RECYCLING OF CARBON CONTAINING ALUMINUM MATRIX COMPOSITES VIA THEIR REMELTING

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Abstract

Nowadays aluminum alloys are strong enough to have good mechanical properties. What is their weak point is stiffness (Young modulus) that cannot be significantly improved be standard alloying elements. Carbon or carbon containing substances like silicon carbide with high values of Young modulus are often used in aluminum matrix composites with intention to increase stiffness of the resulting material. However, carbon bearing aluminum matrix composites are not easily recycled via melting because of reactivity of molten aluminum with carbon or carbides. Direct remelting of Al-SiC composite resulted in silicon containing melt and aluminum carbide while oxide envelop of input material keeps original shape to as high temperature as 850 °C. Addition of fluxes helps to break oxide layers but it does not prevent aluminum from reaction with carbon to form aluminum carbides.

Keywords: metal matrix composites (MMC), recycling, melting, carbides, carbon, aluminum

1 Introduction

So called hard aluminum alloys have good mechanical properties from a point of view of their strength. Ultimate tensile strength (UTS) of aluminum-zinc alloys, e.g. EN AW 7075 T6 exceeds 500 MPa while UTS of special Al-Sc alloys is approaching 1000 MPa [1]. On the other hand, their stiffness is not improved significantly, e.g. EN AW 4032 with over 10% of silicon has Young modulus of 76.8 GPa, ca 10% higher than pure aluminum [2].

In order to increase stiffness of aluminum alloys significantly, addition of particles with very high Young modulus is needed. Silicon carbide is often used as such reinforcement material with elasticity modulus of ca 410 MPa [2]. Another option is to use high strength high stiff carbon fibers, e.g. Torayca M40 with tensile modulus of 392 GPa [3]. Young modulus of such aluminum matrix composites is increased – depending on amount of particles – well over 100 GPa [4].

On the other hand, carbon bearing aluminum matrix composites are not easily recyclable. Direct remelting of composite scraps seems to be most simple way. However, molten aluminum reacts easily with carbon or carbide particles forming aluminum carbide. In order to avoid formation of Al_4C_3 Kamaravan et al. have used ionic melt to recover aluminum via electrolysis of $AlCl_3$ at as low temperature as 103 °C [5]. Disadvantage of above process is relatively low purity of recovered aluminum 98.15% with 1.26 wt.% of copper as main impurity. It should be mentioned however that copper is alloying element of used aluminum alloy matrix. Also energy consumption of 3.2-6.7 kWh per kg of recovered aluminum is not negligible. Therefore Yang et

al. [6] prefer reusing of MMC instead of their remelting. Ravi et al. [7] were successful in using NaCl-KCl flux to separate melt from carbide particles. They claim degradation of SiC particles to such extend as to be unusable for their reuse.

Even more troubling as recycling of aluminum based MMC scrap is recycling of fine wastes like swarfs or saw dusts. Mashadi et al. recommend to cold press fine particles and melt them with flux [8]. This method is facing to the same troubles with interaction of molten aluminum and carbon/carbide particles as that of scrap treatment. Neither direct injection of aluminum swarfs resolves above mentioned problems, although this method is one of the cheapest [9]. An alternative method for production of aluminum matrix composites via plastic deformation of aluminum chips have recommended Gronostajski and Matuszak [10].

Claims of some authors about possibility to reuse of AMC/SiC composites via repeating remelting sounds too optimistic [11-15]. They observed interaction of SiC particles with molten aluminum, however they regard them still functional.

In order to clarify contradictory literature data, the direct remelting of AMC/SiC composite was tested. The intention was to use as simple and as cheap method as possible. Therefore direct remelting on air without any flux has been carried out.

2 Thermodynamic calculations

Results of thermodynamic calculations for temperature range 400-900 °C are shown in **Table 1**. It is seen a discontinuous change of standard enthalpy at a temperature over 650 °C that is caused by phase change – aluminum gets into molten state. Equilibrium constant of reaction (1) shows that formation of pure silicon is not favorable. However, silicon is readily dissolved in aluminum with rather low solubility below eutectic temperature - **Fig. 1**. Therefore activity of silicon dissolved in aluminum should be regarded as non-standard (less than 1). In that case, equilibrium of reaction (1) is shifted towards products of reaction.

$$4Al + 3SiC = Al_4C_3 + 3Si \tag{1.}$$

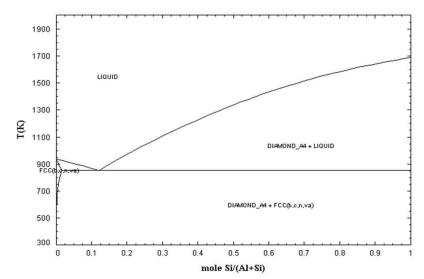


Fig. 1 FactSage calculated phase binary diagram of Al-Si system [16]

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equilibrium constant, logarithm of equilibrium constant					
T [°C]	ΔH [kJ]	∆S [kJ]	∆G [kJ]	K	Log(K)
400	7.809	-18.986	20.589	2.525E-02	-1.598
450	7.389	-19.586	21.553	2.774E-02	-1.557
500	6.850	-20.307	22.550	2.995E-02	-1.524
550	6.172	-21.155	23.586	3.186E-02	-1.497
600	5.340	-22.136	24.668	3.343E-02	-1.476
650	4.334	-23.255	25.802	3.467E-02	-1.460
700	-39.286	-69.983	28.817	2.838E-02	-1.547
750	-39.914	-70.612	32.333	2.235E-02	-1.651
800	-40.490	-71.162	35.877	1.793E-02	-1.746
850	-41.020	-71.644	39.448	1.463E-02	-1.835
900	-41.493	-72.057	43.041	1.212E-02	-1.917

Table 1 Thermodynamic calculations of reaction (1), columns from left to right: temperature, standard enthalpy change, standard entropy change, standard Gibbs energy change, equilibrium constant, logarithm of equilibrium constant

3 Experimental materials and methods

The AMC/SiC composite in a shape of short round bars (ca 100 mm) with diameter of 9 mm were put into stainless steel crucible and heated on air for 1, 2 and 4 hours at 650 and 850 °C. Crucible was covered by BN layer inside to prevent interaction of molten aluminum with crucible walls. Aluminum matrix EN AW 6061 alloy and spherical SiC particles with equivalent diameter of 9 microns were used to extrude AMC/SiC 20 vol.% composite – see **Fig. 2**.

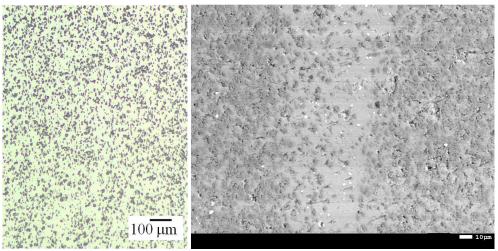


Fig. 2 Microstructure of AMC/SiC 20 vol.% composite with spherical 9 micron SiC particles

4 Result and discussion

The result of melting of AMC/SiC composite on air is shown in **Fig. 3**. Left picture shows material after heating to 650 °C for 4 hours. It is seen that material is still compact although it became very fragile – easily to be broken by hand. No melt was observed on the bottom of crucible. Right picture shows material after heating to 850 °C for 4 hours. Material still keeps shape similar to original input material. It is also very brittle and huge holes are seen parallel to long axis.

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Fig. 3 Samples of AMC/SiC composite after heating on air for 4 hours at a) 650 °C (left) and b) 850 °C (right)

While temperature of 650 °C is close to melting point of aluminum, temperature of 850 °C used for another heating is well above it. Aluminum is not completely molten in case of temperature of 650 °C, so interaction between matrix and SiC particles is limited to the interface only. This causes brittleness of material, Al/SiC interface is not strong and elastic enough to provide material with sufficient tensile properties.

Temperature of 850 °C is high enough to melt aluminum that interact aggressively with SiC interface, dissolving silicon in itself leaving remaining aluminum and aluminum carbide. An Al-Si melt was observed on a bottom of crucible although most of material is kept by oxides in a shape similar to original round bars.

5 Conclusion

Although process of remelting of AMC/SiC composites can be improved by adding of flux in order to brake of oxide envelope and separate molten aluminum with silicon from carbide containing phase(s), this is not promising way of recycling of carbon bearing aluminum matrix composites. Ceramic particles are completely degraded, carbide phases need to be treated in addition operation and resulting Al-Si alloy is low grade recovered material. An additional effort should be directed towards reuse of material without melting, possibly by re-extrusion of AMC/SiC scraps.

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