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Metal-matrix interpenetrating phase composites produced by squeeze casting①

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[Abstract] On the basis of the proposition and manufacture of a new type of metal-matrix interpenetrating phase composites (MMIPCs) by vacuum high-pressure infiltration, squeeze casting method was chosen for further study on this new type of MMIPCs. By employing the highly porous ceramic perform made from SHS reaction of Al-TiO_2-C system, squeeze casting process was studied in detail. By means of OM, SEM and TEM, the obtained highly porous SHS reaction products and the resulting MMIPCs for further understanding were closely examined and analyzed.

[Key words] squeeze casting; MMIPCs; MMCs
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1 INTRODUCTION

Recently, a new kind of composite structure has been prepared of which both the matrix and reinforcement phase are continuous and 3-dimensionally interpenetrating through the microstructure[1~3]. This kind of structure is sharply different from the traditional composite materials that usually possess microstructures combining a continuous phase (matrix) with one or more discrete reinforcement phases, shaped as long fibers, short fibers, whiskers or particles, etc[2]. And the interpenetrating phase composites (IPCs) is interesting since by retaining the continuity of each constituent phase it raises the possibility of developing materials having truly multifunctional characteristics with each phase contributing its own properties to the macroscopic properties of the composites. Examples of such systems include Corning's Vycora™ glass[2], the Lanxide Corporation's DIOMOX™ material[4].

A substantial amount of work has been done on the production characterization and modeling of IPCs focusing on the ceramic matrix composites reinforced with continuous metallic phases[5~8]. To explore this kind of novel structure in the case of metal matrix composites (MMCs), a new type of metal matrix interpenetrating phase composite (MMIPC) was formally proposed breaking a new path in the research field of MMCs[3]. At the same time we have brought forward a way to make this type of MMIPC by making use of highly porous SHS reaction products combined with vacuum pressure infiltration in our previous work[1]. This process is potential, however, the comparatively low infiltration pressure in our earlier experiment limits its potential. In this article, based on the same highly SHS porous products and aiming at elevating the infiltrating pressure thus improved the microstructure and performance of the obtained MMIPC, squeeze casting method is employed in the infiltration process for further exploration and deeper understanding of this new type of MMIPC. Better experimental results are gained and the obtained composites are analyzed in detail by means of OM, SEM and TEM.

2 EXPERIMENTAL

2.1 Self-propagating high temperature synthesis

As described elsewhere, by employing the SHS reaction: $4Al + 3TiO_2 + 3C \longrightarrow 2Al_2O_3 + 3TiC$ and with the addition of volatile agents, the highly porous SHS reaction products Al_2O_3-TiC were made having an interconnected open porosity of 83%. It would be used as the infiltration preforms in the following squeeze casting.

2.2 Squeeze casting

Squeeze casting was employed to intrude the molten metal into the interconnected channels within the obtained highly porous SHS reaction products. The advantages of squeeze casting are that infiltration is achieved under relatively short times, preventing extensive metal/ceramic reactions, and subsequent solidification takes place under externally applied pressure resulting in a fine-grained microstructure free of shrinkage voids. To accomplish this intrusion, the porous ceramics preform was preheated to 800 ℃ and

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then placed in a preheated steel die. A molten 2024 Al alloy, held in a separate crucible at 800 ℃, was then poured onto the preheated preform. A plunger was then activated by hydraulic pressure to force the molten metal into the channels at 70 MPa.

2. 3 Three-point bending strength

The three-point bending strengths of infiltrated samples and the Al alloy matrix in the same condition were measured on as-cast specimens (three for each mean value, approximately 3 mm × 6 mm × 30 mm) using a support span of 25 mm at a constant cross-head speed (0. 5 mm/ min) on a universal testing machine at room temperature. Polished and fractured surfaces were analyzed using optical microscope and scanning electron microscopy. Transmission electron microscopy specimens were ground to a thickness of approximately 100 μm, then mechanically thinned further in a Gatan dimple grinder to a thickness of 5 ~ 10 μm and finally thinned to perforation in a Gatan ion beam milling apparatus. Thinned specimens were examined in a Philips CM12 transmission electron microscope fitted with a Pv9900 EDS.

3 RESULTS AND DISCUSSION

3. 1 Highly porous SHS reaction products

Fig. 1(a) presents a fractograph morphology of the highly porous SHS reaction products showing the interconnected open pore structure of the infiltration preform. The average pore size is about 150 μm. Fig. 2(b) presents the micro-pores morphology of the obtained highly porous ceramics, which is a special phenomenon of the highly porous SHS reaction products. It is believed that these micro-pores are formed when the residual volatile agents and some volatile inclusions are heated to run out of the reaction products through these micro-pore paths during the SHS reaction. The size of these micro-pores is very small, only 1~ 2 μm, which could increase the specific surface of the obtained porous ceramics greatly. However, on the other hand, it also greatly increases the complexity of the interior geometry of the infiltration preform and the difficulties of full infiltration.

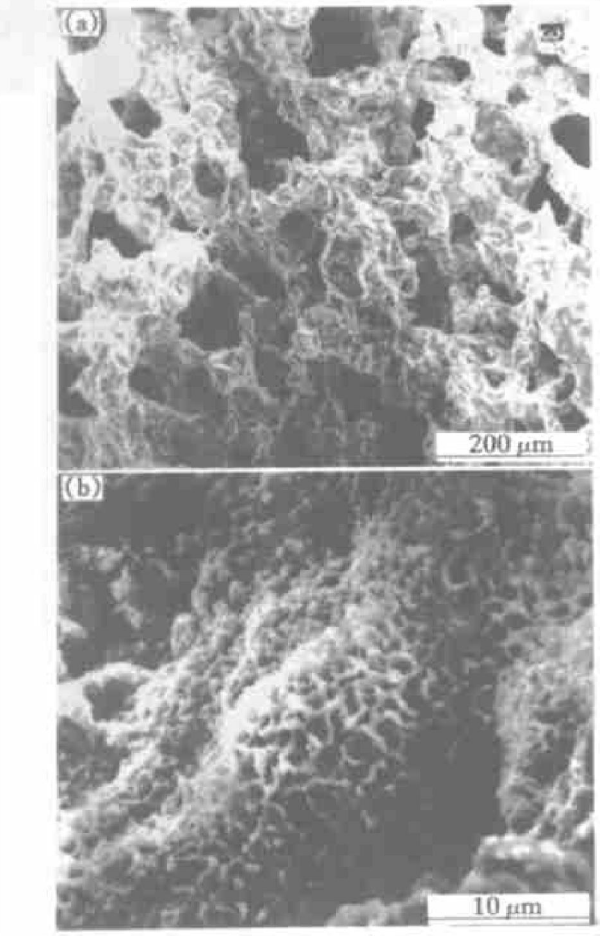

Fig. 1 Fractographs of highly porous SHS reaction products

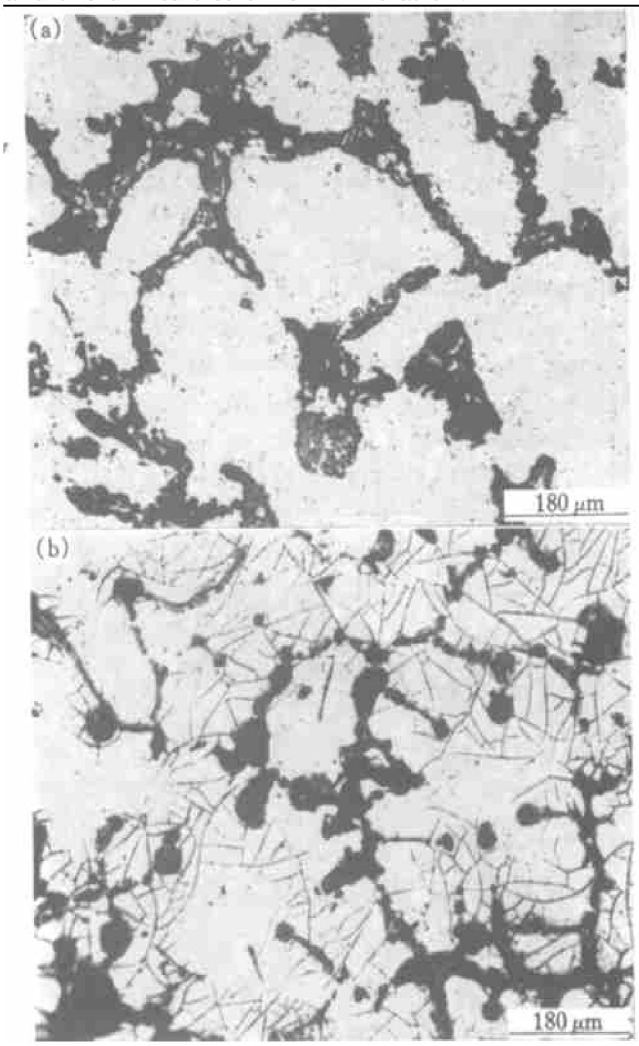

Fig. 2 Optical micrographs of MMIPC infiltrated with 2024 Al alloy

3. 2 Squeeze casting results

Fig. 2 shows optical micrographs of the composite infiltrated with 2024 Al alloy. Polished section of metal intruded materials produced with the highly porous SHS reaction products reveals that the architecture of the metal reinforcement replicates the architecture of the interconnected pores. Metal is observed to intrude into most of the sharp corners of this structure; however, some pores are presented adjacent to the reinforcement. We can see that on the whole, most of the preform is tolerably infiltrated and the interconnected network structure is evident as shown in Fig. 2(a).

Fig. 2(b) presents the stress corrosion observed on the optical micrographs showing great internal stress inherited in the resulting composites. In the squeeze casting process the cooling rate could become very high. It could result in the great internal stress of the obtained composites, which tends to reduce the strength of the obtained composites. The high cooling rate is caused by intimate contact between the melt and die as a result of the applied pressure (70 MPa), which leads to a high heated transfer coefficient and rapid heat extraction[9]. For example, the heat transfer coefficient for gravity casting is typically ~ $2 \times 10^3 W \cdot K^{-1} \cdot m^{-2}$, compared with a value of ~ $1 \times 10^4 W \cdot K^{-1} \cdot m^{-2}$ for a squeeze casting pressure of 100 MPa[9].

Fig. 3 presents the SEM fractographs of the crack surfaces (micro fractograph) of the resulting MMIPC showing an obvious cleavage behavior. From Fig. 3(b) we can see the Al alloy melt coming from different directions met in the center of the preform forming the obvious contacting interface and pore here. This interesting microscopy suggests that as a result of the restriction of the evaporating channels some remnant gases are gathered in the pores producing large internal pressure that inhibits both the syncretization of the melt and the complete infiltration of the pores here. So it is easy to result in great internal stress and origins of flaw in the obtained composites thus reducing the strength of the composite. In vacuum high-pressure infiltration, however, the preliminary vacuum process is obviously very beneficial in removing the gas before infiltration. Probably we could take the merits of both methods, first to infiltrate the preform by vacuum high-pressure infiltration, second to squeeze cast the obtained composites further.

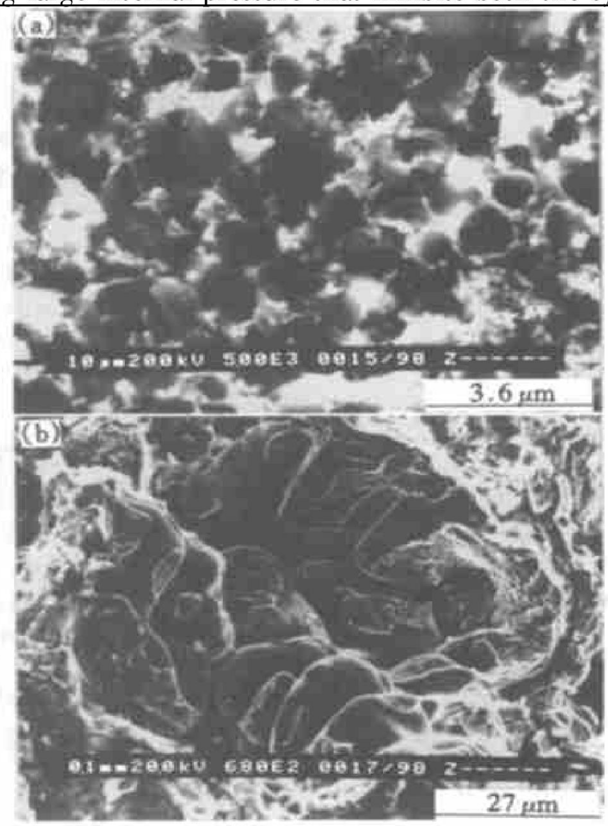

Fig. 3 SEM fractographs of MMIPC infiltrated with 2024 Al alloy

A transmission electron micrograph of an interface in the resulting composites is shown in Fig. 4. Some stacking fault could be seen in the reinforcement. No significant interfacial layer is observed, indicating perfect bonding of the matrix and the reinforcements. Also as revealed in Fig. 3(b), the reinforcement are attached to Al matrix on the crack surface, evincing their good bonding.

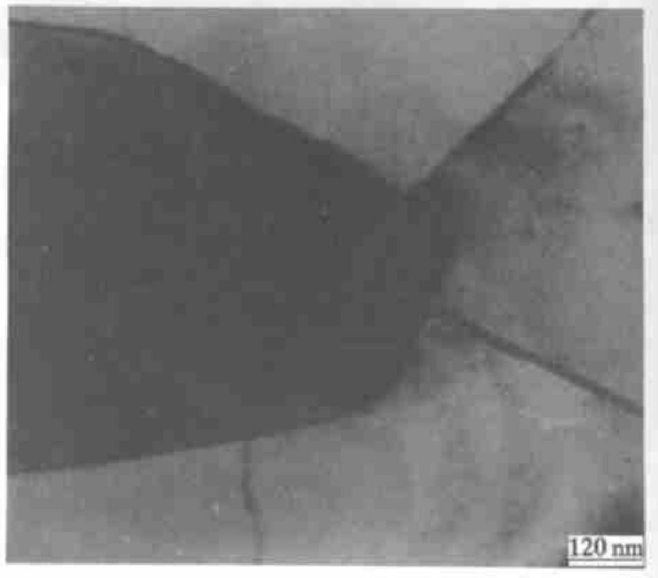

Fig. 4 TEM micrograph of resulting MMIPC showing interface, continuous network reinforcement appearing dark in micrograph

Fig. 5 presents a TEM view of the resulting Al-matrix interpenetrating phase composite. Due to the rapid cooling rate and the large mismatch of the CTE (coefficient of thermal expansion) between the Al and the interconnected network Al_2O_3-TiC reinforcement, large amounts of dislocations are found present in the matrix around the reinforcement. The dislocation density varies with distance from the reinforcement and they are arranged in a network array.

In our previous work[1] by vacuum high pressure infiltration, we reported that the average three-point bending strength of the resulting as-cast MMIPC infiltrated with 2024 Al alloy was 290 MPa. Here by employing squeeze casting method, we intend to reduce the porosity in the microstructure through enlarging the great infiltration pressure applied and the excellent feeding of solidification shrinkage by the

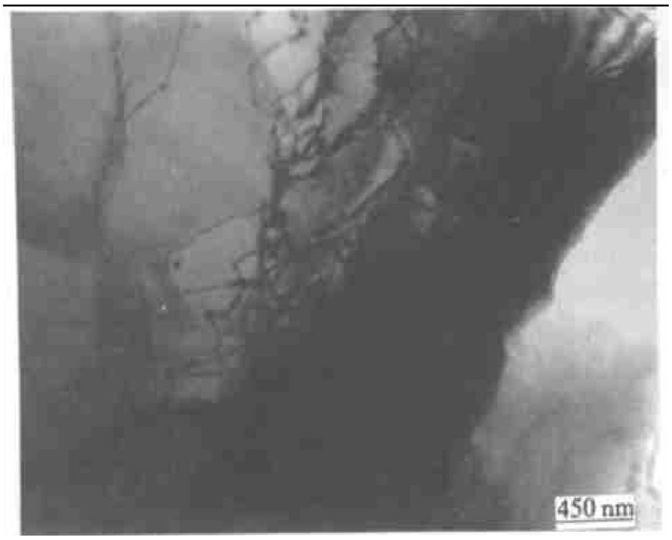

Fig. 5 TEM micrograph of resulting MMIPC illustrating network dislocation array, continuous network reinforcement appearing dark in micrograph

pressurized interdendritic fluid[10] thus increasing the strength and improving the microstructure of the resulting MMIPC further. In the present case a similar cleavage fracture behavior is exhibited, however, the bending strength of the as-cast MMIPC is 308 MPa, only a small increase compared to the earlier results. And according to our test the as-cast 2024 Al alloy matrix has an average three-point bending strength of 303 MPa in the same condition. We can see that the strengthening effect of the continuous network reinforcement phases is still not obvious.

As observed from the above micrograph, we consider one of the major reasons may be that the squeeze cast alloys of this investigation still exhibits considerable porosity contrary to Chatterjee and Das[11] who have reported that porosity in aluminum alloys can be eliminated by squeeze casting. Though, it should be noted that the maximum applied pressure in their work was 360 MPa, while in our case, it is 70 MPa. What is more, it is important to point out that much more complicated interior geometry of the porous ceramics preform has a definitely adverse effect on the squeeze casting process. Different from the situation in squeeze casting the particle or fiber preform, the possible flowing channels for the melt and the evaporating channels for the interior gas in the case of squeeze casting interconnected porous network preform are greatly reduced and restricted owing to the whole interconnectivity of the preform. This leads to great difficulties of both the melt flowing and the gas evaporating in such a rapid squeezing process, which is very disadvantageous to the complete infiltration of the preform raising the possibility of higher porosity thus greatly decreasing the strengthening effect. Concerning the 3-dimensionally interconnected porous network preform, a new model of unidirectional pressure infiltration should be established.

As pointed out in the previous work[1], due to the high porosity and rapid reaction the strength of the porous preform obtained from SHS reaction is comparatively low, which weakens the reinforcing effect of the 3-dimensionally continuous network reinforcements. To improve the strength of the composites, we should increase the strength of the preform by subjoining a sintering process. According to the research of Raymode[12], the SHS reaction products of Al_2O_3-TiC have good sintering ability. They could be sintered pressurelessly at 1 875~ 1 950 ℃.

At the same time we should also note that the squeeze casting method has increased the strength of the obtained MMIPC regardless of the present high porosity. Compared with the vacuum high-pressure infiltration method, it has great potential in improving the microstructure and performance of this new type of MMIPC.

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