Microwave-Induced Combustion Synthesis Of Porous Aluminum Matrix Composite Using A Ni-Based Metallic Glass As Precursor

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***Abstract*—Microwave processing of the Al-$Ni_{52.5}Zr_{15}Nb_{10}Ti_{15}Pt_{7.5}$ powder compacts in a separated magnetic (H-) field has been investigated. Porous Al matrix composites with a porous central region involving nearly spherical pores and a denser outer shell were fabricated by microwave-induced combustion reaction between Al powder and $Ni_{52.5}Zr_{15}Nb_{10}Ti_{15}Pt_{7.5}$ powder under flowing nitrogen gas. The evolution of the structure was obviously affected by controlling the green density of the pellet, stoichiometric ratio of Al and $Ni_{52.5}Zr_{15}Nb_{10}Ti_{15}Pt_{7.5}$, and input microwave power.**

Keywords—Microwave heating; Combustion reaction; Metallic glasses; Microstructure; Porous composite

I. INTRODUCTION

Porous metals exhibit a unique combination of properties, such as high density-compensated strength, high impact energy absorption capacity at low stresses, and the good permeability and acoustic damping properties that cannot be obtained with dense materials or porous polymers and ceramics [1-3]. As such, there is a wide range of possible applications of such materials in areas as diverse as catalysis, fuel cells, hydrogen storage, and thermal and acoustical insulation. Various methods including directly foaming, blowing bubbles, infiltration, powder metallurgy, electro-deposition have been developed for the synthesis of porous metals [1-3]. Among which, the methods that used metal powders as precursors are widely utilized, and show a good control over the cell shape, cell size and porosity distribution. Self-propagating combustion synthesis in two- or multicomponent systems has been widely reported as one of rapid and efficient methods for the synthesis of porous materials [4-9]. In this method, part of the powder compact is heated to ignite a strongly exothermic reaction between the elemental powders. Once the reaction occurs at the heated zone, then the heating of reaction rises at the temperature of the neighboring zone and ignites the reaction again. Hence the reaction propagates throughout the material spontaneously and results in the formation of highly porous materials with the introduction of a gasifying agent or gas bubbles. Generally, the combustion reaction is ignited at the surface of the material by thermal radiation, but a completely different approach is also used in which microwaves are used as the heating source. As microwave heating can offer some advantages over the conventional heating method including selective and volumetric heating, rapid processing time, high energy efficiency and environmental friendliness, microwave-induced combustion process can produce a unique microstructure and lead to a more complete conversion of reactants [10-12].

In fact, metals do not couple well with microwaves at room temperature as good electrical conductors. To ignite self-propagating combustion reactions in metallic systems, an external microwave susceptor, which is a microwave-absorbing material, is often used to heat the surface of metal until it absorbs microwave effectively [12-15]. However, in a few metal powder systems without using susceptor the combustion reaction also has been ignited by microwaves with slower heating rates and microwave coupling efficiency of mixtures of metals is greatly increased with the increase of the temperature [16,17]. Here we demonstrate that a rapid process for fabricating porous metallic materials can be realized in a binary metal-metallic glass powder system by uniting microwave heating and combustion reaction, in which gas bubbles are added to create a porous structure. The simply method involved heating the metal-metallic glass powder compacts in a short time and inducing combustion reactions between metal powder and metallic glassy powder in a separated microwave H-field to produce a porous composite by the introduction of gas bubbles. An ideal metal-metallic glass powder mixture for the present method therefore should have sufficient enhanced microwave absorption capacity with the increasing temperature to cause self-heating reaction at a temperature close to the melting temperature of metal.

II. EXPERIMENTAL PROCEDURES

Pure aluminum (Al) was selected as the matrix material due to its low weight, low melting temperature and good foaming properties. $Ni_{52.5}Zr_{15}Nb_{10}Ti_{15}Pt_{7.5}$ metallic glass was considered as both starting reactant material and microwave susceptor because metallic glasses not only exhibit a combination of properties of metallic and glassy materials and can be heated well in magnetic (H-) field in a form of powder, but also the major constituent elements in this metallic glass have large negative enthalpy of mixing with Al, and hence the Al-$Ni_{52.5}Zr_{15}Nb_{10}Ti_{15}Pt_{7.5}$ powder system is a potential candidate of high-temperature combustion reaction [18-22]. Here, we reported how normally microwave magnetic (H-) field at a frequency of 915 MHz could be exploited for the production of porous aluminum matrix composites using $Ni_{52.5}Zr_{15}Nb_{10}Ti_{15}Pt_{7.5}$ metallic glass as a precursor.

The starting materials were made from commercial powders of $Ni_{52.5}Zr_{15}Nb_{10}Ti_{15}Pt_{7.5}$ with the particle size under 75 μm and Al (99.5 wt.%, about 45μm). Al powder and $Ni_{52.5}Zr_{15}Nb_{10}Ti_{15}Pt_{7.5}$ metallic glassy powder with various volumetric ratios of 9:1, 4:1, 7:3 and 3:2 were used for the powder blends. The powder blends were mixed sufficiently in a mixer for 24 h and then pressed into 20 mm-diameter pellets with the same quality (about 7 g). The green density of the pellet is determined according to its mass and dimension. The microwave system used in this work consists of a 915MHz generator with a power output adjustable from 1 to 5 KW in steps of 40 W, a single mode tunable applicator and a computer control system [23]. All experiments are performed in a separated H-field maximum. Figure 1 shows a schematic view of microwave cavity in the applicator. To achieve the conditions closer to adiabatic, an alumina cylinder was inserted in the waveguide to form a cavity. Each pellet was placed in an open 25 mm-diameter quartz tube. The tube was insulated from the outer quartz cylinder by adiabatic fiber. The process was carried out under flowing nitrogen gas. To order to initiate microwave absorption, the resonating cavity was tuned by a remotely controlled three-stub tuner and the slide plunger at the end of the waveguide. It was necessary for sustaining the effective microwave heating to adjust the three-stub tuner for the minimization of the reflected power. Continuous temperature measurement was made using an infrared pyrometer (IR-FAINNN, temperature range: 350-1600 ℃). XRD analyses of the sample and the initial powder mixtures were performed using a monochromatic Cu Kα radiation (Rigaku, RIHT-Ultima). The thermal stability of the powder mixtures was examined using differential thermal analysis (DTA) at a heating rate of 20 K/min. SEM image of the sample was taken using a Hitachi S-4800 scanning electron microscope. The porosity was also measured by the Archimedes method.

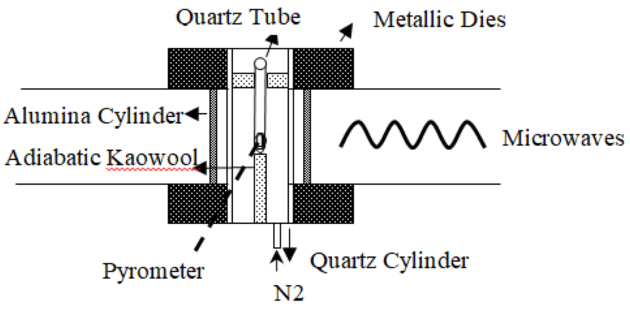

Figure 1 Schematic view of microwave cavity in a single-mode 915 MHz applicator.

III. RESULTS AND DISCUSSION

Figure 2 shows the DTA curves of the powder mixtures of Al and $Ni_{52.5}Zr_{15}Nb_{10}Ti_{15}Pt_{7.5}$ with various volumetric ratios. As can be seen, in the temperature range below 1000 ℃, a strong exothermic peak corresponding to the combustion reaction in the complex Al-$Ni_{52.5}Zr_{15}Nb_{10}Ti_{15}Pt_{7.5}$ system is seen for all the volumetric ratio, indicating the combustion reaction can be induced between Al and $Ni_{52.5}Zr_{15}Nb_{10}Ti_{15}Pt_{7.5}$ metallic glass with a wide composition range. With the increase of the relative amount of Ni-based metallic glassy powders in the starting mixtures, the exothermic peak height becomes more obvious. This means that there is more possibility for Ni-based metallic glass to contact and react with Al. Different from other compositions, the Al-10 vol.% $Ni_{52.5}Zr_{15}Nb_{10}Ti_{15}Pt_{7.5}$ powder mixture exhibits two exothermic events. The primary small one corresponds to the crystallization process of Ni-based metallic glass, while the second one may be a reaction between liquid Al and the crystalline products of $Ni_{52.5}Zr_{15}Nb_{10}Ti_{15}Pt_{7.5}$ metallic glass because it is followed by a small endothermic event corresponding to the melting of Al. With the increase of the content of Ni-based metallic glass, in the Al-20 vol.% $Ni_{52.5}Zr_{15}Nb_{10}Ti_{15}Pt_{7.5}$ powder mixture the endothermic event does not occur, but at the temperature range of 850-900 ℃ the endothermic event corresponding to the melting of the intermetallics induced by combustion reaction is found, which is also seen for the Al-30 and 40 vol.% $Ni_{52.5}Zr_{15}Nb_{10}Ti_{15}Pt_{7.5}$ powder mixtures. With the further increase of the content of metallic glassy powders the exothermic event corresponding to the crystallization of metallic glass becomes smaller until completely disappeared. In the case of 40 vol.% $Ni_{52.5}Zr_{15}Nb_{10}Ti_{15}Pt_{7.5}$ metallic glassy powders, only a large exothermic event is seen, indicating that a complete reaction of Ni-based metallic glass with Al can be attained. These results show that the variation of the volumetric ratios between Al powder and $Ni_{52.5}Zr_{15}Nb_{10}Ti_{15}Pt_{7.5}$ metallic glassy powder obviously affect the thermal process of the mixture.

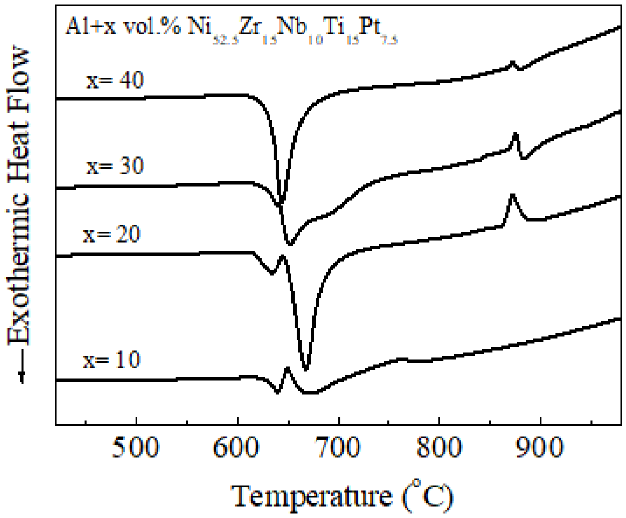

Figure 2 DTA curves of the powder mixtures of Al and $Ni_{52.5}Zr_{15}Nb_{10}Ti_{15}Pt_{7.5}$ with various volumetric ratios.

Figure 3 shows the temperature profiles of the Al-$Ni_{52.5}Zr_{15}Nb_{10}Ti_{15}Pt_{7.5}$ pellets with various volumetric ratios heated in a microwave H-field. It can be seen that, at the input power of 1000 W, microwave-induced combustion synthesis is easily realized in the Al-40 vol. % $Ni_{52.5}Zr_{15}Nb_{10}Ti_{15}Pt_{7.5}$ pellet. Microwaves can couple rapidly with the mixture in H-field, and the peak temperature of about 1100 ℃ can be attained. The heating rate becomes faster with the increasing temperature, indicating that its microwave absorption capacity can be enhanced at higher temperature. At a higher input power of 1400 W, the Al-30 vol.% $Ni_{52.5}Zr_{15}Nb_{10}Ti_{15}Pt_{7.5}$ pellet can be heated rapidly up after a long duration time and microwave-induced combustion reaction led to the peak temperature of about 960 ℃ in H-field. At 3000 W, the combustion reaction is ignited by microwave heating of the Al-20 vol.% $Ni_{52.5}Zr_{15}Nb_{10}Ti_{15}Pt_{7.5}$ pellet and the heating rate becomes slower with the increase of the temperature before combustion reaction. The Al-10 vol.% $Ni_{52.5}Zr_{15}Nb_{10}Ti_{15}Pt_{7.5}$ pellet also couples with microwaves and is heated rapidly up in the initial stage, but the combustion reaction does not occur because microwave energy absorbed is not enough for igniting combustion reaction and supporting the propagation of combustion wave front. The above results show that microwave-induced combustion synthesis between Al and $Ni_{52.5}Zr_{15}Nb_{10}Ti_{15}Pt_{7.5}$ metallic glass can be effectively performed in H-field, and ignited at a lower temperature compared to the conventional combustion synthesis. This means that microwave heating can enhance combustion reaction. Besides the input microwave energy and stoichiometric ratio of the powder mixture, the density of the composite pellet also obviously affects its heating behavior in H-field. Figure 4 the temperature profiles of the Al-20vol. % $Ni_{52.5}Zr_{15}Nb_{10}Ti_{15}Pt_{7.5}$ pellets with various densities in a microwave H-field. Under the same input power higher green density means lower heating rate in the initial stage, and then results in terminating the ignition of the combustion reaction.

To obtain a porous structure without the introduction of a gasifying agent by combustion synthesis, control of the propagation of the combustion wave front is very important because pure liquid metals induced by combustion reaction cannot easily form a porous structure by introducing a flowing gas into them. It can be seen from the above results that the propagation of the combustion wave front depends obviously on the green density and stoichiometric ratio of the pellet and input microwave energy. It also has been proven that high compact density and high thermal conductivity of the material exposed in a single-mode applicator limit or suppress the combustion wave front propagation, and the precise control can be afforded by decreasing the input power step by step on the velocity of propagation of the combustion wave front, and even the propagation can be terminated by turning the power off [24]. Therefore, the combustion reaction process of Al-$Ni_{52.5}Zr_{15}Nb_{10}Ti_{15}Pt_{7.5}$ powder mixtures can be controlled by changing the stoichiometric ratio of the powder mixture and relative density of the compact as well as synchronously decreasing or turning off the input power. Based on the above consideration, porous Al matrix composites were rapidly fabricated in the binary Al-$Ni_{52.5}Zr_{15}Nb_{10}Ti_{15}Pt_{7.5}$ powder mixture by microwave-induced combustion synthesis in H-field. Figure 5 shows the appearance, SEM image and XRD patterns of the Al-20vol.% $Ni_{52.5}Zr_{15}Nb_{10}Ti_{15}Pt_{7.5}$ pellet and its final product with 25% porosity obtained by microwave-induced combustion synthesis in H-field. As can be seen in Fig. 5(a), compared to the initial pellet, its final porous product fabricated by microwave-induced combustion synthesis exhibits an obviously volume expansion in a short time. Its cross-section image shows that a close-cell structure consisting of a porous interior and a dense surface layer or shell is formed. The SEM image of the porous Al matrix composite shown in Fig. 5(b) indicates that the nearly spherical pores are homogeneously distributed in the interior and exhibit a very wide distribution of pore sizes, which is in the range of about 10 to 900 µm. As $Ni_{52.5}Zr_{15}Nb_{10}Ti_{15}Pt_{7.5}$ metallic glass is a multi-component alloy system, the combustion reaction between it and Al is more complex than that between elemental metals. It can be found from Fig. 5(c) that the Al_6NbZr and Al_3Ni phase are detected in the pattern besides pure Al phase. This indicates that these two phases are main reactant products of the combustion reaction between Al and $Ni_{52.5}Zr_{15}Nb_{10}Ti_{15}Pt_{7.5}$ metallic glass.

Combustion synthesis between Al and $Ni_{52.5}Zr_{15}Nb_{10}Ti_{15}Pt_{7.5}$ metallic glasses is ignited by microwaves internally and forced to propagate outward in a controllable manner. Under a non-adiabatic condition microwave heating often induces an inverse temperature gradient where the temperature decreases from the interior to the surface due to the heat loss at the surface of the materials [25]. This makes the ignition temperature be reached first in the center of the sample with the increase of the temperature and high temperature in the interior of the sample forces propagation of a relatively uniform radial combustion wave front to the exterior of the sample, which ensures complete combustion reaction. As the temperature recorded is the temperature of the surface, in fact the interior temperature is higher, even can induce the melting of the reaction products, which is verified by the above results. During this procedure, extensive gas evolution is induced by a flowing nitrogen gas in the central molten region to created bubbles which remained in the structure, and thus formed the spherical pores, while in the outer shell the intermetallics are sintered by viscous flow at a lower temperature. Such structure also has been found in graded porous glass-metal composites produced by microwave heating [26].

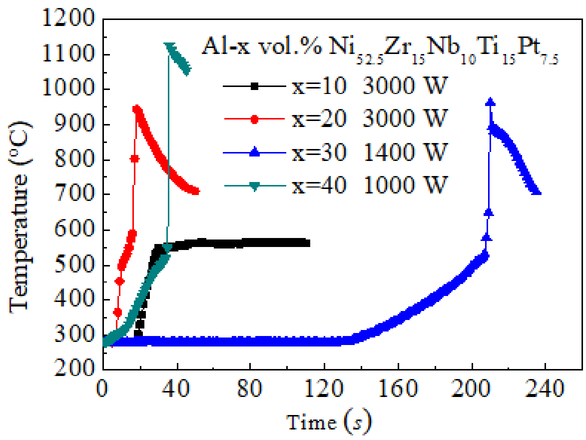

Figure 3 Temperature profiles of the Al-$Ni_{52.5}Zr_{15}Nb_{10}Ti_{15}Pt_{7.5}$ pellets with various volumetric ratios in a microwave H-field.

Compared to the elemental metals or crystalline alloys, the introduction of $Ni_{52.5}Zr_{15}Nb_{10}Ti_{15}Pt_{7.5}$ metallic glass contributes to enhance microwave coupling capacity of Al at room temperature and induces the combustion reaction at a lower temperature because metallic glass is a metastable and multi-component alloy system and exhibits a lower electrical conductivity and melting temperature compared to its crystalline counterpart. Moreover, during the combustion reaction, the heat released by the crystallization of Ni-based metallic glass can accelerate the propagation of combustion wave front and lead to a sufficient combustion reaction. As multi-component metallic glasses can be obtained in various alloy systems with a wide composition range, this offers a possibility for us to synthesis porous metal matrix composite with unique structure and properties using metallic glasses as precursors by microwave-induced combustion reaction.

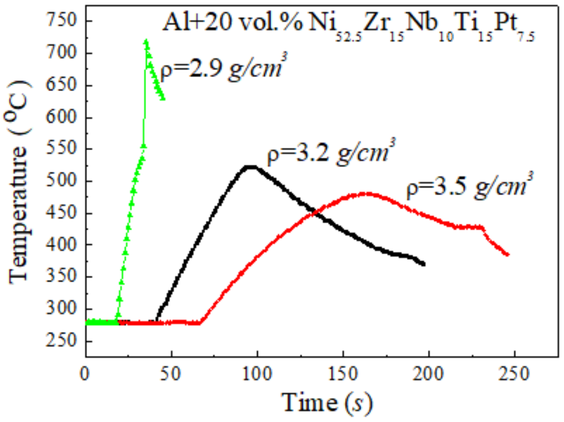

Figure 4 Temperature profiles of the Al-20vol.% $Ni_{52.5}Zr_{15}Nb_{10}Ti_{15}Pt_{7.5}$ pellets with various densities in a microwave H-field.

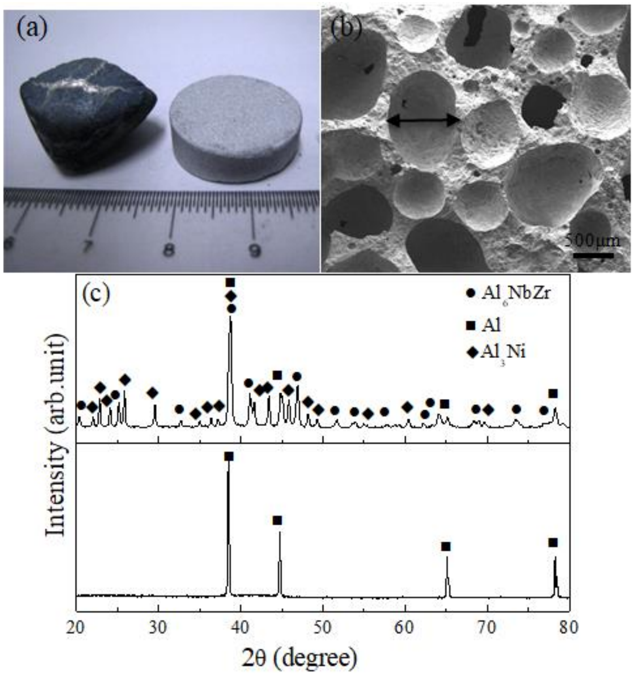

Figure 5 (a) Appearance of the the Al-20vol.% $Ni_{52.5}Zr_{15}Nb_{10}Ti_{15}Pt_{7.5}$ composite and pellet; (b) SEM image of cross-section of the composite; (c) XRD patterns of the composite and initial pellet.

IV. CONCLUSION

A microwave-induced combustion synthesis method to fabricate porous metals was introduced. It was demonstrated that combustion synthesis could be realized in a very short time by microwave heating of Al and $Ni_{52.5}Zr_{15}Nb_{10}Ti_{15}Pt_{7.5}$ metallic glassy particles in a separated H-field using a frequency of 915 MHz. Al matrix composites with a porous central region involving nearly spherical pores and a denser outer shell were fabricated by expandable metal-metallic glass precursor under flowing nitrogen gas. Such close-cell structure was strongly affected by the porosity of the powder compact, blending ratio of Al/ $Ni_{52.5}Zr_{15}Nb_{10}Ti_{15}Pt_{7.5}$, and input power. We believe that this simple technique can be extended for more metallic glasses and used for a wider application in the future.

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