Formation of the Transition Zone "Matrix Alloy - Reinforcing Granules" in the Process of Solid-Liquid Synthesis of Composite Material

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Abstract: Casted composite materials have found certain application in mechanical engineering. Their advantages are lower cost due to the use of casting technology (solid-liquid synthesis), absence of any restriction on the shape and weight of parts. It is known that the transition zone between the matrix alloy and reinforcing phase plays an important role on the level of mechanical properties of cast composites. So, the aim of the study is investigations of transition zone formation during the infiltration of matrix alloy into the frame formed by the reinforcing phase. Copper based alloy was used as a matrix alloy and cast-iron granules were used as a reinforcing phase. Cast composite materials have been exposed for different time during the synthesis. Specimens were studied using electron microscopy. Results on the concentration distribution of main elements (Si, Fe, Cu) in a transition zone are presented. It is shown that the formation of transition zone starts during the infiltration process with further its growth during the exposition at the synthesis temperature. It is established that the formation of the transition zone takes place not only due to elements diffusion but as well due to the dissolution of granules of reinforcing phase. Experiments carried out allow establishing some peculiarities of the transition zone formation. To achieve required level of mechanical properties it is necessary to control the melt temperature and duration of the synthesis of cast composite materials.

Keywords: Synthesis of Casted Composite Materials, Transition Zone, Elements’ Distribution, Dissolution of Granules

1. Introduction

Composite materials are widely used in various fields of technology due to the unique combination of high physical, mechanical and operational properties [1-4]. In the production of composites, metal and polymer materials are used as a matrix, and powders (metals, oxides, intermetallics, etc.), fibers, particles of various shapes are used as a reinforcing phase [5]. Combining these components in various ratios, it is possible to obtain composite materials with required properties for specific operating conditions [6]. A wide variety of composite materials requires different technologies for the synthesis of materials with the given properties and structure [7].

Casting technologies (synthesis of liquid matrix material and solid reinforcing phase) are widely used to produce composite materials [8]. The advantage of casting technologies in comparison with powder metallurgy ones (pressing, sintering) is a lower cost and the possibility of manufacturing parts with various geometries and with practically no size and weight restrictions [9].

A large share in the production of cast composite materials is accounted for matrices based on aluminum, magnesium and copper alloys. The widespread use of aluminum and magnesium alloys is due to their low specific weight, which is extremely important for the aviation and space technology [10]. Copper and its alloys are characterized by high antifriction properties. However, the physical and mechanical
properties of copper are rather low, which limits its use in highly loaded friction units [11, 12]. It is known that the formation of transition zone between reinforcing particles and matrix alloy plays a considerable role in achieving required properties of cast composites [9]. But there is a lack of information concerning the formation of a transition zone and the change of its composition during the synthesis process. Therefore, the study of the transition zone formation during the synthesis process of a composite material based on a copper alloy applying casting technology is of scientific and practical interest for further production of antifriction parts with high operation properties.

2. Study of the Structure and Composition of the Transition Zone

2.1. Some Features of the Cast Composites’ Structure Formation

When choosing the composition of composite materials, both on metallic and non-metallic base, some general provisions should be followed, which can be formulated as: the material should best meet operational reliability, manufacturability and economy [13]. However, due to the heterogeneous nature of cast composite materials, certain problems arise during their synthesis and processing, resulting in specific defects for these materials. Moreover, properties of cast composite materials cannot be determined only by the properties of the components, without taking into account their interaction during the synthesis process [14].

Wetting of reinforcing elements with a melt determines their interaction during the synthesis process and subsequent exposure. The fundamental issue of creating cast composite materials is the possibility of controlling the process of physicochemical interaction at the “matrix - hardening phase” boundary. Without solving this problem, it is impossible to count on an achieving the required level of physical, mechanical and operational properties. It should be noted that the processes at the interphase boundaries are determined by the structure of the substance, which requires a detailed study of these processes to determine methods that prevent the development of an undesirable interaction of composite components with the formation of an unpredictable phase composition [15].

So, one of the main conditions for the formation of cast composite materials is the wetting of reinforcing elements with matrix melt and filling pores, capillaries and voids formed by reinforcing granules (particles). Various options for reinforcing composite materials and various types of components of the reinforcing phase form channels of various shapes and lengths through which the melt flows (infiltrates). In such conditions, the representation of pores (voids, channels) in the form of a system of communicating cylindrical or even capillaries is unacceptable for a number of reasons. Firstly, these are significant differences in real areas (sections, perimeters), as well as differences in wetted perimeters. Secondly, there are significant differences that arise when determining the local velocities of liquids, local pressures, the development of boundary layers and their break points. Thirdly, the interaction of jets (flows) is different in these conditions, first of all for the case of the development of constrained and reverse flows, especially in the sharp corners of the channels.

The wetting of the hardening elements by the melt leads to the close phase contact, which is a necessary condition for the development of a chemical interaction processes at the interface. It is considered mandatory to have a diffusion (transition) zone for a strong connection of two metals [16]. This is due to the fact that in the presence of only mechanical coupling, it is difficult to ensure a high joint strength. The result of purely mechanical coupling is the appearance of cracks in the material under high loads. In addition, a feature of the synthesis of composite materials using casting technologies is the dissolution of the elements of the hardening phase when interacting with a molten matrix alloy. This interaction can be quite long when the mold with a composite material is keeping for degassing the matrix alloy.

The process of atoms transition of the solid component elements into the liquid phase can be considered as a process leading to a decrease in its content in the solid component or as a process leading to its increase in the liquid component. The interaction of solid and liquid components, in most cases, is accompanied by the transition into the liquid phase of particles, molecules, ions that make up the solid component. This process can be considered as an increase in the contact surface between the elements of solid and liquid components when the duration of interaction increases. Then the ratio of the changing surface sizes of the solid component over time to its maximum possible concentration in the liquid phase will express the kinetics of the dissolution process. The process of dissolution of granules during their interaction with the melt can be described by the equation of A. N. Shchukarev:

\[
dM/dt = KS(C^L_0 - C^L) \quad (1)
\]

where \(dM/dt\) is the dissolution rate; \(C^L_0\) is the maximum saturation concentration of the liquid phase over the solid component; \(C^L\) is the current concentration of the solution; \(S\) is the contact surface; \(K\) is the constant of the dissolution rate.

To find the rate of dissolution of the hardening phase during the infiltration of the melt and subsequent exposure, it is necessary to determine the values of \(K\) and \(C^L\).

The task of present investigations was to study the formation of a transition zone in bronze-based composite materials reinforced with cast iron granules and obtained by casting technology. As well, it was intended to determine the value of \(C^L\) for a particular alloy at a certain point in time.

2.2. Experimental Methods

The studies were carried out with composite materials based on bronze containing silicon and manganese, and reinforced with cast iron granules. The use of cast iron granules allows you to additionally control the structure and properties of the material due to various quenching modes.
after the infiltration process as the cast iron structure is very sensible to a quenching rate. This type of composites has been successfully used to normalize the thermal and mechanical state of steam turbines. Parts made of the developed composite materials were used for installation under the surfaces of bearing housings and in dowel units. A feature of the work of these units is high specific pressure at very low velocity, which does not allow the use of conventional lubricants [17].

The synthesis process of the composite material was carried out at a temperature of 1080°C. Molds with samples containing a frame of cast iron granules were infiltrated with molten bronze. Immediately after the infiltration was completed, the first samples were quenched in water to fix the microstructure. The remaining samples were placed in an oven at the given temperature and extracted at intervals of 20 minutes and also quenched in a water. The maximum exposure time of the samples in an oven was 100 minutes.

The scanning electron microscope JSM-5610LV with the chemical analysis system JED-2201, JEOL was used for microstructure study and chemical composition analysis of components. The images were obtained using a back-reflected electron detector (BES), which allows to obtain the maximum phase contrast of the image.

The chemical composition was determined at various cross-section points, starting from a point located in the granule at a distance of 50 µm from the surface (point 1). Then measurements were carried out every 25 µm to a point located in the matrix at a distance of 50 µm from the transition zone (point 6). Points 4 and 5 represent the transition zone.

2.3. Investigation of Specimens' Structure

Figure 1 shows the microstructure of a single granule of the reinforcing phase immediately after the completion of the infiltration process (Figure 1a) and after holding for 100 minutes (Figure 1b). The analysis of microstructures shows that the formation of the transition zone occurs already during the infiltration (the process of filling out the form).

The melt flow regime and its change are related to the kinematic structure of the flow and its stability. The change in the overall configuration of the flow can be influenced by its narrowing, expansion, bending of the axis, change in the cross-section profile. The loss of stability of the flow can be caused by a change in the boundary conditions of the flow, in particular, the appearance of bends in the velocity profile, which will cause a change in the cross-section of the flow. If small disturbances are damped in a laminar flow, then its kinematic structure is constant. The constancy of the kinematic structure of the turbulent flow is determined by the constancy of the size and intensity of changes in the velocity profile.

In the case of a sharp change in the shape of the channel surfaces, the flow velocity profiles change. Such a change leads to a local pressure change, which causes the formation of areas filled with vortices of different sizes. The structure of the flow, the intensity of vortex formation and the size of the vortices are determined by the flow regime. This behavior of the melt flow contributes to the interaction of
granules with the melt already at the stage of filling the mold.

The destruction of the surface layer is visible due to the diffusion of copper into the granule, causing a violation of its continuity. As a result, the dissolution of the surface layer takes place. The thickness of the transition zone increases, which is confirmed by an increase in the content of iron and silicon in it. At the same time, the formation of intermetallic compounds was not seen.

The effect of the exposure time on the change in the composition of the composite material is confirmed by the spectra at point 1 (Figure 2) for the moment of completion of the infiltration process (Figure 2a) and after exposure at the synthesis temperature for 100 minutes (Figure 2b).

Changes in the microstructure of the composite material during the exposure make it possible to approximately determine the values of the $K$ which is included in the equation (1). The $K$ value is determined based on changes in the size and surface of the granules, as well as the exposure time. The calculations carried out gave the value $K \approx 2.238 \times 10^{-9}$ kg/(s·cm$^2$). Values of $C_0^\ell$ are determined from the state diagram, and the current concentration is determined from measurements made using an electron microscope. Calculations show that the granules lose about 35% of their mass during the exposure time of 100 minutes after the completion of the infiltration process, which negatively affects the level of physical and mechanical properties of the composite material.

### 2.4. Elements Distribution in the Transition Zone

The main elements of the composite material are copper, iron, silicon and manganese.

Since the concentration of manganese changed slightly in cross-section over the time, the results of changes in the concentrations only for silicon, iron and copper in the transition zone are given. Moreover, the fact of a high concentration of silicon at the "transition zone – matrix" boundary was unexpected (Figure 3). The maximum amount of silicon (5.14%) was fixed immediately after the completion of the infiltration process. With further time exposure, silicon was redistributed, but even after 100 minutes of exposure, the concentration of silicon in the transition zone (3.7%) was higher than in the granules and matrix.

![Figure 3. Silicon distribution at different exposure times.](image)

The copper content in the transition zone was highest at the end of the infiltration process (7.24%), which decreased to 5.55% after the exposure for 100 min. At the interface of the "transition zone – matrix" the copper content decreased from 36.37% to 13.02%. (Figure 4) The analysis showed an
increase in the copper content in the granule at point 1 (at a distance of 50 microns from the surface) with the increase in the holding time from 3.3 to 4.11%. The increase in the copper content indicates the diffusion of copper into the granule. As a result, more fusible compounds are formed in the granule, which lead to its faster dissolution in the matrix melt.

A different situation was observed for iron. During the exposure of samples at the synthesis temperature, the iron content in the center of the transition zone increased from 85.78% to 91.24%. The iron content at the interface of the “transition zone–matrix” also increased from 56.5% to 80.28% (Figure 5).

An increase in the iron content in the transition zone indicates the dissolution of the granule and the expansion of the transition zone, which is confirmed by a significant decrease in the concentration of copper in the transition zone (Figure 4). A significant increase in the iron content in the transition zone confirms that the main factor is played by the dissolution process of granules, not the diffusion one.

The distribution of iron confirms the formation of the transition zone during the infiltration of the matrix alloy. To reduce the thickness of the transition zone during the synthesis of the composite material, it is necessary to reduce the infiltration time. This can be achieved, for example, by applying the excessive pressure on the matrix melt during the infiltration process.

Knowing the changes in the concentration of the element in the solution at certain points in time, it is possible to determine the rate of the granule dissolution in time for the calculated value of the dissolution constant. As well, it is possible to calculate the time required for the formation of a transition zone, knowing the rate of dissolution and taking into account the diffusion process.

As well one can calculate the time when granules lose less than 2% of their mass. In this case, a strong connection between granules of the reinforcing phase and the matrix alloy is formed. The deviation from the predicted physical-mechanical properties does not exceed 3%.

3. Conclusion

The studies carried out allowed us to determine some features of the transition zone formation between the granules of the reinforcing phase and the matrix alloy based on a bronze. A change in the nature of the distribution of the main elements in the transition zone after the exposure at the synthesis temperature has been established. The analysis showed that in order to achieve the predicted mechanical properties, it is necessary to observe the temperature and time parameters of the synthesis of cast composite alloys as accurately as possible. The holding time after the infiltration required for a melt degassing should not exceed 10 minutes. In this case, the physical and mechanical properties of the composites will correspond to the predicted level.

![Figure 5. Iron distribution at different exposure times.](image-url)

References


