Relationship between microstructure and mechanical property of sintered Cu specimens prepared via particulate extrusion

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Abstract

Sintered Cu specimens were prepared via particulate extrusion of different Cu feed materials. The feed materials were differentiated due to different Cu powder shape, size and content. A multi-component polymer mixer, B4550, comprising low density polyethylene (LDPE): paraffin wax (PW): stearic acid (SA) with a ratio of 45:50:5 by weight, was selected as a binder. Volume ratios between the Cu powder and the binder were varied, e.g., 60:40, 65:35 and 70:30. The feed materials were extruded downward, with an extrusion ratio of 10:1, into a rod-shape part with a diameter of 5.0 mm. Only the extrudable Cu specimens were debinded, sintered and tested. The sintered Cu specimens produced from small powder particles showed better mechanical properties than those of sintered coarse Cu powders. Amongst small Cu powders, irregularity of the powder provided superior mechanical properties. Microstructures of the sintered Cu specimens provided some clues to the property differences amongst them. Smaller powder particle size and with higher irregularity provide better sinterability, which yields a higher numbers of metallic bonds or strengths.

Keywords: Particulate extrusion, microstructure, mechanical property and copper powder

1. Introduction

There are several methods for fabricating structural metal/alloy parts. By employing a certain metal forming process, it is impossible to produce the whole engineering parts with different shapes and sizes. One process may be suitable for some kinds of parts. For example, when a metal part with high aspect ratio and good surface finishing is needed, metal extrusion is considered as an optimized choice of metal processing. Extrusion is one of metal forming processes involving squeezing of a metal billet to flow through a die and form a part with a desired shape. In bulk metal extrusion, a metal can be deformed and flowed only when a high pressure is exerted to a metal billet, made from either a metal ingot or a powdered metal compact, under considerably high temperature [1, 2]. For example, pressure required for extrusion of aluminium alloys is up to 1000 MPa and extrusion temperatures for the same materials are around 400-500 °C [2].

Because of high pressure and high temperature, a heavy extrusion press machine and heating instruments are needed. Energy consumption for a conventional metal extrusion is therefore high. A new approach for powder extrusion (PE) based on shaping of metal powder-binder mixes not only reduces energy consumption but also provide opportunity for shaping porous parts with high aspect ratio. The PE process involves steps of (i) mixing of the metal powder with a binder granulate to form a "feed material", (ii) extrusion of the feed material, (iii) removing of the binder (debinding) and (iv) sintering of the debinded or brown parts. Different shapes of powder affect rheology of the feed material and inter-particle friction of the green part (extrudate) [3]. Spherical metal powders from gas atomization are suitable component of the feed material because of their high packing density, high flow rate and fluid-like characteristics. However, the cost of gas atomized metal powders is relatively high. Their low inter-particle friction also affects component shape retention. In contrast, the cost of irregular shape powders is low. The irregular shape results in a high inter-particle friction with improved shape retention. The packing density of irregular shape powders is low and shaping is difficult due to thigh viscosity of the feed material [4].

In our previous work [5], the PE process involving steps of (i) mixing of the metal powder with a binder granulate to form a "feed material", (ii) extrusion of the feed material, (iii) removing of the binder (debinding) and (iv) sintering of the debinded or brown parts has been preliminarily studied. Five different binder formulae comprising low density polyethylene (LDPE); paraffin wax; and stearic acid were studied. The binder B4550 was the most promising for preparation of the feed materials of spherical and irregular Cu powders. The optimum debinding step consists of solvent and thermal debinding. Properties of the sintered extrudates are influenced by not only by the sintering time but also by the shape of the powders. In this article, relationship between microstructure and mechanical property of the sintered Cu specimens has been examined. Powder characters controlling microstructures of the sintered materials have also been investigated in order to fully understand the particulate extrusion of metal powder feed materials.

2. Experimental Procedure

The metal powders used in this work were Cu powders with spherical and irregular shapes. The powders were classified into three size fractions, namely < 45, 45-75 and 75-125 μm. A multi-component binder, B4550, was prepared from mixtures of low density polyethylene (LDPE), paraffin wax (PW) and stearic acid (SA) [5]. Mixing of the Cu powders with the binders was carried out in an internal mixer with twin screw counter rotor at 130 °C and speed of 25 rpm for 60 min. Designation of the feed material of spherical Cu powders was simply made by putting “S” in front of the binder code. For example, SB4550 represented the feed material of spherical Cu powders mixed with the binder B4550. Similarly, the feed material designation of the irregular Cu powders began with “I”. Volume ratios between the Cu powder and the binder were varied as 60:40, 65:35, 70:30 and 75:25. The feed material was put into the extrusion chamber and then extruded with an extrusion pressure of 30 bar at 95 °C. Debinding was performed in two stages, (i) solvent debinding by immersing the extrudate in hexane solvent at 60 °C for different times to identify the optimum solvent debinding conditions and (ii) thermal debinding with the following heating cycle: heating from room temperature to 100 °C with heating rate of 5 °C/min and from 100 to 250 °C with lower heating rate of 1 °C/min., holding at 250 °C for 90 min., heating from 250 to 450 °C with heating rate of 2 °C/min., holding at 450 °C for 60 min., and finally cooling in the furnace. Sintering was performed at 1030 °C under H_2 atmosphere. Holding times for sintering were varied from 60 to 240 minutes. A universal testing machine (Instron model 8801) was employed to measure mechanical properties of the sintered parts. Microstructural observation was carried out using optical microscopy.

3. Results and Discussion

(1) Extrudability of the cu feed materials [6]

Relationship between microstructure and property of the sintered Cu specimens produced by particulate extrusion was constructed from experimental data of the extrudable materials. It is thus important to declare extrudability of the feed materials here. Flowability of the feed materials might be dependent on some parameters, related to powder characters and processing natures. This property determines extrudability of the feed materials. Experimental results (Table 1) showed that extrusion of Cu powders was limited by three parameters, namely powder shape, size and metal powder to binder ratio. For spherical powder, metal powder to binder ratio was a limiting factor. The feed materials prepared from spherical Cu powders with particle sizes up to 125 μm could be extruded. Extrusion of the spherical Cu feed material was limited when binder content ≤ 30 vol. %. For irregular powder, particle size and binder content were limiting factors. Only the powders with particle sizes of < 75 could be extruded. This was attributed to flow inhibition caused by inter-particle friction between irregular powder particles. Increasing of binder content may increase flowability of the irregular Cu powder feed materials. However, higher binder content will need long time and high amount of chemicals for debinding.

Table 1 Extrusion performance of the Cu feed material

Powder size range	Spherical powder				Irregular powder			
	60 % Cu	65 % Cu	70 % Cu	75 % Cu	60 % Cu	65 % Cu	70 % Cu	75 % Cu
75-125 μm	✓	✓	✓	×	×	×	×	×
45-75 μm	✓	✓	✓	×	✓	×	×	×
< 45 μm	✓	✓	✓	×	✓	✓	×	×

(2) Microstructural observation

(2.1) Effect of powder particle size and sintering time

Microstructures of the sintered Cu specimens 60 vol. % Cu clearly depended on original powder particle size. The matrix showing relationship between spherical Cu powder particle size, sintering time and microstructure (**Fig. 1**) provides useful explanation why the sintered specimens of small Cu powders showing superior mechanical property. With a fixed sintering time (along the column), microstructures indicate that the sintered Cu specimens prepared from small powders show good signs of sintering (numbers of metallic bonds or sintered necks) compared to those prepared from coarser powder particles. **Fig. 1** indicates that number of metallic bonds decrease with increasing powder particle size. In general, smaller powder particles have higher surface areas hence surface energy. The driving force for sintering process is that the powder particles need to reduce their surface energy [7-9]. That means under the same sintering conditions, small powder particles are favored to weld by forming interparticle bonds or sintering necks. The numbers of interparticle bonds directly relate to mechanical property of the sintered materials.

Powder character | **Sintering time (hr.)** 1, 2, 3

Spherical, < 45 µm

Spherical, 45-75 µm

Spherical, 75-125 µm

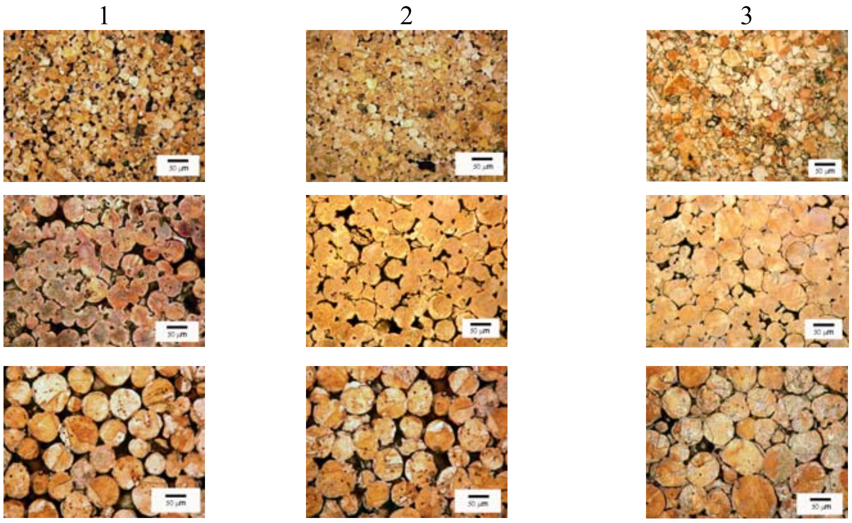


Fig. 1 Microstructures of sintered Cu materials prepared by extrsion of Cu powders with defferent particle sizes and sintering times.

(2.2) Effect of powder particle shape and sintering time

Effect of powder particle shape under the same sintering condition was constructed in the form of the matrix relating powder character, sintering time and microstructure (**Fig. 2**). In the case of Cu powders with particle sizes < 45 µm, comparison of microstructures of the sintered Cu specimens prepared from the spherical and irregular powders, as given in **Fig. 2**, may give no different information if only number of interparticle bonds and pores are compared. In fact, interpretation of sintered microstructures needs to take sintering stages into account. The sintering stages can be indicated by pore morphology difference. The specimen prepared from spherical powders < 45 µm and sintered for 1 hr. showed some interconnected pores and some pores with sharp corners. This indicates premature sintering. In contrast, the specimen prepared from irregular powders < 45 µm and sintered for the same period of time showed pores with round corners indicating mature sintering. The matured-sintered materials give optimum mechanical properties [8]. Microstructures of the sintered Cu specimens prepared from Cu feed materials using the powders with particle sizes between 45-75 µm showed clearer evidences of sintering stage difference. Under the same sintering conditions, different sintering stages experienced by the extruded Cu specimens may be controlled by surface areas/energies of the powders. For powders with the same particle sizes, irregular powders have more surface areas hence surface energies. This may be the important reason why irregularity of powders causes different mechanical properties given in #(2).

Powder character | **Sintering time (hr.)** 1, 2, 3

Spherical, < 45 µm

Irregular, < 45 µm

Spherical, 45-75 µm

Irregular, 45-75 µm

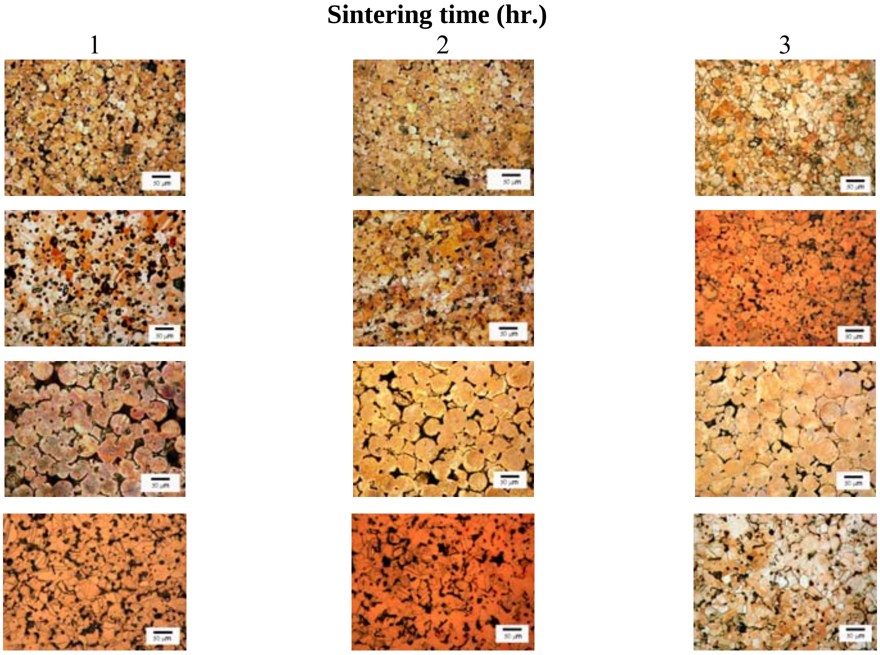
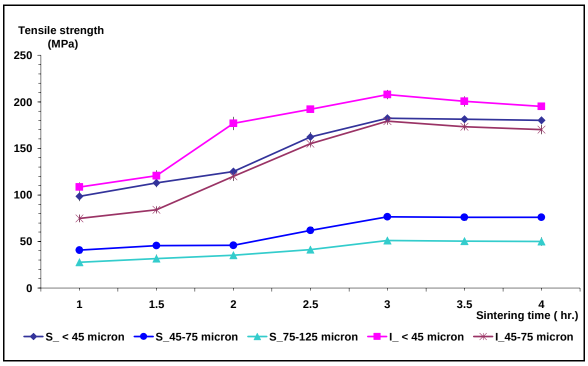


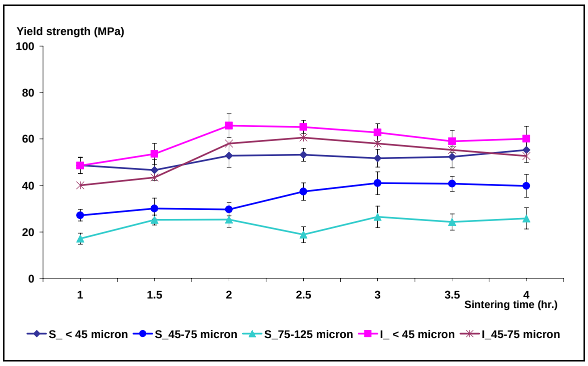
Fig. 2 Microstructures of sintered Cu materials; the effect of powder particle shapes.

(3) Mechanical property of the sintered Cu material

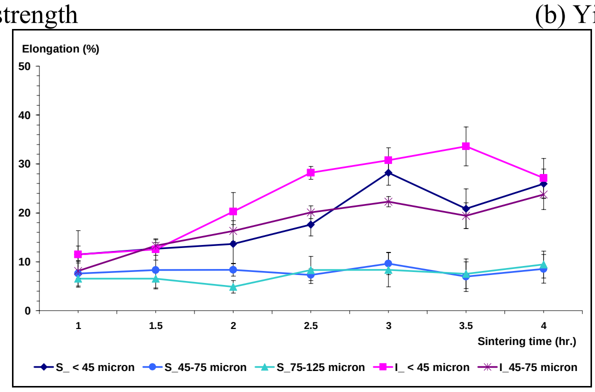
Mechanical properties of the sintered Cu extrudates were controlled by powder size and shape (**Fig. 3**). The sintered materials produced from small Cu powders showed better mechanical properties than those of sintered coarse Cu powders. Amongst small Cu powders, irregularity of the powder provided superior mechanical properties although it prohibited flowability of the feed material. The powder irregularity may increase points or areas of contact between powder particles. The points and areas of contact are important places where material transport starts. Microstructures of the sintered material would confirm this hypothesis. Microstructural observation will be further investigated.



(a) Tensile strength



(b) Yield strength



(c) elongation

Fig. 3 Mechanical properties of the sintered Cu extrudates.

Information obtained from #(2) microstructural observation and #(3) mechanical property leads to a simple interrelationship between powder irregularity, powder size and sinterability as given in **Fig. 4.** Smaller powder particle size and with higher irregularity provide better sinterability, which yields a higher numbers of metallic bonds or strengths.

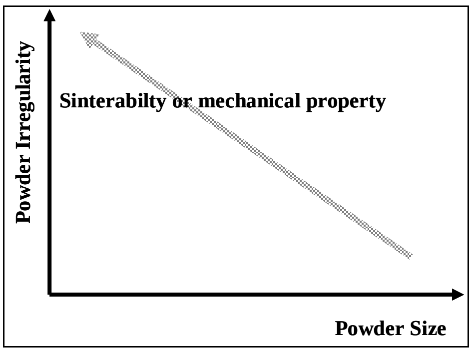


Fig. 4 A simple interrelationship between powder size, powder irregularity and sinterability or mechanical property of the sintered Cu specimens.

Conclusions

By using the same binder system, extrudability of the Cu feed materials depends on powder particle size, shape and content. Powder particle size also affects sinterability of the materials prepared from particulate materials. Experimental results indicate that smaller powder particles are sintered easily than the coarser ones due to their higher driving force to reduce surface areas/energies. The experimental results also indicate that with the same powder particle size, irregular powders can be sintered easier than the spherical ones. Enhanced sinterability in the irregular powders is resulted from higher surface areas/energies. Difference in sinterability of the powders with different size and shape is a good explanation for difference in mechanical properties of the sintered materials prepared from powders with different characters.

Acknowledgements

The authors would like to express their sincere gratitude to Materials Technology Division, School of Energy Environment and Materials, King Mongkut University of Technology Thonburi, Bangkok, Thailand and National Metal and Materials Technology Center (MTEC), Pathum Thani, Thailand, for financial support.

References

1. P. K. Saha, 2000, Aluminum Extrusion Technology, ASM International, Ohio.
2. T. Sheppard, 1999, Extrusion of Aluminium Alloys, Kluwer Academic Publishers, London.
3. S-B. Li, J-X. Xie, 2007, "Fabrication of thin-walled 316L stainless steel seamless pipes by extrusion technology"Materials processing technology, vol.183, pp. 57-61.
4. C. Karatas, S. Saritas, 1998, "Rheological properties of MIM feedstocks produced from gas and water-atomized 316L stainless steel powders", Journal of Turkish Engineering ad Environmental Sciences of TUBITAK, vol.22, pp. 445-451.
5. R. Wichianrak, N. Thavarungkul, N. Poolthong and R. Tongsri, 2008, "Copper Powder Extrusion: A Smart Processing for Energy and Environment Conservation", *Advanced Materials Research*, **55-57**, 357-360.
6. R. Wichianrak, M. Morakotjinda, T. Yodkaew, N. Torsangtum, R. Krataithong, A. Daraphan, O. Coovattanachai, B. Vetayanugul, N. Thavarungkul, N. Poolthong and R. Tongsri, 2008, "Extrusion of Cu Feed Materials", submitted to MSATV, 16-19 September 2008.
7. H. E. Exner and E, Arzt, 1996, "Sintering process", Physical metallurgy, Elsevier Science, Amsterdam, pp. 2628-2662.
8. R. M. German, 1996, Sintering theory and practice, Wiley Interscience, New York.
9. A. Bose, 1995, Advances in particulate materials, Butter worth-Heinemann, Boston.