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Development of pore-free Ti-Al-C MAX/Al-Si MMC composite materials manufactured by squeeze casting infiltration

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ABSTRACT

An innovative method of manufacturing of Ti-Al-C MAX/Al-Si MMC composite materials was developed using squeeze casting infiltration of open-porous MAX phase preforms. Self-propagating High-temperature Synthesis (SHS) in microwave-assisted mode was applied for the creation of preforms in the Ti-Al-C system, that were subsequently infiltrated with Al-Si alloy to create dense composite materials. Microstructure and phase composition, structural defects and potential impacts between constituents of manufactured composites were characterized by the means of SEM and TEM microscopies and XRD analysis. No undesired reaction at the interface was observed, but TiC inclusions were identified in the material. Among the mechanical properties, the instrumental Young's modulus and Vickers hardness were established. The hardness and the elastic modulus of the matrix were enhanced 4 to 5 times. Wear behaviour was tested with a "pin-on-flat" method with the reciprocating motion for different load values (0.1, 0.2 and 0.5 MPa) for tool steel counterpart. Wear resistance of the composite material (WR = 1.6–2.3 × 10^{-4} mm^3/Nm) was twofold higher than for the sole matrix (WR = 3.5–4.8 × 10^{-4} mm^3/Nm). The developed manufacturing method allows the effective fabrication of pore-free MAX phase based MMC composite materials, with significantly higher wear resistance than the widely applied Al-Si alloys.

1. Introduction

MAX phases – also called machinable ceramics – consist of a mixture of mostly hexagonal ternary carbides or nitrides, which allows them to combine the advantages of ceramic and metallic materials. To date, approximately seventy MAX phases have been synthesized. Originally, most of the interest was directed at Ti_3SiC_2-based ceramics [1], but later even more attention was paid to titanium aluminium carbides (Ti_3AlC_2 and Ti_2AlC), which are the most lightweight and oxidation-resistant of the MAX phases [2]. Compared with the others they are characterized by high thermal and electrical conductivity, high strength and impact resistance, and low coefficient of thermal expansion. Last but not least, Ti_3AlC_2 is the only MAX type phase exhibiting increased plasticity at higher temperatures [3].

Composites reinforced by three-dimensional ceramic skeletons have shown promise as candidates for wear-resistant applications, such as bushing or brake materials. During friction, a stiff ceramic network carries most of the load, limiting plastic deformation of the matrix [4]. However, the above can be achieved only as long as all of the fine channels separating the intricate branches of the ceramic skeleton are fully infiltrated with liquid metal. Therefore, fabrication of such composite materials requires a pressure-assisted method to fill all available space with the liquid metal, overcoming its poor wettability and simultaneously improving the crucial matrix-reinforcement bonding [5].

Self-propagating High-temperature Synthesis (SHS) is an efficient method of manufacturing MAX phase-based materials, but it usually results in a highly porous product. This fact opens the way for infiltration with a metal and the obtaining of metal matrix composites (MMC). Yeh et al. [6] presented a study concerning the *in situ* fabrication of composite materials based on Ti_2AlC or Ti_3AlC_2 and Al_2O_3, but the obtained materials were lacking in density. Only slightly better results were obtained with a Pressure-Less Melt Infiltration (PLMI)

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technique used for the manufacture of Ti_2AlC/TiC/Mg composites by Anasori et al. [7,8]. Based on a spontaneous flow of molten metal through a porous Ti_2AlC preform, the technique leads to the creation of many imperfections such as misruns and similar. Nevertheless, the tensile strength was improved to 420 MPa, and ultimate compressive strength to 1028 MPa.

Another attempt to produce such an MMC was undertaken by combining SHS with pressure melt infiltration (PMI) [9]. In that case, the porous Ti_2AlC preform was subjected to pressure infiltration with liquid Al alloy under 28–35 MPa for 20–22 s, but still large parts of the preform remained non-infiltrated. Hu et al. [10] proposed another method named Current Activated Pressure Assisted Infiltration in which Ti_2AlC powder was first sintered with a space holder material producing easily penetrated channels, and next infiltrated with AA6061 alloy. Although the specific strength of the composite was 1.5 times higher than that of the matrix, metal-free cavities were still identified in the material. Although almost 97% of the open porosities were filled, the closed ones were still present in the material. The above indicates that, even though progress in this area is evident, a method for producing ceramic skeletons allowing full metal infiltration is needed to produce materials with even better properties than those obtained to date.

This paper presents a fast and reproducible method of fabrication of Al-Si MMC composite based on squeeze casting infiltration. The novelty in the subject of MAX type phases is the use of Microwave Assisted Self-propagating High-temperature Synthesis (MASHS) for the creation of open-porous MAX phase preforms in the Ti-Al-C system. This process, being significantly shorter, cheaper and more energy-efficient, can be used instead of the conventional powder metallurgy methods. Next, the preforms were infiltrated by Al-Si alloy to form a pore-free composite. The infiltration techniques of porous MAX phase preforms described so far in the literature require a lot of time and energy and do not provide a sufficient degree of filling of open porosities, what can be solved by the use of squeeze casting infiltration. The beneficial influence of MAX phase-based reinforcement on mechanical properties of the Al-Si matrix such as wear resistance, hardness and instrumental Young's modulus was then tested.

2. Materials and Methods

Commercial powders of Ti (99.5% Ti, −325), Al (99.9% Al, −325, Alfa Aesar) and graphite (99.5% C, −325, SGL Carbon Ltd) were used in the molar ratios 2:1:1 to prepare a stoichiometric reactant mixture and fabricate Ti_2AlC and Ti_3AlC_2. First, the Ti, Al and C powders were mixed with ZrO_2 balls for 10 min, or 3 h in the case of mechanical activation, with BPR 10:1. Subsequently, the powders were uniaxially cold-pressed into cylindrical samples in the shape of pellets with 22 mm diameter under a pressure of 900 MPa for 10 s. The prepared samples were subjected to a Microwave-Assisted Self-propagating High-temperature Synthesis (MASHS) process. The microwave magnetron power was applied in the range 300–400 W. The whole reaction took place under the flow of Ar atmosphere. The temperature was controlled by a Raytek Marathon MM pyrometer with measuring spot dia. 0.6 mm. Other processing details were presented elsewhere [11].

The prepared MAX skeletons were subsequently used for pressure infiltration – squeeze casting with an Al-Si eutectic alloy, AC 44200 (10–13.5% Si, 0.4% Fe, 0.05% Cu, 0.4% Mn, 0.1% Zn, 0.15% Ti, remainder Al). Molten metal (770 °C) was pressurized (90 MPa) onto the ceramic skeleton, preheated to 750 °C, for 1 min. The obtained composite was characterized with a Hitachi S-3400 N scanning microscope equipped with SwiftED3000 chemical analyser (Hitachi, Japan) and Ultima IV X-ray diffractometer (Rigaku, Japan, 40 kV/40 mA). Microstructures were analyzed by a Hitachi H-800 transmission microscope (150 kV) (Hitachi, Japan). Discs for TEM observations were ground down to a thickness of 5 μm with the Gatan Dimple Grinder and finally polished with a Gatan DuoMil ion polisher. The samples were also analyzed in scanning (STEM) mode with the convergent beam with the use of Transmission Analytical Electron Microscopy using a JEOL 2100 Electron Microscope operated at 200 KV and equipped with JEOL JED-2300 T EDS unit for the analytical measurements (JEOL, Japan).

The wear resistance tests were carried out by a “pin-on-flat” method with the reciprocating motion of average speed 0.3 m/s for different load values (0.1, 0.2, 0.4 and 0.5 MPa). The flat counterpart was made of CT70 tool steel with hardness 67 HRC and roughness Ra = 0.4–0.6. Before the test, both of the tribosurfaces were degreased with acetone. Each test was repeated minimum three times. From the difference in heights the wear volume was calculated. Other mechanical properties, such as instrumental hardness and Young's modulus, were established with the use of a Nanoindenter NHT (Anton Paar, Austria) by the Oliver–Pharr method [12].

3. Results and Discussion

3.1. Microwave-assisted Self-propagating High-temperature Synthesis

SHS synthesis was carried out for a range of process parameters. As an example, curves obtained for samples produced from green compact compressed under 930 MPa and 465 MPa and those subjected to mechanical activation in a ball mill (3 h, BPR 10:1, ZrO_2 balls) are presented in Fig. 1. This shows that the applied force does not affect the combustion temperature, while the mechanical activation decreases it significantly. In the case of mechanical activation of the powders, the obtained preform was less porous, contrary to the aim of the planned experiment. Therefore, the conditions selected for further experiments consisted of regular mixing and compaction at 930 MPa. Based on the thermal-derivative analysis, it could be assumed that in area A the temperature fluctuation corresponds to a peritectic reaction between Ti_2AlC and TiC at 1570 °C and to a much lesser extent to solidification of residual amounts of titanium (possibly saturated with aluminium). Further, during cooling, the small peak observed on the first derivative curve (area B) represents a thermal effect that may be related to the transformation of sub-stoichiometric titanium carbide TiCx in the 1000–1200 °C temperature range.

The SHS synthesis of the reactant pellet causes its expansion, which can be explained by the solid–liquid reaction mechanism. Firstly the metallic particles forming Al-Ti melt with solid Ti cores, then TiC is formed at 900–1200 °C in a strongly exothermic reaction causing an additional temperature jump, similar to that found in the experiments of Hashimoto et al. [13]. When the combustion temperature increases above ~1660 °C even the titanium cores are melted. Subsequently, most of the TiC dissolves in the Al-Ti liquid forming an Al-Ti-C liquid phase. At such high temperature Al may partially evaporate, and when the temperature decreases some Ti and C solidify to form TiC again [14]. During the cooling process, below 1300 °C, the Ti_2AlC and Ti_3AlC_2 MAX phases are precipitated from the solution of TiC and Al-Ti melt in a

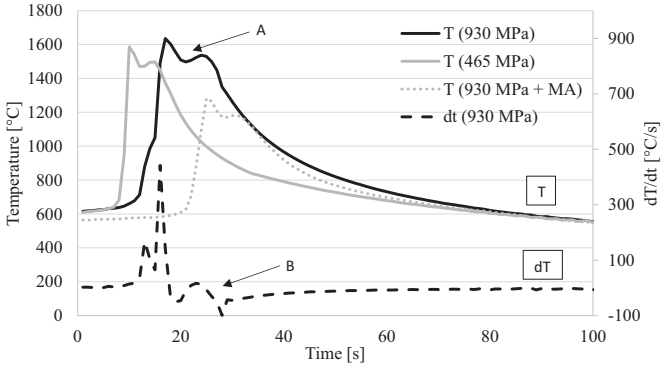

Fig. 1. Temperature-time dependences of SHS synthesis in Ti-Al-C system for different compaction pressures applied during uniaxial pressing of green compacts, MA – Mechanical Activation.