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Powder-Metallurgical Free-Form Microchanneling Process for Producing Metallic Microreactors

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Abstract

We verify effectiveness of the concept of powder-metallurgical free-form microchanneling with functional microchannel lining. It is based on a microscopic infiltration phenomenon during liquid phase sintering of a powder mixture of metals with different melting points. A shaped compound of an organic binder and a powder metal with lower melting point is used as the sacrificial core that gives the shape of the microchannel. The resultant microchannel is surrounded by an alloy layer, which is expected to have useful properties.

Key words

Microreactor, Microchannel, Free-form Microchanneling, Powder Metallurgy, Infiltration

1. Introduction

In recent years, remarkable progress has been made in microreactor technology.[1-3] Metallic microreactors for catalytic reactions have also been investigated actively. The high surface to volume ratio of their microchannel structure is suitable for catalytic reaction, and it allows keeping the reaction temperature throughout the reactor even if the reaction is highly exothermic or endothermic. In addition, the metallic materials are better adapted to the catalyst support or substrate used for high temperature reactions because of their good heat resistance, high thermal conductivity, and ease of property control by means of alloying.

One of the most important topics in the study of the metallic microreactors is a configuration of the method to fabricate microchannels in metal bodies. Some microchanneling processes have been proposed, including precision mechanical machining, photoetching, micro-electro discharge machining, laser or electron beam machining, and galvanization.[3] All these processes are methods of manufacturing structured surfaces and require many processing time and steps to construct the microreactors. Methods to fabricate microchannels directly in bulk metals would be desirable from a practical viewpoint.

Recently, we proposed a powder-metallurgical free-form microchanneling process.[4-5] Figure 1 illustrates the schema of the process. In this process, two kinds of powder metals with different melting points are used. One is the body metal that has higher melting point and is to make the device body. Another is the sacrificial-core metal. A shaped compound of a sacrificial-core metal powder and an organic binder is used as the sacrificial core that gives the shape of the microchannel. A body-metal powder compact containing a sacrificial core is sintered at a temperature between the melting points of the two metals.

The organic binder in the sacrificial core is removed during heating of the powder compact, and then molten sacrificial-core metal migrates to the body-metal powder region by infiltration and partly by diffusion. As a result, a microchannel is produced at the site initially occupied by the sacrificial core. In addition, the body metal and the sacrificial-core metal in many cases produce an alloy layer lining the microchannel.

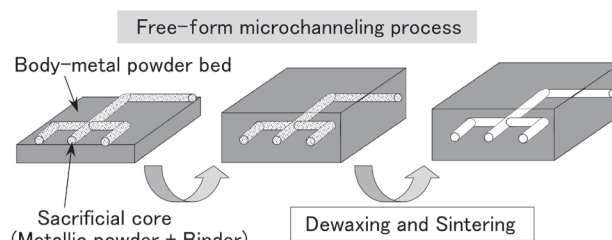


Fig. 1 Concept of the free-form microchanneling process

It was also shown that the composition of the lining layer was changed by addition of an alloying element into the sacrificial-core metal.[4-5] This result indicates that the structure and function of the microchannel wall can be controlled by our microchanneling process. We call this new concept "functional microchannel lining". In this concept, the sacrificial core acts as not only the spacer [6-7] but also the source of the alloying element to control the properties of the lining layer. Furthermore, the use of alloy powder for the sacrificial core in our process makes it possible to fabricate a microchannel network with a complicatedly diverged configuration even if the alloy has poor workability.

In the present study, we examined the feasibility of our process, fabricating microchannels with various configurations, including T-shaped, converging/diverging and long zigzag channels. We also investigated the process parameters that control the structure and composition of the lining layer.

2. Experimental Procedure

A combination of titanium (the body metal) and aluminum (the sacrificial-core metal), designated as Ti/Al, is used in the experiments to produce T-shaped and converging/diverging channels, and a combination of copper and tin (Cu/Sn) was examined for a long zigzag channel. Table 1 lists the purity and average particle size of each metal powder.

The organic binder was prepared from yellow wax and pine resin blended in a mass ratio of 1:1, which was mixed

with a sacrificial-core metal powder in a given volume ratio, 1:1 or 7:2, to produce the sacrificial-core compound. The compound was extruded into a wire 0.5 or 1.0 mm in diameter. The 1.0 mm-diameter wire was used only to produce converging/diverging channels. Figure 2 depicts the wire and our extrusion equipment. The wires were then cut, shaped and joined for the sacrificial core.

Table 1 Purity and size of the metal powders used for free-form microchanneling experiments

Metal	Diameter, d [μm]	Purity, c_p [mass%]
Ti	under 45	99
Cu	45-75	99.9
Al	100	99.7
Sn	38	99.99

Eighteen grams of the body-metal powder, in which the sacrificial core was included, was cold-pressed into a cylindrical green compact in a metal mold. The green compact had a diameter of 20 mm. Although the initial cross section of the wire was round, it was distorted into an oblate figure by unidirectional pressing.[4-5]

Figure 3 presents the heating patterns for the microchanneling heat treatment, including dewaxing and sintering. The sintering temperatures were 1473 K for the Ti-Al system and 1123 K for the Cu-Sn system. All the heat treatments were carried out in an argon gas atmosphere.

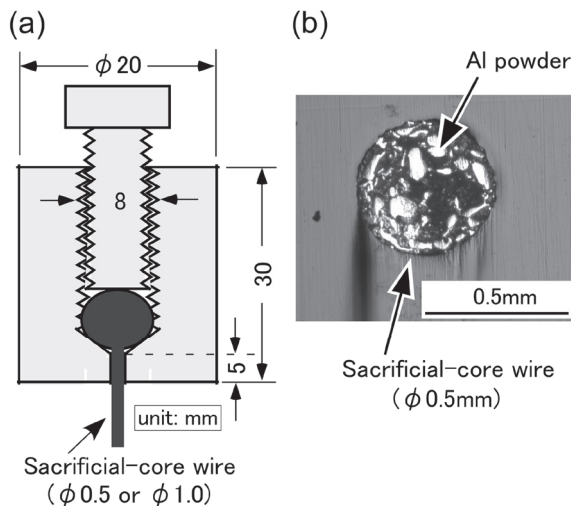


Fig. 2 Fabrication of sacrificial-core wire. (a) Compound-injection equipment. (b) Optical micrograph of a cross section of a sacrificial-core wire. The volume ratio of the binder to the sacrificial-core metal, V_B/V_M , was 1

We conducted several microchanneling experiments to investigate three process parameters that would influence the lining layer formation: the compacting pressure to

prepare the powder compact, the combination ratio of the binder and the metal powder in the sacrificial-core compound, and the kind of third element added to the sacrificial-core metal. The sacrificial core used in each experiment was a straight wire 0.5 mm in diameter and 15 mm in length.

The combinations Cu/Sn and Ti/Al were used to examine the first and the second parameter, respectively. For the third parameter, titanium was used as the body metal, and two kinds of Al-Si based alloys, an ADC12 alloy (an Al-11.2mol%Si alloy containing 1.1mol%Cu) and a SP8 alloy (an Al-15.8mol%Si alloy containing 0.5mol%Cu-1.0mol%Fe-2.2mol%Mg-0.5mol%Ni), were used as the sacrificial-core metal. Both Al-Si based alloy powders were prepared by gas-atomizing. The average diameter of the ADC12 alloy powder was 50 μm , and that of the SP8 alloy was 120 μm .

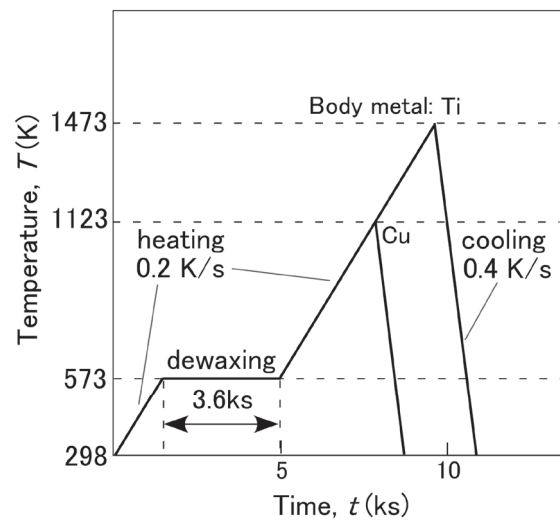


Fig. 3 Time-temperature patterns for the heat treatments

3. Results and Discussion

3.1 Formation of microchannels with various shapes

Figure 4 presents examples of optical micrographs of transverse cross sections of the microchannels fabricated by the free-form microchanneling process with titanium as the body metal and aluminum as the sacrificial-core metal. The compacting pressure for producing the powder compact, P_c , was 624MPa and the relative density of the compact was 0.89. In Fig. 4, it is observed that the shape of the transverse cross section of the microchannel is not influenced by the volume ratio of the binder to the metal in the sacrificial-core compound, V_B/V_M .

Figures 5(a) and (b) show longitudinal cross sections of a T-shaped channel and a converging/diverging channel. In each experiment, shaping and joining of the sacrificial-core compound were easily performed, and the resulting microchannel has the same shape as the sacrificial core. This fact demonstrates the one of the advantages of using the sacrificial-core compound.

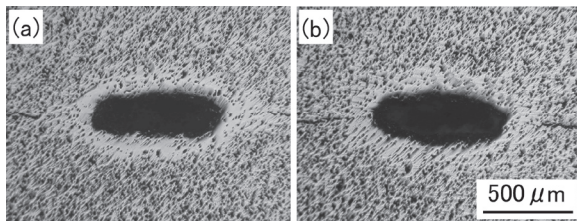


Fig. 4 Optical micrographs of cross sections of the microchannels in the Ti-Al specimens. Volume ratio of the binder to the sacrificial-core metal: (a) $V_B/V_M=1$, (b) $V_B/V_M=3.5$

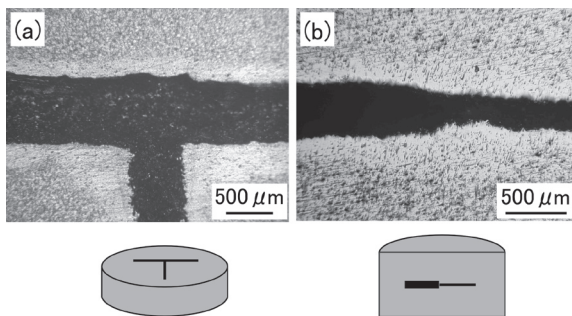


Fig. 5 Optical micrographs showing the formation of a T-shaped channel (a) and a converging/diverging channel (b) in Ti-Al specimens. $V_B/V_M=3.5$

Figure 6 shows a result of a flow examination for a long zigzag channel in a Cu/Sn specimen. The compact pressure of the specimen was 156MPa and the resulted relative density was 0.68. As shown in Fig. 6, the water flowed through the microchannel and jetted out from the outlet. The result shows that the microchannel was open throughout its entire length. Furthermore, it's interesting to note that a small amount of water seeped through the sintered body. This indicates that an open porous body and a microchannel network can be produced simultaneously. Such a structure may be suitable for a transpiration-cooling device which is applied to the thermal protection of machines such as a disaster preventing robot [8].

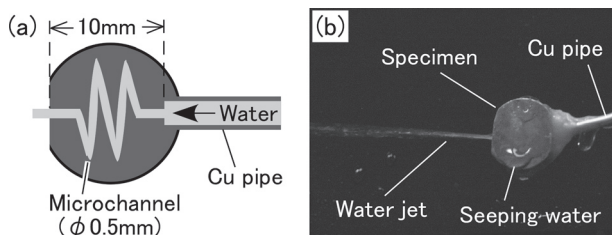


Fig. 6 Flow examination for a long zigzag channel produced in a Cu/Sn specimen ($V_B/V_M=3.5$ and $P_c=156\text{MPa}$). (a) Schema of the specimen. (b) Result of the examination

3.2 Process parameters to control the structure and composition of the microchannel lining layer

Figure 7 presents optical micrographs of microchannels produced in Cu/Sn specimens, showing the influence of the compacting pressure on the structure of the device. Both the degree of circularity of the cross-sectional shape of the microchannel and the porosity of the sintered body decreased as the compacting pressure increased. Therefore, the pressing pressure is one of the important parameters to control the performance of the transpiration-cooling device noted above.

Figure 8 shows aluminum concentration profiles across the lining layers in the Ti/Al specimens produced using different sacrificial-core compounds. For the case of $V_B/V_M=1$ [5], the aluminum concentration is about 7-26 mol% higher than the case of $V_B/V_M=3.5$. The binder/metal volume ratio affected not the shape of the microchannel, as shown in Fig. 4, but the composition of the lining layer.

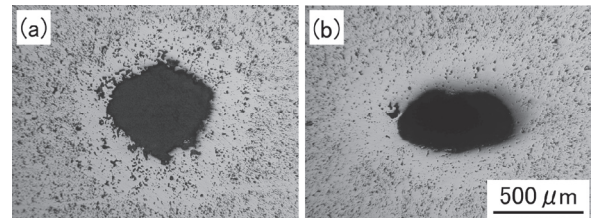


Fig. 7 Influence of the compacting pressure for producing the powder compact, P_c , on the structure near the microchannel in the Cu/Sn specimen ($V_B/V_M=1$). (a) $P_c=156\text{MPa}$. (b) $P_c=234\text{MPa}$

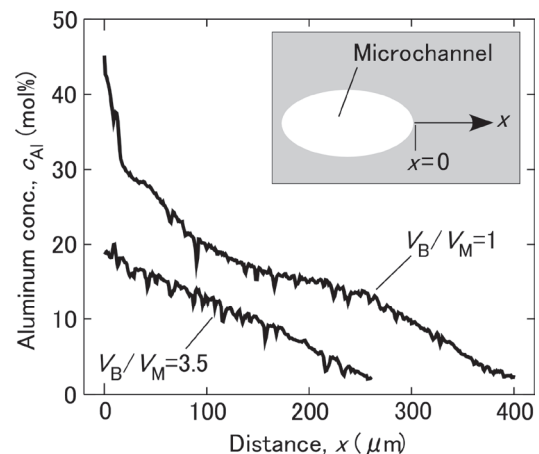


Fig. 8 Influence of the binder/metal volume ratio in the sacrificial core compound, V_B/V_M , on the aluminum concentration profile across the lining layer surrounding the microchannel in the Ti/Al specimens

Figures 9 and 10 depict the results of third-element addition to the sacrificial core metal.

Figure 9 shows the structure and the aluminum- and copper-concentration profiles in a Ti/Al-34mol%Cu alloy specimen.[5] The average copper concentration in the region from $x=0$ to $100\ \mu\text{m}$, C_{Cu} , was 7.9 mol%, and the average aluminum concentration, C_{Al} , was 17.4 mol%. Figure 10 shows the result for the case of a Ti/ADC12 specimen. The average silicon concentration in the region from $x=0$ to $100\ \mu\text{m}$, C_{Si} , was 5.5 mol%, and C_{Al} was 30.1 mol%. In each specimen, the lining layer was rich in the third element added to the sacrificial core metal. However, the concentration profile varies from one element to the next. In Fig. 9, the ratio of the average copper concentration to the average aluminum concentration, $C_{\text{Cu}}/C_{\text{Al}}$, was 0.45, which is 27% lower than the composition ratio in the sacrificial-core metal. On the other hand, in Fig. 10, the ratio, $C_{\text{Si}}/C_{\text{Al}}$, was 0.18, and was 38% higher than the composition ratio in the sacrificial-core metal. This difference is probably caused by that in the diffusion rate of each element in titanium or reaction-product phases.

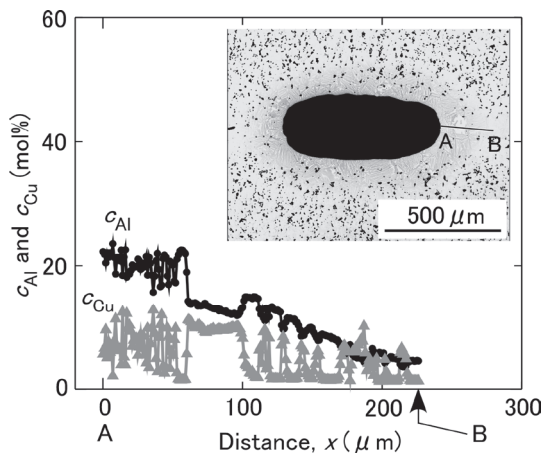


Fig. 9 Back-scattered electron image and aluminum- and copper- concentration profiles near the microchannel produced with titanium as the body metal and an Al-34mol%Cu alloy as the sacrificial-core metal.[5] $V_B/V_M=1$

Figure 11 presents back-scattered electron images of a cross section of the microchannel fabricated using the combination Ti/SP8. As discussed in a previous paper [5], the copper addition to the sacrificial-core metal increases smoothness of the inner wall of the microchannel (see Fig. 9). This is probably caused by dissolution of titanium into copper-rich Al-Cu alloy melt. In contrast, silicon addition tends to produce a concave-convex surface as shown in Figs. 10 and 11.

Control of roughness of the microchannel wall is very important in practical applications. For example, it may enable flow-resistance control in microchannel flow systems, or it provides an essential part of the fabrication process of a microchannel wall with a high-specific surface area in the microreactors for catalyst reactions.

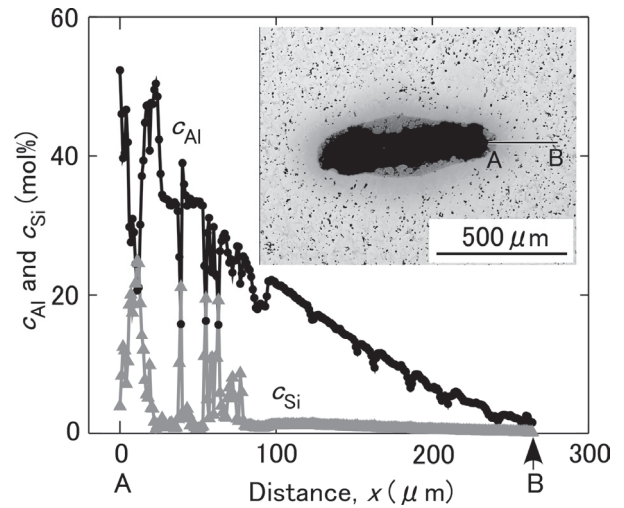


Fig. 10 Back-scattered electron image and aluminum- and silicon- concentration profiles near the microchannel produced with titanium as the body metal and an ADC12 alloy as the sacrificial-core metal. $V_B/V_M=1$

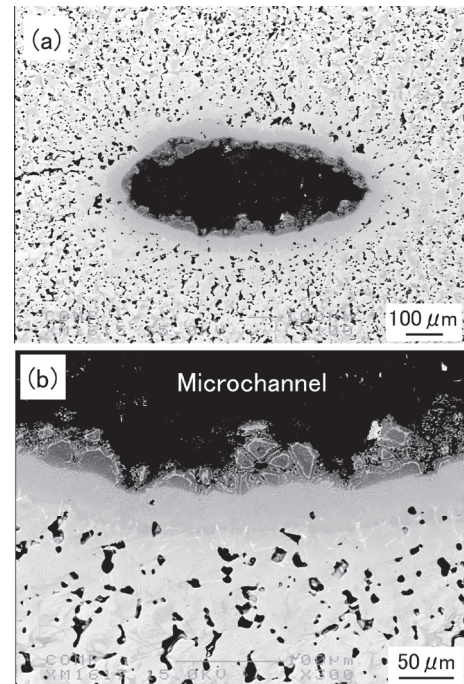


Fig. 11 Back-scattered electron image near the microchannel produced with titanium and an SP8 alloy. $V_B/V_M=1$. (a) Whole image. (b) Magnified view of the lining layer

It should be also noted that these Al-Cu and Al-Si alloys have extremely poor workability and are difficult to shape into a network of fine wire by machining processes. The above results demonstrate the effectiveness of the concept of free-form microchanneling.

4. Conclusions

We verified practical effectiveness of the free-form microchanneling process, fabricating T-shaped, converging/diverging and long zigzag microchannels. We also investigated the process parameters to control the structure and composition of the lining layer. The results of our investigation can be summarized as follows.

(1) T-shaped and converging/diverging channels can be produced by the free-form microchanneling process.

(2) The binder/metal volume ratio in the sacrificial-core compound influences not the contour of the transverse cross section of the microchannel but the composition of the lining layer.

(3) The compacting pressure for preparing the powder compact affects the degree of circularity of the cross-sectional shape of the microchannel and the porosity of the sintered body. Both of them decrease as the compacting pressure increases.

(4) The roughness of the microchannel wall and the composition of the lining layer are influenced by third-element addition to the sacrificial-core metal. They are affected by the kind and the concentration of the third element.

Acknowledgements

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References

- [1] Kiwi-Minsker, L. and Renken, A.: *Catalysis Today*, **110** (2005), 2-14.
- [2] Meille, V.: *Applied Catalysis A: General*, **315** (2006), 1-17.
- [3] Mills, P.L., Quiram, D.J. and Ryley, J.F.: *Chemical Engineering Science*, **62** (2007), 6992-7010.
- [4] Ohmi, T., Takatoo, M., Iguchi, M., Matsuura, K. and Kudoh, M.: *Journal of JSEM*, **5** (2005), 234-238.
- [5] Ohmi, T., Takatoo, M., Iguchi, M., Matsuura, K. and Kudoh, M.: *Mater. Trans.*, **47** (2006), 2137-2142.
- [6] Hakamada, M., Asao, Y., Kuromura, T., Chen, Y., Kusuda, H. and Mabuchi, M.: *Scripta Mater.*, **56** (2007), 781-783.
- [7] Hakamada, M., Asao, Y., Kuromura, T., Chen, Y., Kusuda, H. and Mabuchi, M.: *Mater. Letters*, **62** (2008), 1118-1121.
- [8] Yano, T., Imai, R. and Isomura, K.: *Nagare (Journal of Japan Society of Fluid Mechanics)*, **21** (2002), 55-61.