, implying the filling of the molds with the liquid-state alloys, as well as thixotechnologies, implying the filling of the molds with the two-phase-state alloys, are the most appropriate.

Specific feature of the cast MMCs is the need to provide equal distribution of the disperse particles in the melt volume for all stages of cast ingot produced by the traditional casting methods (gravitation casting, high-pressure casting, etc): in the process of particle incorporation into the melt or the particle formation initiation in the melt in the melting unit or in a special device, in the process of liquid-state treatment in the intermediate (pouring) device, when running through the gating system, and, finally, in the process of ingot formation in the mold. Therefore, the problem of sedimentation stability of the cast composites is closely connected not only with MMCs property and structure formation but with the implementation of the casting production methods.

MMCs based on the aluminium and magnesium alloys are the prospective materials for the various branches of industry due to low unit weight and higher, in comparison with the matrix alloy, level of the properties (wear resistance, hardness, local strength, bearing capacity, heat resistance, damping, antifriction, transport and other properties).

Magnesium matrix composites possess unique specific performance, various countries develop this trend [2-4]. Magnesium alloys with high tendency to vitrification and thermostability are of the particular interest as matrixes for the MMCs. Due to the satisfactory wetting and chemical stability in the magnesium melt, among the ceramic reinforcers SiC is used. Low oxygen solubility in magnesium allows to use oxides, and fine wetting of the metals by magnesium – refractory metal powders (Cu, Ni, Ti). When using transition metals in in-situ processes, the produced intermetallides provide high wear resistance and tribological properties of magnesium MMCs.

Nowadays, aluminum alloys are of the high demand and wide-spread among the non-ferrous metals materials, possessing high specific strength and a general-purpose complex of mechanical, service and special properties, creating preconditions for the use of aluminum-based materials to develop engine-building, aeronautical and space engineering.

The possibilities to enhance the service properties of aluminum alloys, produced by the traditional technology of component alloying, their treatment in the liquid state, and shaping, and used for the articles of various branches of engineering, are exhausted to a large degree. Due to this fact all technically developed countries started the research and development in the field of the alumomatrix composites (AMCs) synthesis.

Liquid phase technologies of AMCs production by incorporating a sufficient amount (up to 20 vol. %) of micrometric refractory particles into the melt have been brought to the industrial application (MC 21, Duralcan, Science Application Intern. Corp., Alcoa Inc. (USA), British Alcan, Talon Composites, Aerospace Metal Composites Ltd (Great Britain), Deutsche Edelstahlwerke GmbH (Germany), Alloytic Co. Ltd. (Korea), Hitachi Metals Ltd. (Japan), etc.).

The structure and properties of the composites are determined by the matrix melt properties, by the composition, form, dimensions, quantity of incorporated or formed in the melt disperse particles, and also by the interaction activity and processes on the “disperse particle-melt” boundary. These factors determine the possibility of producing of the metallic suspension, the pouring of the latter into the molding cavity (casting mold, molding tool) provides the production of the articles with the given properties. However, the role of the incorporated into the melt or formed in the melt particles differs considerably in various stages of the finished article production.

Microsize particles in the composites, in particular, alumomatrix composites, serve as reinforcers, and the character of interactions on the “particle-melt” boundary is mainly determined by their wettability by the melt. The research in the field of alumomatrix composites, reinforced with widely used microsize SiC, Al_2O_3 , B_4C , TiC and the experience of use of the articles proved that these alloys provide high level of mechanical and service properties and may be used to produce shaped articles with casting methods.

For example, Duralcan company, using the mechanical mixing of SiC microparticles into the melt, produces gravitation casting (F3S.20S (AA 359/SiC/20p) and high-pressure casting (F3N.20S (AA 360/SiC/20p) composites, working in normal and high temperatures, in corrosive medium.

Talbor company specializes on producing alumomatrix composites, which incorporate SiC, B_4C , Al_2O_3 microparticles in the process of mechanical mixing. In comparison with B_4C , silicon carbide is better wetted by the melt, provides high level of the composite hardness, more commercially appropriate, however, Al_4C_3 creates danger of brittle failure of the articles. The presence of adsorbed gases does not allow to weld and thermally treat the composite. Even if the matrix alloy can be welded, high SiC density with the same incorporated quantity leads to weighting of the articles, tends to the sedimentation. Al_2O_3 is the densest of the three reinforcers, minimally active towards aluminium, provides higher strength of the composites, but its segregation ability in the casting process is high. For the cast composites (sand mold casting and metal mold casting, high-pressure casting, liquid forging) the company uses B_4C as reinforcing and providing the composite with the ability to withhold hard radiation particles. With the density similar to the density of aluminium, B_4C provides higher sedimentation stability of the slurry, allowing of producing thick-section castings by the methods of gravitation casting, using ceramic filters, melt outgassing with argon or nitrogen, B_4C particle passivation, special construction of the gating system.

In a number of cases, in order to improve the wettability, the disperse particles are plated by the elements, enhancing their wettability by the melt (for example, technological coatings of Fe and Cr are used for SiC). For aluminum alloys, Mg, Sn, Sb, Bi serve as interphase-active elements, improving the wettability of the incorporated refractory particles.

To improve the wettability and homogeneity of disperse particle distribution in the melt, the two-stage process of the incorporation of these particles is employed: the overheated melt cools to a two-phase state, heated particles are incorporated into the melt when the melt is in the semi-solid state, in the mixing process the melt fuses completely and is poured into mold using the traditional casting methods. The mixing of the particles to the state of a semi-solid slurry, obtaining higher viscosity, facilitates the gas blanket surface destruction and improves the wettability of the particles by the melt [5].

New generations of military, aerospace and civil equipment require the development of the new structure and functional materials, obtaining properties, unattainable in the traditional structure materials and composites, reinforced with ultradisperse micrometric particles.

At present, special attention is drawn to the works dedicated to the production of the metal matrix nanocomposites (MMNCs) by means of the multipurpose use of ex-situ and in-situ processes (polyreinforcement), implying nanosize structural constituents in the form of the introduced from the outside nanosize thermostable particles and intermetallides, produced as a result of the occurring processes. All useful properties of the microsize composites in the nanocomposites are implemented at a higher level [6-10]. However, high energy potential, high specific surface and interphase particle energy, high surface tension of the melt impede the incorporation of the nanoparticles into the melt, and the tendency of the nanoparticles to develop aggregates, the possibility of their fusion in the melt, require special technology solutions, different from the technologies of the microsize compound production. Thermal activation of the incorporated particles in the process of their preparing to the incorporation into the melt, and the processes occurring in the liquid composite during the dwell time, pouring into the mold and finished article solidification, contribute to the intensification of the diffusion and recrystallization processes, to the disappearance of the non-equilibrium phases, residual stress relief and, correspondingly, to the modification of their unique properties. The higher is the technological temperature, the more intensively all these processes occur, the lower is the viscosity of the melt.

Most researches consider the nanoparticles, incorporated into the aluminum alloys, as modifiers. It is noted that SiC microparticles are mainly located at the boundaries of the eutectic grains, and SiC nanoparticles – in the dendrites of the primary solid solution. The tendency of the microparticles to locate at the boundaries of the grains leads to the lower fracture toughness, strength and hardness at high temperatures, and also to the machining deterioration. Nanosize SiC and Al_2O_3 particles affect the size and morphology of the intermetallic compounds formed in the melt, as a result of the interaction of the nanosize SiC and Ti particles, the reinforcing phase TiC is formed.

It is evident that irrespective of the influence of the nanoparticles on the crystallizing alloy, they remain as isolated inclusions in the solid composite, affect the processes of composite destruction, depending on the place of their location regarding the boundaries of the structure elements.

In the process of cast nanocomposite production it is necessary to provide the inoculation of the nanoparticles into the melt and their equal distribution in the volume of the slurry; sedimentation stability of the melt, prevent nanoparticles from aggregation in the process of feeding the composition into the molding cavity, and in the recycling process of the composites and reheating before the shaping of the previously produced nanocomposite charges. Concurrently, the problem of providing the peak level of the necessary complex of composite properties by affecting the processes of crystallization and structure formation of the matrix melt is of great value.

The inoculation of the nanoparticles into the melt and their uniform distribution in the slurry is mainly implemented by means of the mechanical and MHD *stirring* of the particles, and reinforcing particle powders may be introduced not only in the initial state using plasma torch, injection in the gas current, but also in the form of the pellets, briquettes, flux cored wire, extended pressed compositions. In the process of the mechanical mixing the optimum performance of the mixer provides the onset of the shear deformation in the melt, especially when mixing the melt in the liquid-solid state, prevents the agglomeration of the particles, provides better wetting and uniform distribution of the particles in the melt volume. The ensuing dwell time of the melt in the mixer with low mixing speed allows of its transporting to the casting molds using various casting methods.

Plasma synthesis method along with the biplanar MHD-mixing and incorporation of the nanoparticles into the melt in the form of nanosize powder composites, produced by mechanical alloying in high-energy mills [11] possesses a number of advantages. Biplanar MHD mixing of the wide-interval alloys, in particular, hypoeutectic silumins, in the liquid and two-phase states allows of solving the problem of equal distribution of the particles in the melt volume, ensuring the degeneration of the dendrite structure and the possibility to use all the advantages of thixocasting for producing nanocomposites [11-14].

High-power ultrasound treatment (UST) of the alloys in the liquid and two-phase state is one of the most efficient methods of effecting the processes of alloy structure formation [15-19]. The impact of ultrasound diminishes with increasing the distance between the alloy and the ultrasonic horn, therefore, it is advisable to use ultrasound treatment along with the MHD-mixing. This allows of solving the problem of non-dendrite structure formation in the process of composite thixocasting at the stage of the primary treatment of the melt, considerably decreasing the duration or excluding the reheating of the blank aimed at final “degeneration” of dendrites. The use of thixoforming in the process of cast products production out of the composites allows to cure the defects, characteristic to this type of product – gas porosity, occurring in the process of the mechanical mixing of the particles into the melt.

The problem of the particle inoculation into the melt may be simplified due to the preliminary production of rich nanocomposite alloys in the liquid-solid state with their further incorporation into the melts.

Parameter optimization of the complex process of the nanoparticle inoculation, their equal distribution the melt volume, of the external action on the liquid and crystallizing alloy, create the preconditions for the production of nanocomposite blanks, in which the total degeneration of the dendrite structure occurs in the process of primary crystallization. This, in its turn, allows to heat the cut-to-length sections only to the temperature of shaping.

The development in the field of production of MMNCs using as reinforcing complexes nanocarbon materials, in particular, fullerenes C_{60} , nanotubes, nanodiamonds, nanosize products of the modification of the natural carbonaceous rock (shungites), is highly prospective [20].

Special attention is given to the innovative development of cast and wrought MMNCs industry - the superdeep penetration (SDP) phenomenon. It may be regarded as the new physical instrument to affect the existing materials. The new concept of the physical phenomenon of the SDP is based on the consequent implementation of the complex of the physical effects, such as higher energy density (accumulation) in the local zones of the barrier material due to the shutdown sistem, creation of dynamically stable local zones of high pressure, the level of the latter sufficient to implement the dynamic phase transition. The use of SDP allows to incorporate into the volume of the solid body the alloying elements tens and

hundreds millimeters deep at an interval of 10^{-3} - 10^{-7} second. In the volume of the solid body the fibrous elements, obtaining specific nano- and microstructures, allowing of producing the materials with unique properties, are created [21-23]. Nowadays SDP is used for the solid-state processes, however, there is a reason to believe that complex technologies would make it possible to use SDP for the liquid phase methods of metallomatrix nanocomposite production.

As a rule, the foundry specialists are concerned with the production of the shaped castings out of the casting alloys by various casting methods. However, the development of the thixoforming (thixocasting, thixoforging) processes proves the effectiveness of the cooperation of the foundry specialists and the specialists in the field of the forging processes. The field of mutual cooperation may include the production of sheets and shapes out of nanocomposites using the methods of the ingotless rolling.

At present the granular technologies develop rapidly, especially the new material science branch of nanostructure granular composites, combining the advantages of the metallurgy of granules and the principles of producing the volumetric composite out of granules. As a rule, uniform (isostatic), hydrostatic or gas-static pressing is used for the compaction. Considering the experience of the foundry specialists in the field of suspension casting, the possibility of the use of the nanostructure granules for the inoculation of the nanoparticles into the melt, the cooperation of the foundry specialists with the specialists of the metallurgy of granules in the field of the development of the hybrid processes, capable of providing the production of the shapes out the new class of nanocomposites, is highly prospective.

2. CONCLUSION

1. The theory of nanostructured metal matrix, in particular, alumomatrix, composite alloys is in the making. It is necessary to define the role of the nanoparticles of various composition and origin in the processes of composite crystallization and recrystallization, in the processes of particles destruction in various in-use conditions. This requires, in particular, determination of thermal and physical characteristics of the composites and the development of the corresponding mathematical and physical models.

2. In the process of nanocomposite development it is necessary to ponder not only over the given complex of the mechanical and special properties, but also to consider as obligatory the constructional features of the specified blanks (parts), method of their production, special features of the mechanical and thermal treatment.

3. The problems of nanocomposite article production using liquid phase technologies require the system approach to the solving of the whole complex of the occurring problems, involving specialists in various fields (thermodynamics, physics and chemistry of the melt and solid state, fracture mechanics, technologies of the production and treatment of the alloys in the liquid and two-phase state, etc.)

4. The prospective technologies are the complex technologies of MMNC production, implementing the external influence (UST, MHD) on the liquid and crystallizing matrix alloy with incorporation of the reinforcing particles along with the thixoforming of the finished article production.

One of the present-day approaches to the development in this field is the adjustment of the characteristic features of the SDP processes to the liquid phase technologies of the MMNCs production.

References

- [1] Zheng Wu, Reddy, *Advanced Engineering Materials* 5(3) 2003) 167-173.
- [2] Hassan S. F., Gupta M., *J. Mater. Sci.* 37 (2002) 2467.
- [3] Lu L., Thong K. K., Gupta M., *Comp. Sci. Techn.* 63 (2003) 627.
- [4] Cai Y., Tana M. G., Shen G. J., Su H. Q., *J. Mater. Sci. Engng. A.* 282 (2000) 232.
- [5] Zhon W., Xu Z. M., *J. Mater. Proc. Techn.* 63 (1997) P.368.
- [6] Kalashnikov I. E., Bolotova L. K., Chernyshova T. A., *Tsvetnye Metally.* 9 (2009) 67-71.
- [7] Kondratenko A. N., Golubkova T. A., *Composite Materials Constructions* 1 (2009) 24-28.
- [8] Prusov E. S., Panfilov A. A., Kechin V. A., *Russian Foundryman.* 12 (2011) 35-40.
- [9] Koch C. C., *Nano structured materials: processing, properties, and potential applications'*, 423-526; 2002, Norwich, NY, Noyes Publications.
- [10] Schurack F., Borner I., Eckert J., Schultz L.: in «*Metastable, mechanically alloyed and nanocrystalline materials*», (ed. A. Calka and D. Wexler), 312-314; 1999, Zurich, Trans. Tech. Publ.
- [11] G. A. Kosnikov, V. A. Baranov, S. Y. Petrovich, A. V. Kalmykov, *Liteinoye proizvodstvo* 2 (2012) 4-9.
- [12] Szajnar J., Stawarz M., Wróbel T., Sebzda W., *J. of Achievements in Materials and Manufacturing Engineering* 34(1) (2009) 95-102.
- [13] S. Ji, Ma Qian, Z. Fan., *Metallurgical and Materials Transactions A.* 37A. (2006) 779-787.
- [14] Lilian Ivanchev, Siggibo Templeton Camagu, Gonasagren Govender. Semi-Solid High Pressure Die Casting of Metal Matrix Composites Produced by Liquid State Processing, *Journal Solid State Phenomena* (Volumes 192-193), Semi-Solid Processing of Alloys and Composites XII, pp. 61-65.
- [15] Abramov V. O, Abramov O. V., Artemiev V. V., Gradov O. M., Kolomeets N. P., Prihod'ko V. M., Eldarkhanov A. S., *High-power ultrasound in metallurgy and machine building*. Edited by Abramov O. V., Prihod'ko V. M., Moscow, Yanus-K., 2006, P.688.
- [16] G. A. Kosnikov, O. L. Figovsky, A. S. Eldarkhanov, V. H. Mezhidov, S. S. Yusupov, *Scientific Israel- Technological Advantages* 15(2) (2013) 93-96.
- [17] Eldarkhanov A. S., Nuradinov A. S., Gerikhanov A. K., Yusupov S. S. Low-frequency and ultrasonic treatment of steel in the process of ingot production and continuous casting. *International conference "Development of nanotechnologies: problems of international and regional science-and-training and science-and-production centres"*. AltGU, Barnaul. September 12-15, 2012. Pp. 188-190.
- [18] L. Zhang, D. G. Eskin, A. Miroux, L. Katgerman. Formation of microstructure in Al-Si alloys under ultrasonic melt treatment, *Light Metals* (2012), 999-1004.

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- [19] Yao Lei, Hao Hai, J. I. Shouhua, Fang Canfeng, Zhang Xingguo, *Trans. Nonferrous Met. Soc. China* 21 (2011) 1241-1246.
- [20] V. M. Prokhorov, V. D. Blank, G. I. Pivovarov, L. F. Solovyeva. Preparation of aluminium - fullerene alloys with nano-components and study of their mechanical properties. *Proc. Int., Sci. Conf. New promising materials and processes of their preparation* (NPM-2004). Volgograd, 20-23 Sept., 2004, pp. 124-126.
- [21] Oleg L. Figovsky, et al., *Journal of Technical Physics* 49(1) (2008) 3-25.
- [22] Figovsky O., Usherenko S., Usherenko Yu. *The creation of metal composite materials. The Physics and technics of high-energy processing of materials*. Dnepropetrovsk: The Art-Press, 2007. Pp. 218-235. (in Russian).
- [23] Usherenko S., Figovsky O., Usherenko Y., *Scientific Israel – technological advantages* 9(1,2) (2007) 28-32.

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