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Effect of sintering temperature on the thermal expansion behavior of $ZrMgMo_3O_{12p}$/2024Al composite

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ABSTRACT

A 2024Al metal matrix composite with 10 vol% negative expansion ceramic $ZrMgMo_3O_{12}$ was fabricated by vacuum hot pressing, and the influence of sintering temperature on the microstructure and thermal expansion coefficient (CTE) of alloys was investigated. Experimental results showed that all $ZrMgMo_3O_{12p}$/2024Al composites sintered at 500–530 °C had a similar reticular structure and exhibited different linear expansion coefficients at 40–150 °C and 150–300 °C. The addition of 10 vol% $ZrMgMo_3O_{12}$ decreased the CTEs of 2024Al by ~ 16% at 40–150 °C and by ~ 7% at 150–300 °C. This addition also increased the hardness of 2024Al by ~ 23%. The density of the composites and the content of Al_2Cu in $ZrMgMo_3O_{12p}$/2024Al increased as the sintering temperature increased. The CTEs of the composites decreased, whereas hardness increased. Thermal cycling from 40 °C to 300 °C caused the CTEs of the composites to decrease gradually and reach a stable value after seven cycles. The lowest CTEs of 15.4×10^{-6} °C^{-1} at 40–150 °C and 20.1×10^{-6} °C^{-1} at 150–300 °C were obtained after 10 thermal cycles and were reduced by ~ 32% and ~ 17%, respectively, compared with the CTE of the 2024Al. Among the current reinforcements, $ZrMgMo_3O_{12}$ negative expansion ceramics showed the highest efficiency to decrease the CTE of Al matrix composites.

1. Introduction

With the rapid development of aerospace technology, strict requirements have been established for the primary physical properties of a material used in inertial instruments. The high elastic modulus and low CTE of materials are necessary to provide good dimensional stability for inertial instruments. As first-generation inertial materials, Al alloys are widely available because of their excellent processing performance [1]. However, Al alloys have become unattractive because their high CTEs may cause severe thermal mismatches and microcracks when these alloys are combined with other materials. These severe thermal mismatches and microcracks can result in the poor performance or even premature failure of devices. As a second-generation inertial material [1], beryllium is initially used as an inertial guidance device in America because of its high elastic modulus (E ≈ 280 GPa) and CTE similar to that of steel (11.4×10^{-6} °C^{-1}), but the application of beryllium has been restricted because of its toxicity, carcinogenicity, and high cost [1,2]. Al matrix composites, especially SiC_p/2024Al composites, which are reinforced with ceramic particulates, have been extensively investigated as the most representative alternative materials because of their high elastic modulus, high thermal conductivity, low density, low CTE similar to that of beryllium, and lower cost than that of beryllium [3,4]. Nevertheless, a high volume fraction (30–40 vol %) SiC_p should be added to a composite to guarantee the low CTE of the SiC_p/2024Al composite. This process deteriorates the machinability of materials, reduces strength, toughness, and compactness, and consequently degrades the performance of composites [5]. Therefore, the volume fraction of reinforcements in composites should be reduced to ensure a good combination of mechanical properties, physical properties, and machinability.

Various negative thermal expansion (NTE) ceramics, such as antiperovskite manganese-based compounds (e.g., $Mn_3Ga_{1-x}Ge_xN$) [6], fluorides [7,8], cyanides ($Ag_3[Co(CN)_6]$) [9], oxides (AMO_5, AM_2O_7, AM_2O_8, A_2O, and AM_3O_{12}) [10–12], nanostructures (Au and CuO nanoparticles) [13], perovskite structure compounds ($PbTiO_3$) [14], and magnetic materials (Th_2Zn_{17}) [15], have been developed. Metallic alloys reinforced by NTE ceramics may be effective in decreasing the thermal expansion of metallic alloys by introducing a small amount of NTE ceramics. Limited studies have demonstrated that NTE ceramics/Al or Cu composites have controllable and low CTEs. Zhang [16] found that 90 vol% α-$Cu_2V_2O_7$/Al composite fabricated through a solid-state method has a CTE of 0.49×10^{-6} °C^{-1} from room temperature to

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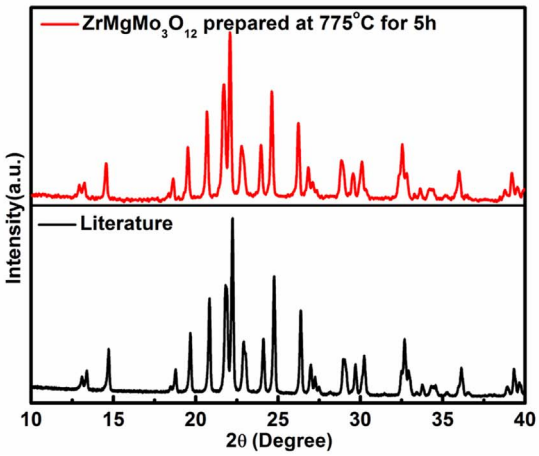
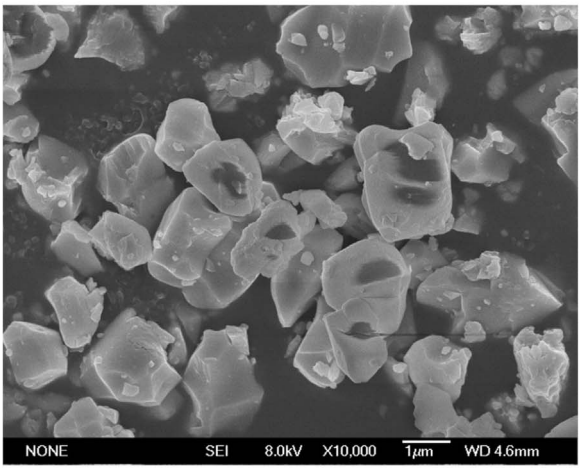
500 °C. Wu [17] found that 50 vol% ZrW_2O_8/Al composite fabricated with squeeze casting method yields a CTE of $(6 \pm 0.5) \times 10^{-6}$ °C^{-1} from 35 °C to 120 °C. However, its CTE is influenced by squeeze pressure and temperature. Zhou [18] reported that ZrW_2O_8/Al composite fabricated through spark plasma sintering shows low thermal expansion (4.83×10^{-6} °C^{-1}) from −70 to 20 °C if sintering temperature is controlled rationally. Similar to Al matrix composites reinforced with NTE ceramics, copper matrix composites reinforced with NTE ceramics have low CTEs. Sheng [19] observed that the effect of NTE on tetragonal cubic phase transition in $Sr_{0.2}Ba_{0.8}TiO_3$ particles can be utilized to reduce the CTE of $Sr_{0.2}Ba_{0.8}TiO_3$/Cu composite. Zhang et al. [20] showed that the CTEs of ZrW_2O_8/Cu composites fabricated with vacuum hot pressing (VHP) are controllable by adjusting the volume fraction of ZrW_2O_8. Although composites fabricated with low hot-pressing pressure have a relatively loose microstructure, a stable CTE of 1.6×10^{-6} °C^{-1} is obtained for 68 vol% ZrW_2O_8/Cu. Gao [21] reported that the CTEs of Cu/$Sc_2W_3O_{12}$ composites are controllable, and a low CTE of 2.1×10^{-6} °C^{-1} from room temperature to 300 °C is developed for 60 vol% $Sc_2W_3O_{12}$/Cu. Xu [22] indicated that the copper coating of $ZrWMoO_8$ powder slightly influences the CTEs of $ZrWMoO_8$/Cu composites. Furthermore, 50 vol% $ZrWMoO_8$/Cu composite fabricated with copper-coated $ZrWMoO_8$ powder has a CTE of 3.377×10^{-6} °C^{-1} from room temperature to 250 °C.

A NTE ceramic is an effective reinforcement to decrease the thermal expansion of Al matrix composites. However, various factors, such as hygroscopicity in atmospheric environments, decomposition, phase transition, and reaction of NTE ceramics with a metallic matrix at high temperatures and pressures [23], influence the thermal expansion behavior of NTE ceramics. Thus, some limitations are observed when NTE ceramics are combined with Al to fabricate NTE ceramics/Al composites. Wu [17] found that the reversible order–disorder phase transition at approximately 167 °C and the irreversible cubic–orthorhombic phase transition at a pressure of 40.21 GPa cause the instability of Al-ZrW_2O_8 composite. Zhou [18] reported that the phase transition of ZrW_2O_8 influences the stability of the CTEs of ZrW_2O_8/Al composites. Fabrication at a low sintering pressure or a temperature below the phase-transition temperature of ZrW_2O_8 can avoid the phase transition of ZrW_2O_8 and enable the low and stable thermal expansion of ZrW_2O_8/Al composites. Wu and Sumithra [24,25] found that the hygroscopicity of $Y_2Mo_3O_{12}$ and $Y_2W_3O_{12}$ hinders the production of thermostable $Y_2Mo_3O_{12}$/Al and $Y_2W_3O_{12}$/Al composites, although $Y_2Mo_3O_{12}$ (CTE = -9.36×10^{-6} °C^{-1}) and $Y_2W_3O_{12}$ (CTE = -7.34×10^{-6} °C^{-1}) have large negative CTEs from room temperature to 930 °C.

The key to fabricating thermostable aluminum matrix composites reinforced by NTE materials is restraining or avoiding the hygroscopicity and phase transition of negative expansion ceramics. Song et al. [26] and Romao et al. [27] reported a new $ZrMgMo_3O_{12}$ negative expansion material that remains unhygroscopic, and maintains structural stability below 500 °C, and possesses a near zero or negative CTE. On the basis of these characteristics, Xiao et al. [28] used this material to fabricate stable Al-$ZrMgMo_3O_{12}$ composite with controllable thermal expansion and found that 80 wt% $ZrMgMo_3O_{12p}$/Al composite yields a CTE of 0.77×10^{-6} °C^{-1} from room temperature to 400 °C and corresponds to a near-zero thermal expansion dielectric material. Furthermore, 60 wt% $ZrMgMo_3O_{12p}$/Al composite has a CTE of 8.72×10^{-6} °C^{-1}, which is one-third of that of Al (23.79×10^{-6} °C^{-1}). These preliminary results have indicated that $ZrMgMo_3O_{12}$ shows potential as an alternative to NTE reinforcement to fabricate Al matrix composites with a low and stable CTE. In this study, $ZrMgMo_3O_{12}$ was selected to reinforce 2024Al, and 10 vol% $ZrMgMo_3O_{12p}$/2024Al composites were fabricated by VHP. The influence of sintering temperature on the phase constitution, microstructure, mechanical property, and thermal expansion properties of $ZrMgMo_3O_{12p}$/2024Al composites was examined.



(a)



(b)

Fig. 1. The XRD patterns (a) and morphology (b) of $ZrMgMo_3O_{12}$ powder.

2. Experimental procedures

2.1. Experimental materials

The 2024Al powder with a nominal size of 10 μm and a chemical composition (mass fraction, %) of 3.852% Cu, 1.62% Mg, 0.042% Mn, 0.183% Fe, 0.087% Si, 0.2% Zn, and Al balance was supplied by Changsha TianJiu Metal Material Co., Ltd., Hunan, China. $ZrMgMo_3O_{12}$ power was synthesized using commercially analytical-grade ZrO_2 (≥ 99% purity), MgO (≥ 99% purity), and MoO_3 (≥ 99% purity) powder. ZrO_2, MgO, and MoO_3 were mixed at a molar ratio of 1:1:3 and then grounded for approximately 2 h with moderate anhydrous alcohol. The mixed powder was dried, sintered for 5 h at 775 °C in a box-type resistance furnace, and cooled in air to obtain $ZrMgMo_3O_{12}$ powder. Fig. 1 presents the X-ray diffraction (XRD) pattern and the morphology of $ZrMgMo_3O_{12}$ powder sintered at 775 °C for 5 h. In reference to Fig. 1(a), pure $ZrMgMo_3O_{12}$ powder was synthesized on the basis of the previously reported XRD patterns of $ZrMgMo_3O_{12}$ [26,27]. Fig. 1(b) presents the morphology of $ZrMgMo_3O_{12}$ powders. The particle size of $ZrMgMo_3O_{12}$ powder was 1–3 μm.

2.2. Composite fabrication

The 10 vol% $ZrMgMo_3O_{12p}$/2024Al composites were fabricated by VHP. The $ZrMgMo_3O_{12}$ powder and 2024Al were first milled for 2 h in a planetary ball mill in an alcohol medium. The ratio of the ball to the material was 10:1, and the milling velocity was 150 r/min. The mixed