TEM INVESTIGATION OF MATRIX - SAFFIL™ FIBER INTERFACES IN ALUMINIUM ALLOYS BASED COMPOSITES

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Abstract
The microstructure of metal matrix composites of AA2024, AA 6061 and EN AC 44300 alloys with 10 vol. % Saffil fibers were investigated using transmission electron microscopy. The fibers in the preform were stabilized using liquid glass binder and fired at 800°C resulting in formation of amorphous silicon oxide joints. The preforms infiltration with AA2024 leads to reaction of liquid metal with SiO_2 binder substituting it with fine-crystalline mixture of MgO, $\Theta-Al_2Cu$ and silicon. Similar operation with AA6061 leads to binder substitution with porous –amorphous Al_2O_3 and MgO. Only squeeze casting of EN AC 44200 with Mg<0.6 % left most of the binder unchanged and allowed to obtain practically clean Saffil/ matrix interfaces.

Keywords: metal matrix composites, TEM, Saffil fibers

1 Introduction

The aluminium alloys found already wide application in automotive industry for production of engine blocks, piston heads and others, but stronger and stiffer materials would be very much welcomed. The experiments with reinforcing of high strength aluminium alloys, like AA6061, 2024, 7075 or AlMgCuAg with up to 20 vol. % of Al_2O_3 Saffil™ fibers showed roughly 20% of UTS increase already at ambient temperatures [1-4]. On the other hand, a composite based on AC 8B casting alloys just reproduced the matrix maximum hardness [2]. Simultaneously, both groups of composites retain their ambient temperature strength by ~100°C higher, than their matrixes. The reason of different response of matrixes to the same reinforcing Saffil™ fibers, as well as further optimization of properties of these composites, could be obtained by finding the detail nature of metal/ceramic interfaces in these materials.

The problems with production of such composites were overcome partly by pressure infiltration and partly by wetting enhancement with alloying additions, like magnesium, routinely introduced into commercial aluminium alloys. However, switching from the Al – Al_2O_3 system in to AlMg – Al_2O_3 causes that reactive infiltration takes over. In that case, at the metal/ceramic interface an MgO oxide and at 4<Mg<8 wt. % an even more brittle $MgAl_2O_4$ spinel may form [1]. The situation in that area is worsened by abundance of oxygen from SiO_2 binder easily dissolved in contact with liquid metal containing magnesium.

The observations of Al4Cu1Mg0.5Ag matrix with Saffil™ fibers bonded with SiO_2 showed, that in multi-component alloy the $MgAl_2O_4$ may form along the MgO at very low magnesium additions (~1 wt.%) [3,5]. Similarly, the $MgAl_2O_4$ was found on β-Al_2O_3 particles strengthening Al1Mg0.6Si0.3Cu (AA6262) alloy [6]. The type of phases formed at the Saffil™ fiber/matrix interface are important, but their microstructure, as well as presence of voids easily nucleated during such phase transformation might be even more detrimental on the properties of such composites. However, information about that problem is limited.

This project was aimed at investigation of the type and microstructure of phases formed at the Saffil™/ metal matrix interface in a series of aluminium alloys used in car industry applications.

2 Material and Experimental Methods

Performs made of Safill™ fibers (10 vol. %) were stabilized with liquid glass (hydrated SiO_2), dried and fired at 800°C. Next, they were squeeze cast with an AA2024, 6061 or EN AC 44200 alloys (**Table I**). Finally, composite materials were solution heat treated and aged to peak hardness.

The microstructure was investigated using Tecnai SuperTWIN FEG 200kV transmission electron microscope with EDAX microanalysis system. Thin foils were cut using Quanta 3D focused ion beam equipped with Omniprobe lift-out attachment.

Table 1 Additions (wt.%) to alloys used for infiltrating the preforms

Alloys	Mg	Si	Cu	Mn	Fe	other
AA2024	1,2-1,8	0,4-0,6	3,8-4,9	0,3-0,9	<0,5	<0,1 [1)]
AA6061	0,8-1,2	0,4-0,8	0,1-0,4	<0,15	<0,7	<0,4 [2)]
EN AC 44200	0,2-0,6	6,5-7,5	<0,1	<0,3	<0,5	<0,2 [3)]

[1)] Cr <0,1, Ti <0,15, Zn <0,25, [2)] Cr 0,04-0,35, Ti <0,15, Zn <0,25, [3)] Ni<0,1, Zn<0,1, Pb<0,1,

3 Results

Immersing of Saffil™ fibers into liquid glass covers them with thin layer of binder forming bridges between individual fibers during subsequent forming and firing (**Fig.1a**). The firing of such preform at high temperature fixes the bridges into permanent form and allows retaining the preform shape during squeeze casting with liquid aluminium alloy. The microstructure observations showed, that rough fiber surface is well covered with the binder, which solidifies in an amorphous form (**Fig. 1b**).

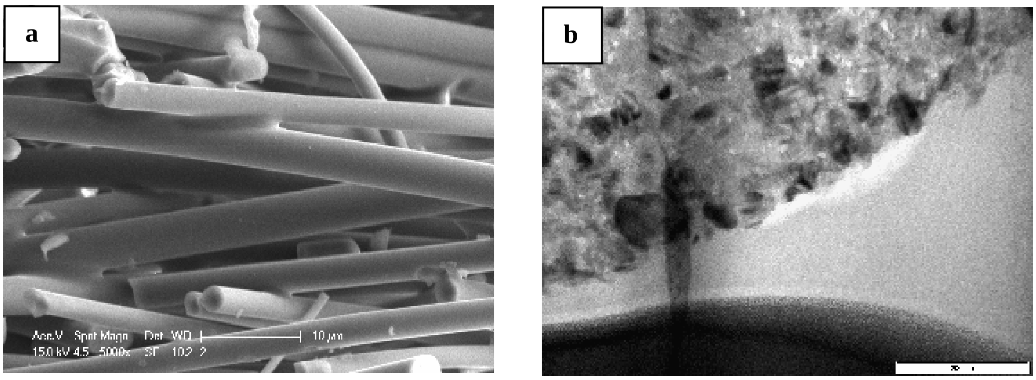

Fig.1 Scanning (Secondary Electrons) image of preform after firing process (a) and transmission image of SiO_2 bridge in-between adjacent Saffil™ fibers

The squeeze casting of such preform with AA2024 alloy caused reaction of liquid metal with the binder substituting it with dense nano-crystalline material (**Fig.2**). The local chemical analysis using EDS system indicated, that at areas with bigger amount of binder, i.e. so called bridges the surface of the Saffil™ fibers covers copper-aluminium crystallites, i.e. probably $\Theta{-}Al_2Cu$ phase (**Fig. 3**). The centre of such post-binder area is filled with the mixture of the $\Theta{-}Al_2Cu$ and silicon crystallites immersed in remnants of SiO_2 binder phase, while at their outskirts the MgO oxide forms. However, outside the joints, i.e. where Suffil™ were covered with thinner layer of binder the latter was totally dissolved and fine crystallites of the MgO precipitated directly on the fibers surface.

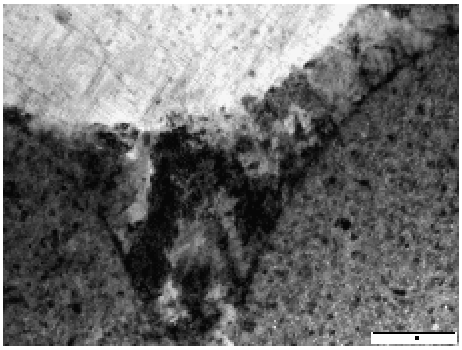

Fig.2 Transmission image of products of reaction between SiO_2 binder and liquid AA2024 alloy

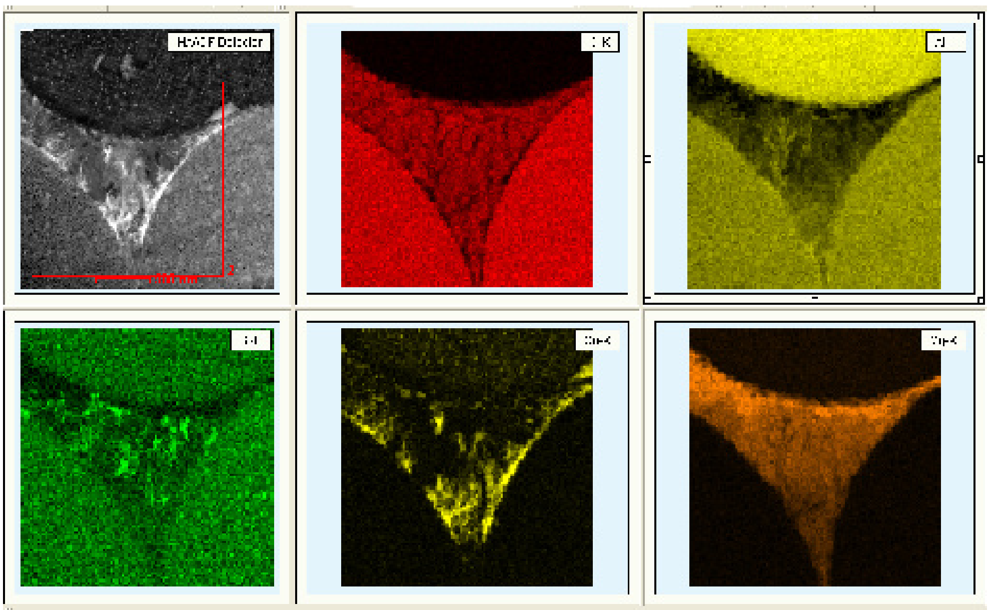

Fig.3 Scanning-transmission (HAADF) image and series of maps presenting distribution of O, Al, Si, Cu and Mg obtain from area of reaction between SiO_2 binder and liquid AA2024

The infiltration of preform with AA6061 alloy resulted in substitution of SiO_2 binder with an amorphous highly porous material (**Fig. 4**). Aside of porosity, larger voids at the Saffil™ /metal matrix were also observed. The maps presenting local chemical composition helped to determine, that the amorphous material consist of alumina with traces of silicon oxide (**Fig.5**). Small amount of SiO_2 present inside Al_2O_3 fiber was introduced into them already at their production stage, i.e. the RF grade contain up to 4 at. % Si preventing grain growth. The manganese oxide was located in small amount and only at the border of amorphous areas and matrix. The copper containing Θ–Al_2Cu type precipitates are usually nucleated on these amorphous areas like the one visible in the centre of HAADF image in Fig.5, or directly on Saffil™ surface.

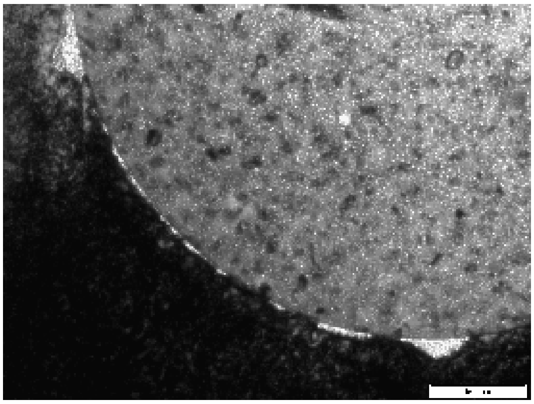

Fig.4 Transmission image of products of reaction between SiO_2 binder and liquid AA6061 alloy

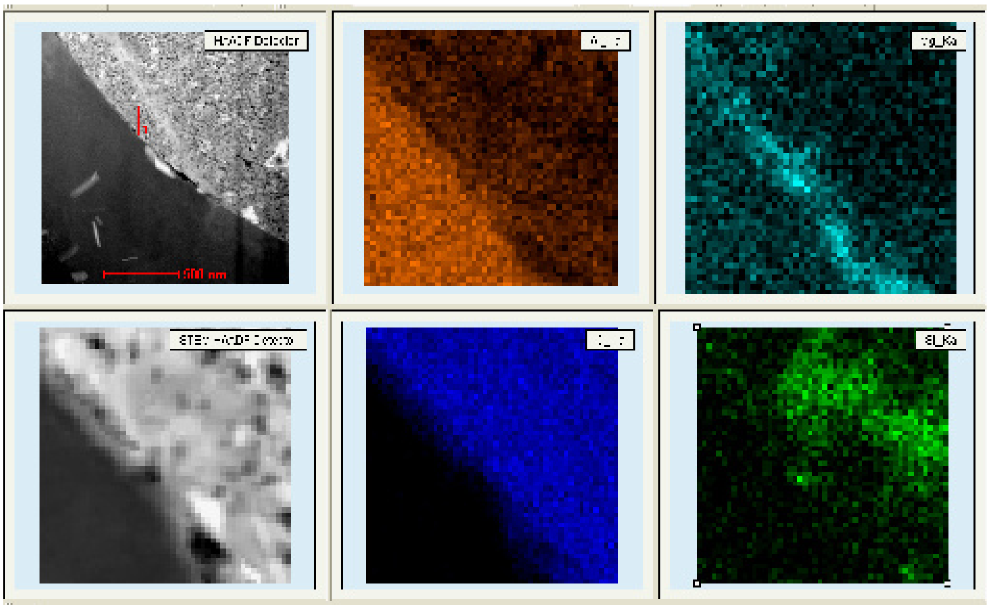

Fig.5 Scanning-transmission (HAADF) image and series of maps presenting distribution of O, Al, Si and Mg obtain from area of reaction between SiO_2 binder and liquid AA6061

The same experiments were also performed with EN AC 44200 alloy. In that case, most of the binder bridges withstand contact with liquid metal preserving connections in-between Saffil™

fibers (**Fig. 6**). More detailed observations of the binder showed that it contain small petal-type voids, which may indicate either start of dissolution of amorphous SiO_2 or just represent the effect of sample preparation with high energetic Ga+ beam. The precipitation of silicon was noted both within matrix and at the fiber surface – some of it even within the binder area. The maps of local chemical composition showed, that aside from bridges between fibers, the a thin layer of binder is uniformly covering the fibers surface (**Fig. 7**).

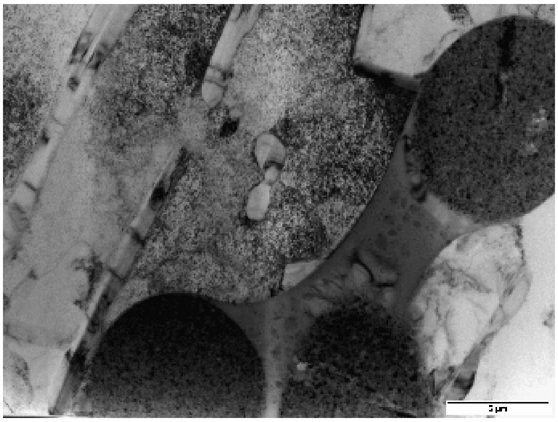

Fig.6 Transmission image of SiO_2 binder bridges between Saffil™ in perform force infiltrated with AC 44200 alloy

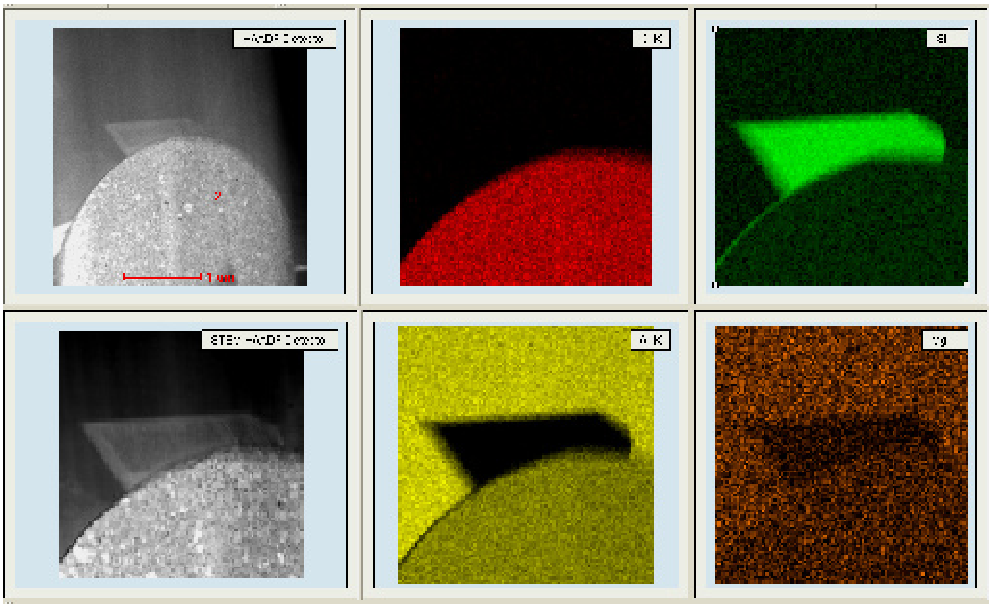

Fig.7 Scanning-transmission (HAADF) image and series of maps presenting distribution of O, Al, Si and Mg obtain from Saffil™/ AC 44200 matrix interface

4 Summary

The squeeze casting of ceramic preforms made of Saffil™ fibers using liquid AA2024, 6061 or EN AC44200 alloys produced dense metal matrix composites. However, an improvement in

mechanical properties of these composites could be gained only after ascertaining a proper connection between these high strength (~2 GPa) nano-crystalline slightly porous (5 – 10 %) of 3 -5 μm diameter Al_2O_3 Saffil™ fibers and their matrix. The Vaucher and Beffort [1] indicate, that a proper bonding between Al_2O_3 fibers and aluminum alloy depends to large extent on amount of magnesium additions deciding on type of oxide phases, like MgO or $MgAl_2O_4$ formed as a consequence of the production process. They pointed also, that presence of the spinel forming from 4<Mg< 8 wt. % content is the most detrimental for composite mechanical properties. Even, as this amount is higher than usual magnesium additions in commercial alloys, though presence of SiO_2 bonding phase might cause nucleation of $MgAl_2O_4$ also in case of such alloys, i.e. like Al1Mg2Cu0.5Ag [3] or Al12SiCuMgNi [5]. Present experiments with AA2024 and AA6061 alloys containing 1.6 and 1 wt. % Mg respectively showed however only presence of MgO precipitates. The EN AC44200 alloy with even lower magnesium was free from any magnesium oxides. The observed differences between literature and experiment are probably connected with varying squeeze casting conditions, like infiltration time, pressure and other.
The lack of $MgAl_2O_4$ spinel at Saffil™ fiber/matrix interface, generally considered as a necessary condition for obtaining proper composite strength improvement, is not an only requirement. The microcrystalline mixture of Θ-Al_2Cu, Si and MgO material found in Saffil™/AA2024 composite has close similarity to the one with spinel obtained after infiltration Saffil™ with Al4Cu1Mg0.5Ag [3]. Even, as such a mixed microstructure seems of dubious value for load transfer, though the measurement of the latter indeed showed significant strength increase. However, the porous mixture of amorphous Al_2O_3 and MgO substituting bonding phase in Saffil™/AA6061 composite definitely stand no chance at all at keeping fiber-matrix together. What is more, the voids also present in this area might serve, as good crack initiators. In this situation, the Saffil™/AC44200 composite with preserved amorphous SiO_2 bonding phase at fiber surface and therefore with clean interfaces should present the highest relative property improvement, i.e. as compared with its matrix.
The performed microstructure observation aimed at characterization of Saffil™ fiber/ aluminum matrix interfaces clearly indicated, that the composite mechanical properties are strongly depended not only on type of phase formed over there, but also on defects like porosity or voids. The decreasing magnesium content in the AA2024, 6061 or EN AC44200 alloys used as matrixes in investigated composites resulted in diminishing or even eliminating formation of MgO at fiber interfaces. However, the above alloys differ also in amount of other alloying additions and consequently in melting temperature, so the magnesium content was just one of parameters influencing above changes at Saffil™/matrix interfaces.

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