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Phases created during diffusion bonding of aluminium and aluminium bronze chips

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Materials

ABSTRACT

Purpose: The main aim of the paper is to apply the new sintering criterion to determinate the sintering conditions and to investigate the phases created during bonding aluminium and aluminium bronze and their effect on the composites properties.
Design/methodology/approach: An original concept of producing a composite, consisting in the mixing and bonding of aluminium chips with aluminium bronze chips through press moulding and extrusion, has been developed. As a result of the reciprocal diffusion of copper and aluminium during extrusion and heat treatment applied just after extrusion a creation of hard phases leading to an increase of wear resistant takes place.
Findings: Heat treatment applied after extrusion improves tribological properties and hardness of composites by increase of diffusion bonding and creation of new phases. To obtain the bonding of particles separated by a layer of oxide and a highly compacted product, two conditions must be fulfilled: thin aluminium oxide and large shear plastic deformation
Practical implications: The proposed new sintering criterion allows to determined condition of sintering and deformation in order to obtain good product. This criterion can be applied in FEM.
Originality/value: Diffusion bonding process of aluminium and aluminium bronze chips leads to creation of phases typical for Cu-Al alloys. The bonding takes place during extrusion of cold compacted mixture of aluminium and aluminium bronze chips and during heat treatment applied after extrusion. In this way, without participation of metallurgical processes good bearing materials can be manufactured.
Keywords: Aluminium; Bonding; Phases; Chips; Sintering

1. Introduction

A technology which has been developed systematically in recent years is that of manufacturing composites with predetermined properties. Among the different production possibilities of such materials, one of them is manufacturing composites from waste products. During the recycling of the waste by remelting a lot of the metal is lost as a result of oxidation, especially in the case of aluminum and its alloys, and the costs of labour and energy as well as the expenditures of environment protection are very high. Thus the great interest has been shown in chips recycling processes other then remelting [1].

So different ways of waste products recycling, consisting in the direct conversion of waste into compact metal by granulation, remoulding and hot extrusion or hot forging, where melting is eliminated, were elaborated. This kind of recycling can be applied not only to aluminum [2-5] and its alloys but also to iron, copper and, to some extent, to cast iron [6-9].

In the metallurgical process no more than 54% of the aluminum and aluminum-alloy chips is recovered. In the case of the direct conversion of the same metals chips into compact metal by extrusion ultimately 95% of the metal is recovered [10-12].

The benefits of the direct conversion of aluminum and aluminum-alloy scrap into compact metal include also a possible reduction in the funds spend on the labour, energy and environment protection as a result of the reduced consumption of ores and energy carriers, and less degradation of the natural environment because of reduced air-pollution emission. Flow charts of the conventional recycling of aluminum and aluminum-alloy chips and of their direct conversion are shown in fig. 1.

Factors that contribute significantly the bonding of aluminum and aluminum-alloy chips contain: degree of granulation of the aluminum and aluminum-alloy chips, remolding parameters, stress and strain states in consolidation processes, temperature and rate of consolidation processes, lubrication method and the lubricants used. In case of composites with additional introduced reinforcing phases the amount, form and size of these phases are also very important. For good bonding of granulated chips the large plastic deformation is needed. Such deformation can be obtained in extrusion process with extrusion ratio, at least 4.

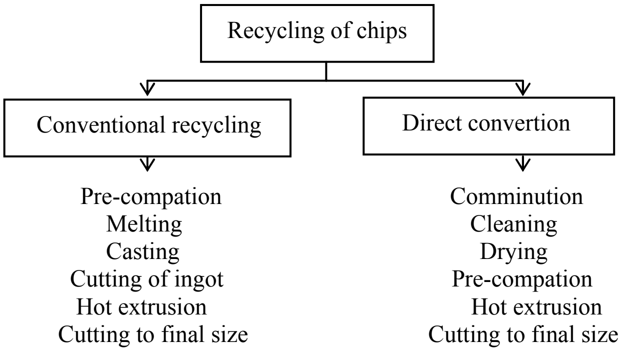

Fig. 1. Flow charts of the conventional recycling and direct conversion of chips [1]

The direct conversion recycling process was applied to aluminum and its alloy chips without reinforcing phases [2-4] and with the following reinforcing phases: tungsten, carbon, silicon carbide, ferro-chromium and aluminum oxide [5,13,14]. On the one hand reinforced phases decrease free movement of dislocations and increase the yield and the tensile strength but on the other hand increase the porosity and reduce these properties as well.

In recent years, researches have been aimed at producing composite materials based on aluminum for the manufacturing of bearings [15-17]. Such composites are conventionally produced from aluminum powders with silicon, silicon-carbide and graphite additions [18].

An original concept of producing such a composite, consisting in the mixing and bonding of aluminum chips with aluminum bronze chips through press moulding and extrusion, has been developed [19]. The mechanical properties of composites can be correlated to phase transformation in Al-Cu equilibrium diagram, similar like properties of aluminum bronzes [20]. The particle dispersion has distinct effect on the process kinetics [21] and conditions of sintering and extrusion can be analysed by using deformation processing map [22, 23].

The main aim of the paper is to investigate the new created phases and their effect on the composite properties.

2. Sintering criterion

The elaboration of the best method of chips recycling was based on sintering criterion. The sintering criterion [24] was created at following assumptions:

- contribution of clear surface of particles which are exposed during working processes as a result of surface layer brittle fracture to whole particle surface should be as much as possible,
- normal stresses acting on the clear surface of particles should bring them together on the atomic distances.

The fracture of oxide surface layer and atomic contact of clear surface take place in such area where exist the biggest tensile strain and compression stress. So it can be stated that the greater tensile strain the lower normal stresses needed for good junction of particles. The greater compression stress the easier way that metallic bonding could be created. So the sintering criterion can be dependent on both factors in the following form:

$$dW_s = f(\sigma_n, d\varepsilon_1) \tag{1}$$

where: dW_s – sintering indicator characterising the local quality of particles junction,

$d\varepsilon_1$ – increment of largest tensile strain,

σ_n – largest compression stress normal to the direction of largest tensile strain

For isotropic materials there is consistence of principal stress and strain directions and the normal compression stress σ_n is equal to the largest principal compression stress σ_3.

Taking into account that the greater outspread of native surface the lower value of stresses is needed for good sintering, the indicator characterising the local quality of sintering of particles can be described by the product of both factors as follows.

$$dW_s = \sigma_3 d\varepsilon_1 \tag{2}$$

Junction of the particles in the whole considered volume take place when sintering indicator W_s obtain the some critical value C_{cr}:

$$W_s = \int_0^{\varepsilon_1} \sigma_3 d\varepsilon_1 = C_{cr} \tag{3}$$

For axial symmetric metal forming processes of incompressible materials component of strain state ε_1 can be expresses by other component of strain state ε_3

$$d\varepsilon_1 = -2d\varepsilon_2 = -2d\varepsilon_3 \tag{4}$$

Taking into account that sintered materials especially during manufacturing do not keep incompressible condition the relation (4) should be expressed by:

$$d\varepsilon_1 = -2\frac{1}{\beta}d\varepsilon_3 \tag{5}$$

where: β – compressible coefficient, Combining equations (3) with (5) the following relation is obtained:

$$W_s = \int_0^{\varepsilon_3} \frac{2}{\beta} \sigma_3 d\varepsilon_3 = C_{cr} \tag{6}$$

It means that for sintering of particles the defined work of largest principal compression stress on the largest suitable displacement is needed. For application of such criterion the critical value of C_{cr}, which secures good junction of particles has to be known. This parameter is dependent on compressible coefficient β, which can be determined on the base of changes of density of composites before and after sintering.

The parameter C_{cr} can be determined experimentally or analytically. Experimentally determination of the parameter is very labour-consuming and strenuous, so much easier is to apply into analysis of sintering in metal working processes the theoretically calculated values of C_{cr} parameter.

Assuming that n times increase of particles surface is enough to exposure of native structure, as a results of fracture of brittle surface layer, the following consideration can be performed. If the analysis is restricted to surface of particles parallel to the largest elongation (more complex particles are considered in the paper [26]), e.g. in extrusion process to the extrusion direction, it can be easy stated that between expansion of surface of individual particle and degree of deformation the following relation exists:

$$S_p = \sqrt{R_p} \tag{7}$$

where: S_p – degree of expansion of the surface of individual particle given by following relation,

$$S_p = S_{pf} / S_{po} \tag{8}$$

where: S_{po}, S_{pf} – initial and final surface of particle respectively,
R_p – reduction of cross section of individual particle given by relation,

$$R_p = d_{po}^{\ 2} / d_{pf}^{\ 2} \tag{9}$$

where: d_{po}, d_{pf} - initial and final substituted diameters of particle.

Extending the relation (7) on the whole volume of extruded composite the compressible coefficient has to be included. Then the following relation is obtained:

$$S = \beta \sqrt{R} \tag{10}$$

where: R – reduction of cross section of ingot,
S - ratio of initial surface of ingot to final surface of product:

For surface expansion S = n the limit strain needed for good junction of particles is given by relation;

$$\varepsilon_l = \ln \frac{d_o}{d_f} = \ln \sqrt{R} = \ln \frac{S}{\beta} = \ln \frac{n}{\beta} \tag{11}$$

From that relation it can be stated that the lower compressibility of composites the smaller deformation is needed to obtain a good junction of particles.

The critical values of plastic work needed to obtain good junction of particles can be calculated from the following relation:

$$C_{cr} = \int_0^{\varepsilon_l} \frac{1}{\beta} \sigma_p d\varepsilon_{\mathrm{int}} = W_s \tag{12}$$

For known stress – strain of the composite materials the calculation of critical values of the plastic work by solution of above given equation can be performed (Fig.2).

Additional condition of good junction of particles is uniformity of plastic flow during extrusion, because in other way the heterogeneity of deformation can lead to surface layers fracture caused by tensile stress.

In some cases, presintering was employed to set in motion the diffusion transport of matter between the aluminum and aluminum-alloy particles and the hardening phases. Because of the relatively small number of contact bridges between particles and the intensive oxidation of their surfaces, the extent of diffusion transport during sintering, which would be free of large plastic strains, could be very limited. Therefore hot extrusion was employed as the final operation.

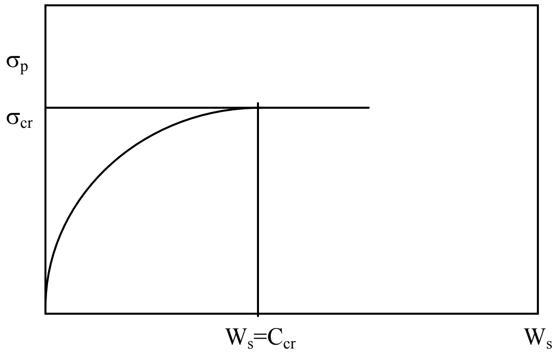

Fig. 2. Relation between flow stress and plastic work needed to manufacturing of composite

3. Materials and processing

Sintering criterion indicates that for good metallic bonding the chips during manufacturing of the composites should be subjected to large plastic deformation, so the matrix of composite should be made from ductile material. In the industry a big amount of aluminum and its alloys chips as waste materials are produced. So applied of direct conversion method in such cases was a very valuable achievement.

An original concept of producing a composite, consisting in the mixing and bonding of aluminum chips with aluminum bronze chips through press moulding and extrusion, has been developed. As a result of the reciprocal diffusion of copper and aluminum during extrusion and heat treatment applied just after extrusion a creation of hard phases leading to an increase of wear resistant takes place.

As the starting materials the granulated chips of aluminum of two fractions, i.e. below 2 mm and 2-4 mm were mixture with different amount of reinforcing phases: 15, 22, 30 and 45%. For the reinforcing phase the aluminum bronze containing 8% of aluminum was chosen. High-power ball mill with a horizontal axis of rotation filled with 20 mm-diameter steel balls up to 45% of its volume was used for the mixing of granulated aluminum chips with particles of reinforcing phase. The mixtures were subjected to compacting, sintering and hot extrusion. The cold compacting was performed in a device with a floating die under the constant pressure of 400 MPa. The sintering was employed in vacuum of 10^{-3} HPa during 1 hours. Then hot extrusion was applied as the final operation to bring about the diffusion bonding between particles by crushing the layer of oxides and actuating diffusion processes under a high pressure and temperature. Hot extrusion was carried on in the temperature range of 500-525^{0}C. As lubricant was used mixture of zinc stearynian with graphite.

The additional operation - heat treatment was applied at 545 ^{0}C for a different time, changing in the range of 0,5 – 10 hours. Using optical and scanning electron microscopes and image analysis system VISILOG-4 the structure was investigated.

4. Results and discussion

Elements distribution along the strait line perpendicular to aluminum matrix - aluminum bronze particle boundary shows that diffusion in extruded composites is very small (Fig. 3).

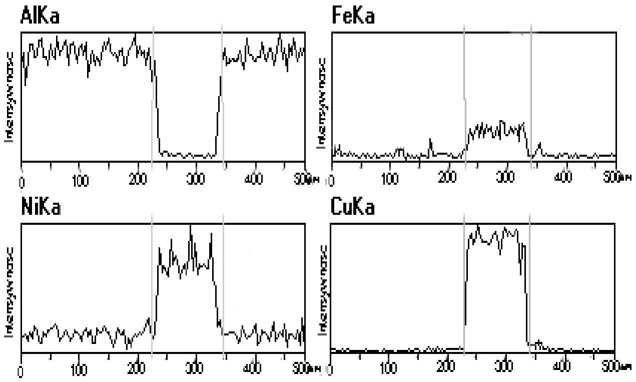

Fig. 3. Linear distribution of elements at grain boundary before heat treatment of composite containing 22 % of aluminum bronze

After heat treatment the distinct diffusion of elements is observed, and typical structures at the primary boundary between matrix and reinforcing phase are shown in the Fig.4

The phase's identification performed by diffraction method in Ferrous Institute in Gliwice shows, that during heat treatment following new phases were created: γ_1, δ, ξ_2, η_2 and θ. All the phases are typical for phases in Cu-Al equilibrium diagram [25]. As a result of diffusion, the aluminum matrix of composites was slightly enriched in copper, and CuAl8 reinforcing phase in aluminum also. In this way the typical structure of bearing materials was obtained, it means the large, hard, load-carrying particles created from phases γ_1, δ, ξ_2, η_2 and θ distributed in soft matrix. A structure like this cannot be obtained in the case of solid aluminum alloys.

Typical distribution of element concentrations at the aluminum matrix-reinforcing particle boundary after heat treatment is shown in Fig.5.

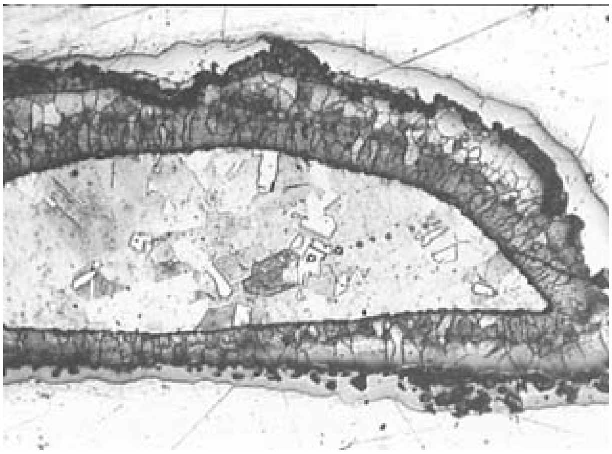

Fig. 4. Structure of composite containing 22% of aluminum bronze after annealing at 545^{0}C during 7 hours

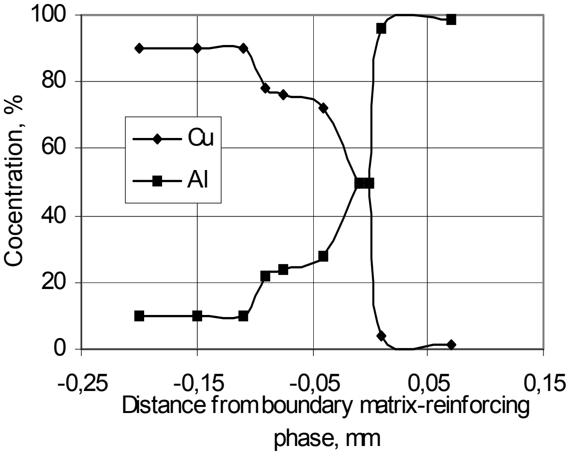

Fig. 5. Linear distribution of elements at grain boundary after 7 hours annealing at 545^{0}C of composite containing 22 % of aluminum bronze

From the figure one can notices that the good diffusion bonding between matrix and reinforcing phase were obtained. The distribution of elements is dependent on the time of annealing. With longer time the diffusion of copper reaches deeper layer of matrix.

This way, without participation of metallurgical processes good bearing materials can be manufactured.

The effect of CuAl8 reinforcing phase content on the wear intensity of composites is shown on Fig.6.

From the figure it can be seen that the wear intensity decreases rapidly with content of reinforcing phase to about 15 %.

The further increase of reinforcing phase content until 30 % caused negligible decrease of wear intensity. In the range of 30-40 % of reinforcing phase content the wear intensity is nearly constant. Such changes of wear intensity is observed for fine (below 2 mm) and coarse (2-4 mm) reinforcing phases. In the whole range of reinforcing phase content the wear intensity is lower for coarse phase.

The differences of mechanical properties observed in tensile test (Fig.7) and compression test (Fig.8) indicate on the weak bonding between particles of matrix and matrix and reinforcing phase. The weak bonding could deteriorate the wear of composites, because small particles could be pull out from the surface by collaborating sliding element. To improve the bonding the heat treatment of composites after extrusion was applied.

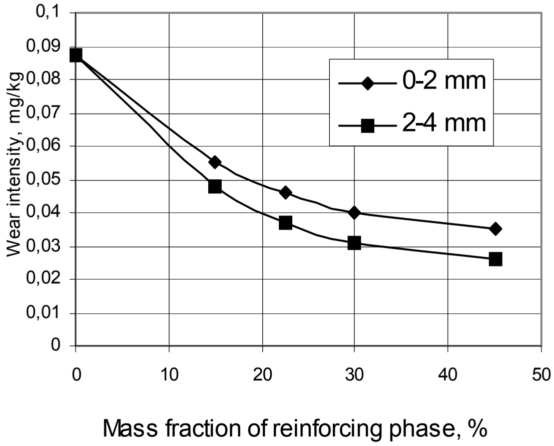

Fig. 6. The wear intensity as a function of mass fraction of reinforcing phase

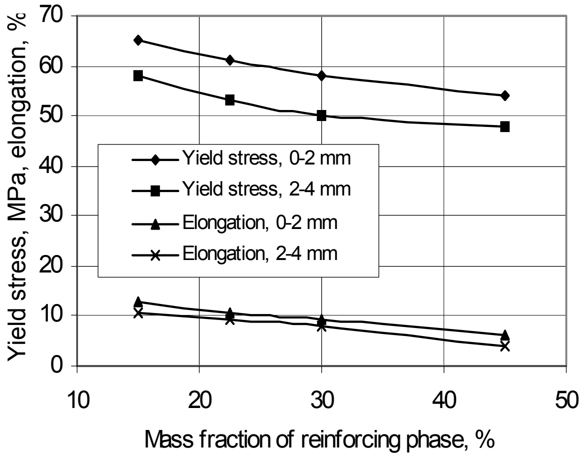

Fig. 7. Tensile yield stress and elongation as a function of reinforcing phase content

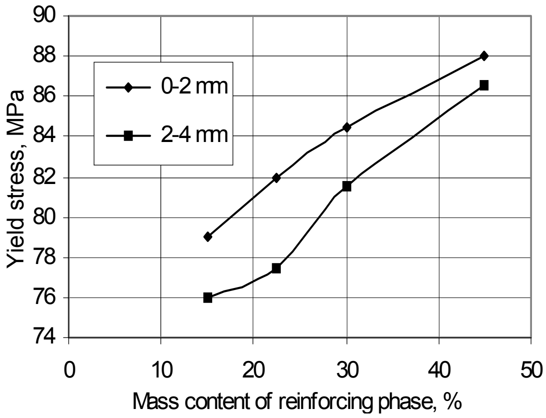

Fig. 8. Compression yield stress as a function of reinforcing phase content

The effect of annealing time on the mass wear and friction coefficient of composites is shown on Fig. 9.

From the figures it can be seen that the mass wear of composites and friction coefficient decreases with time of annealing. The distinct decrease of friction coefficient is observed with increase of annealing time over 4 hours, but the decrease of mass wear in the whole applied time is nearly constant. Increase of fractional reduction has very small effect on the wear of composites.

Heat treatment has positive effect on mechanical properties of composites determined in tensile as well as in compression tests.

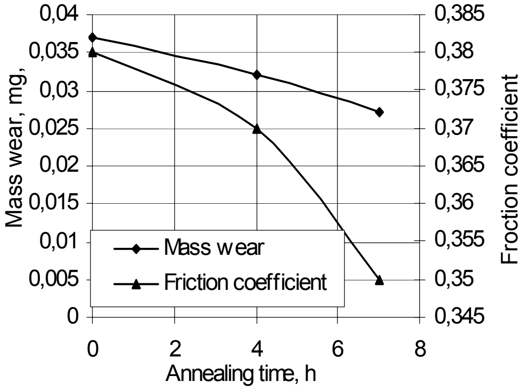

Fig. 9. The mass wear and friction coefficient of composites with coarse reinforcing particles extruded with 16 fractional reduction as a function of annealing time

5. Conclusions

On the base of presented investigation of manufacturing composites from granulated aluminum and CuAl8 aluminum bronze chips it has been conclude that:

- by hot extrusion of cold compacted mixture of aluminum and aluminum bronze chips and by heat treatment applied after extrusion the hard phases are created,

- hard phases have positive effect on the tribological properties of composites,
- the diffusion bonding takes place during extrusion of cold compacted mixture of aluminum and aluminum bronze chips and during heat treatment applied after extrusion.
- new method of manufacturing bearing composites from aluminum and aluminum bronze chips without metallurgical process was elaborated,
- heat treatment applied after extrusion improves tribological properties and hardness of composites by increase of diffusion bonding and creation of new phases,
- to obtain the bonding of particles separated by a layer of oxide and a highly compacted product, two conditions must be fulfilled: thin aluminum oxide and large shear plastic deformation

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