

Original Research

# Forming of magnesium alloy microtubes in the fabrication of biodegradable stents

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## Abstract

Magnesium alloys have, in recent years, been recognized as highly promising biodegradable materials, especially for vascular stent applications. Forming of magnesium alloys into high-precision thin-wall tubes has however presented a technological barrier in the fabrication of vascular stents, because of the poor workability of magnesium at room temperature. In the present study, the forming processes, i.e., hot indirect extrusion and multi-pass cold drawing were used to fabricate seamless microtubes of a magnesium alloy. The magnesium alloy ZM21 was selected as a representative biomaterial for biodegradable stent applications. Microtubes with an outside diameter of 2.9 mm and a wall thickness of 0.2 mm were successfully produced at the fourth pass of cold drawing without inter-pass annealing. Dimensional evaluation showed that multi-pass cold drawing was effective in correcting dimensional non-uniformity arising from hot indirect extrusion. Examinations of the microstructures of microtubes revealed the generation of a large number of twins as a result of accumulated work hardening at the third and fourth passes of cold drawing, corresponding to the significantly raised forming forces. The work demonstrated the viability of the forming process route selected for the fabrication of biodegradable magnesium alloy microtubes.

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**Keywords:** Vascular stent; Microtube; Magnesium alloy; Extrusion; Drawing

## 1. Introduction

In recent years, balloon-mounted vascular stenting has become the primary choice for the treatment of atherosclerosis, coronary artery diseases in particular. At present, more than three million stent implantations to reopen stenosed vessels are performed every year [1,2]. The vast majority of the stents currently in clinic use are made of stainless steels or cobalt–chromium alloys [3] and permanently stay in blood vessels. Although these metallic materials in general possess excellent mechanical properties and acceptable biocompatibilities, such stents may produce serious postoperative side-effects such as thrombosis and in-stent restenosis [4,5], thereby limiting the long-term success of stenting [6]. One of the approaches to reducing and even circumventing these side-effects is to implant temporary stents made of biodegradable

materials. Polymer-based biodegradable stents developed so far still suffer from the major drawback of limited mechanical performance. Much attention has recently been paid to metallic biomaterials for biodegradable stents. Pure magnesium, as biodegradable and biocompatible metallic material, presents itself as a promising material for biodegradable stents [7]. However, alloying, although making the biocompatibility issue complex, is necessary to enhance the biocorrosion resistance and mechanical properties of magnesium. By adding appropriate alloying elements to magnesium, magnesium alloy stents are expected to be able to perform their biomechanical functions by supporting the arterial wall during the remodeling process over a period of 6–12 months and then progressively degrade. Rare earth (RE) elements have been found to be able to serve the purposes of alloying better than most of other bio-tolerable elements. RE-containing magnesium alloys such as WE43 alloy and AE21 have therefore been subjected to extensive in vitro investigations [8]. Furthermore, a number of in vivo evaluations of RE-containing magnesium alloy stents have been performed. Encouraging results have been obtained and new

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strategies to optimize the performance of metallic biodegradable stents have been mapped out, focusing on alloying, coating and microstructure control.

In developing metallic biodegradable stents, developing the fabrication technology is of equal importance, as the fabrication technology determines the feasibility of converting a piece of starting stock to a functional medical device in a cost-effective manner. The research in this aspect has however received insufficient attention. In the case of fabricating permanent metallic stents, tubing is produced in the seamless form, followed by tube drawing and laser slitting. In the case of magnesium alloys, however, it is very difficult to produce cold-drawn seamless tubes with exact dimensions on the microscale due to their poor plasticity at room temperature. Only a few studies have so far been conducted on developing the fabrication technology for magnesium alloy microtubes and improving the mechanical properties as well as the dimension accuracy of the semi-finished products for stents [9,10]. Ge et al. obtained ultrafine-grained (UFG) tubes of magnesium alloy ZM21 through severe plastic deformation using the equal channel angular pressing (ECAP) method [11]. Faraji et al. succeeded in producing AZ91 UFG tubes by means of multi-pass tubular channel angular pressing (TCAP) [12]. With the ultrafine-grained structure created during these severe plastic deformation processes, the mechanical properties of magnesium alloy tubes could be enhanced considerably [13] and even biocorrosion resistance could be improved to a certain extent, both of which are beneficial for stent applications. Furushima et al. successfully produced AZ31 magnesium alloy microtubes from ECAP-processed and extruded billets by using the multi-pass dieless drawing method [14]. Local heating was applied during dieless drawing to make the heated area with an initial outside diameter of 2 mm and a wall thickness of 0.5 mm thinner. However, the process did not allow the change of the ratio of the inside diameter to the outside diameter of microtubes and as such its applicability is rather limited. Moreover, the refined grain structures from the ECAP process would increase the difficulties in subsequent forming processes to the near net shape of stents by greatly enhancing the forming force. It would therefore be of practical significance, if magnesium alloy microtubes could be drawn from the billets without going through a severe plastic deformation process.

The present study was an attempt to develop a forming process route for the fabrication of magnesium alloy microtubes through simple forming processes, i.e., indirect extrusion and multi-pass cold drawing without involving a severe plastic deformation process. Pre-extruded ZM21 alloy billets were used as the starting material. Dimensional accuracies and microstructures of extruded and cold-drawn tubes were examined to establish the viability of the forming process route.

## 2. Forming methods and experimental setup for stent microtubes

A material for tubular stents must possess (i) appropriate mechanical strength and ductility and (ii) microscale dimensions [15]. Considering these two basic requirements, hot extrusion appears to be a suitable method to convert a large-

dimension as-cast ingot into a small-dimension as-extruded bar. During the process, in addition to large plastic deformation, coarse grains formed during casting are significantly refined through dynamic recrystallization (DRX), leading to enhanced workability for subsequent processing. Obviously, the direct extrusion process does not allow the production of seamless tubes and the indirect extrusion process must be utilized to convert a tubular billet to a thin-wall hollow tube. Moreover, during the indirect extrusion process, the force requirement is reduced because of the decreased friction between the billet and the mandrel. Prior to indirect extrusion, machining is applied to cut the extruded bars into cylindrical hollow billets. These hollow billets are indirectly extruded to produce seamless thin-wall tubes at an elevated temperature, followed by multi-pass cold drawing where dimensional inaccuracies arising during indirect extrusion are rectified in addition to generating the near net shape of stents and enhancing their mechanical performance.

The whole forming process route for the fabrication of stent microtubes [16] is schematically shown in Fig. 1.

### 2.1. Development of the high-precision extrusion press for the hot indirect extrusion of microtubes

A lab-scale horizontal extrusion press was developed to extrude a hollow billet into a thin-wall seamless tube (Fig. 2). The container and the mandrel were mounted on a movable platen driven by a hydraulic piston. The die was fastened to the front fixed platen. Heating elements surrounding the container were used to heat the container and the billet. Extrusion temperature was measured by a thermocouple inserted into the container to realize closed-loop temperature control around the pre-set value. During hot extrusion, the piston together with the container and the mandrel moved forward, thereby pressing the hollow billet into the die, and a thin-wall tube was extruded through the gap between the die orifice and the mandrel. Fig. 3 schematically illustrates the indirect extrusion process for the fabrication of magnesium alloy microtubes.

### 2.2. Development of the special tooling for the cold drawing of microtubes

After indirect extrusion, the wall thickness of extruded tubes would need to be reduced further and moreover the variations

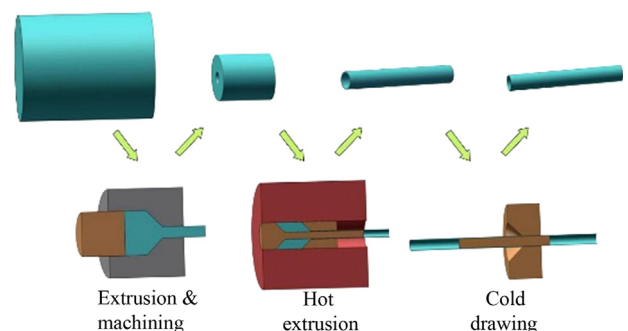


Fig. 1. Schematic illustration of the forming processes for the fabrication of seamless microtubes.

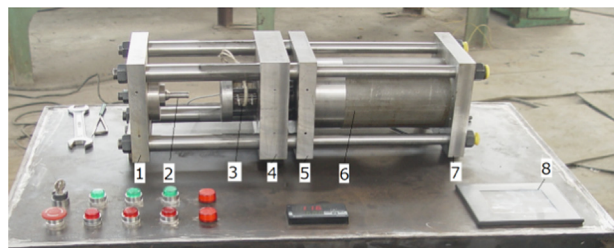


Fig. 2. Lab-scale press for the hot indirect extrusion of magnesium alloy microtubes. 1: front fixed platen, 2: die, 3: container with heating elements (mandrel is moulded inside), 4: movable platen, 5: middle fixed platen, 6: hydraulic cylinder, 7: rear fixed platen, and 8: console.

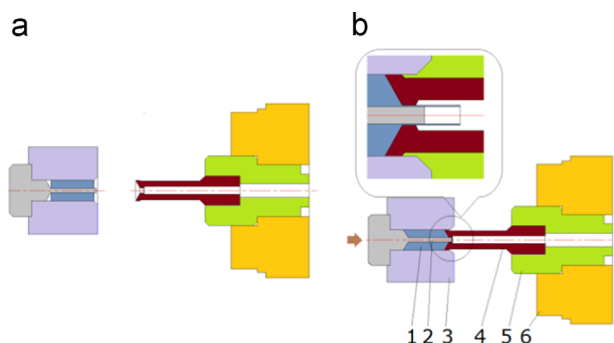


Fig. 3. Schematic illustration of hot indirect extrusion: (a) before extrusion and (b) during extrusion. 1: hollow billet, 2: mandrel, 3: container, 4: die, 5: die set, and 6: front fixed platen.

in wall thickness arising during hot indirect extrusion must be reduced. A floating mandrel method was chosen for cold drawing. With this method, the friction force between the tube and mandrel would be considerably decreased, as compared with the fixed mandrel method. The wall thickness uniformity of microtubes would be better, as compared with the mandrel-free method. The tooling shown in Fig. 4 was mounted on a universal mechanical testing machine. The reduction ratio in the cross-section area of the drawn tube at each pass would be rather restricted, considering the poor formability of magnesium at room temperature. After a certain number of passes annealing would be necessary to regain the workability of the material.

### 3. Material and experimental details

#### 3.1. Material

Magnesium alloy ZM21 ingots were cut into rods with a length of 100 mm and a diameter of 48.3 mm for extrusion. Billets were extruded at a temperature of 375 °C, a reduction ratio of 20 and a ram speed of 1 mm/s to produce bars with a diameter of 11.2 mm, using an extrusion press having a container with a diameter of 50 mm. Extruded bars were then machined into tubular billets with an outside diameter of 10.9 mm and a length of 20 mm. These billets were used in the subsequent hot indirect extrusion process.

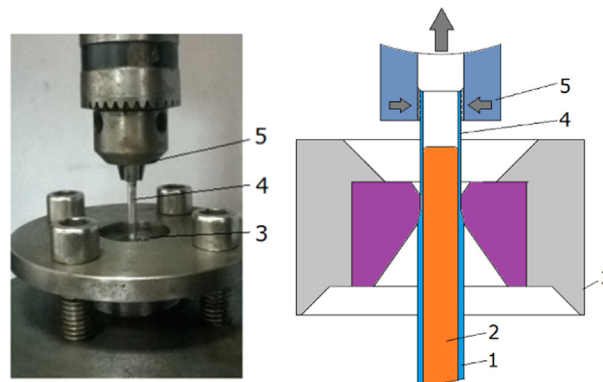


Fig. 4. Cold drawing of magnesium alloy tubes. 1: tubular billet, 2: mandrel, 3: inlaying die, 4: drawn tube, and 5: gripper.

Table 1  
Dimensions of drawing tools and area reductions of microtubes.

| Pass no. | Diameter (mm) |         | Area reduction (%) |
|----------|---------------|---------|--------------------|
|          | Die           | Mandrel |                    |
| 1        | 3.05          | 2.50    | 4.6                |
| 2        | 3.00          | 2.50    | 9.9                |
| 3        | 2.95          | 2.50    | 10.8               |
| 4        | 2.90          | 2.50    | 11.9               |

#### 3.2. Experimental procedures

Indirect extrusion to produce thin-wall tubes was performed at a temperature of 480 °C, an extrusion speed (i.e. the mandrel speed) of 0.13 mm/s and a reduction ratio of 50. The container was heated to the pre-set temperature and held for 10 min to ensure that the billet inside the container reached the same temperature.

Extruded tubes with an outside diameter of 3.16 mm and an insider diameter of 2.66 mm were cold drawn by using the drawing tooling fixed on a universal mechanical testing machine so that accurate displacement and force data could be recorded during the drawing process (Fig. 4). The dimensions of the mandrels and dies used at four individual passes in the cold drawing process are listed in Table 1. To ensure the success of each drawing pass, reduction ratio in cross-section area was limited to 12%. Machine oil, recommended by Hanada et al. [3] after comparison with other lubricants in cold drawing, was used to decrease the drawing force in the experiments.

Cold-drawn tubes at each pass as well as the as-extruded tubes were cut on the cross section and hot-mounted in an epoxy resin matrix. After grinding, polishing and etching in a picric acid-based etchant, the microstructures of the samples were observed using a Zeiss Axio Scope.A1 optical microscope. The accuracies of microtube dimensions were reassured with the aid of the micrographs taken at low power (a magnification of 50).

### 4. Results and discussions

#### 4.1. Forces at each forming step

A typical force–mandrel displacement diagram during hot indirect extrusion to produce a thin-wall tube is shown in

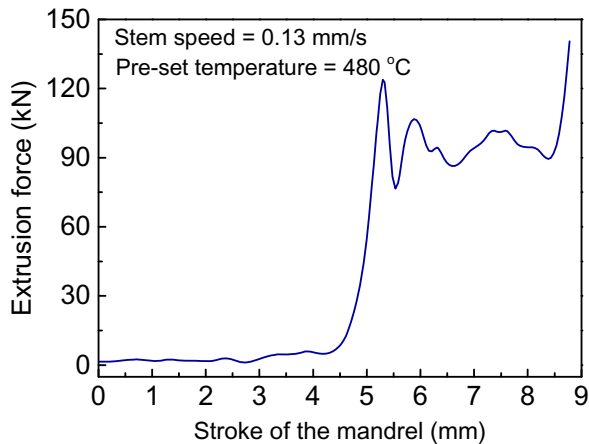


Fig. 5. Variation of force with mandrel stroke during hot indirect extrusion to produce a magnesium alloy tube.

Fig. 5. The extrusion force increased significantly when the billet was upset in the container and the material started to flow through the die bearing. After reaching the peak value of 130 kN, the extrusion force oscillated between 80 and 120 kN. During this period, the tube was formed. At the end of mandrel displacement, the container touched the front platen and the force increased abruptly and significantly.

To extrude microtubes with even smaller dimensions, the extrusion force requirement would become extremely high and a much more powerful hydraulic system would be needed to drive the container and mandrel forward. This would put the mandrel at risk, because during extrusion the mandrel was a highly vulnerable tool and prone to break, due to strong friction forces arising from the flowing billet material and likely due to bending forces as a result of any slight misalignment.

Multi-pass cold drawing to bring the extruded tubes to the near-net dimensions of stents needed much lower forming forces than hot indirect extrusion (Fig. 6). The forming force was only 10 N at the first pass, as a result of a relatively low reduction ratio chosen. Actually, the first pass was used primarily to straighten the as-extruded tube. During cold drawing at the second and third passes, the diameter of the die orifice decreased step by step and the forming force increased to 50 and 150 N. As a result of the accumulation of work hardening at the previous passes, the plasticity of the material became progressively lower and the force required at the fourth pass exceeded 300 N.

#### 4.2. Dimensional accuracies of drawn tubes at each pass

The microtubes after cold drawing at different passes are shown in Fig. 7. Towards the fourth pass, both the outside diameters and wall thicknesses of the tubes decreased. In addition, as will be shown later, the wall thickness uniformity improved along with the progressive drawing passes.

Fig. 8 shows the average values of the wall thicknesses of tubes after indirect extrusion and multi-pass cold drawing, as well as their standard deviations. It is clear that after hot

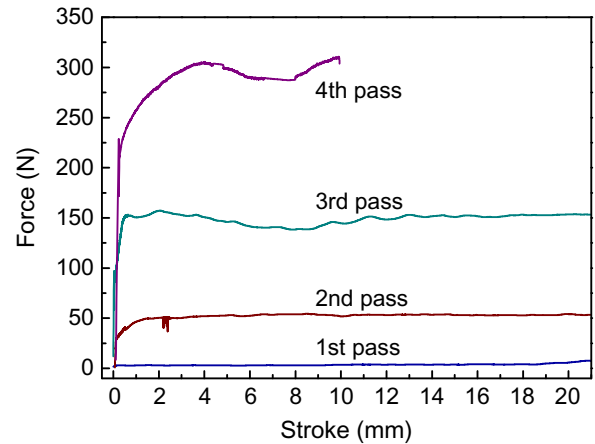


Fig. 6. Variations of force during multi-pass cold drawing to produce magnesium alloy microtubes.

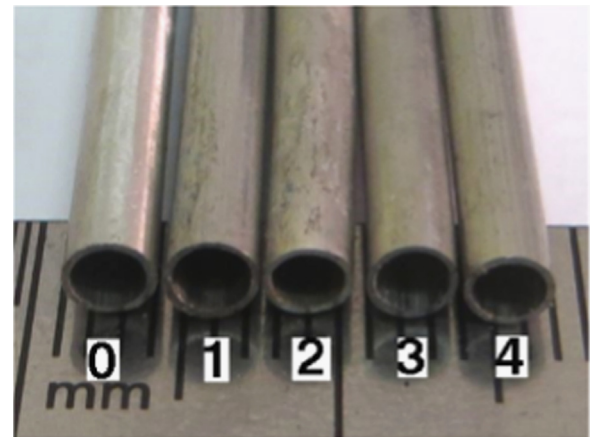


Fig. 7. Extruded tube and drawn tubes (0: as-extruded tube, 1: after the first pass, 2: after the second pass, 3: after the third pass, and 4: after the fourth pass).

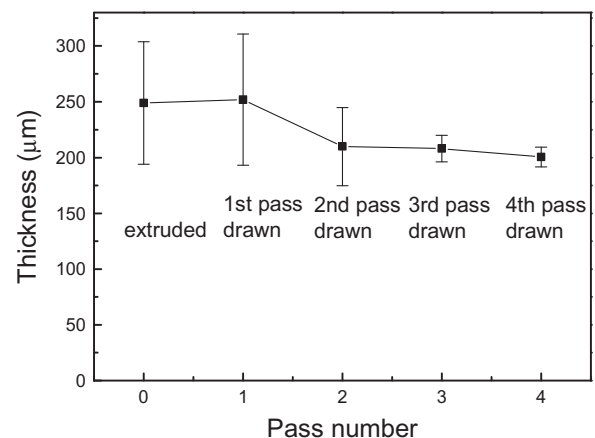


Fig. 8. Changes of tube wall thickness along with multi-pass drawing. Extruded and drawn tubes.

extrusion, the wall thickness varied considerably. The wall thickness of the extruded tube had the largest value of 304.3  $\mu\text{m}$ , while the thinnest part had a wall thickness of only

194.5  $\mu\text{m}$ . Even after the first pass of cold drawing, the significant variation in wall thickness remained. At the subsequent drawing passes, the material at the thicker region flowed into the thinner region. The uniformity of wall thickness was obviously improved at the last three drawing passes. After the fourth pass, the average wall thickness of the drawn tube was 200.5  $\mu\text{m}$  and the standard deviation was 8.9  $\mu\text{m}$ , which was only 16.14% of the value before cold drawing. The inside and outside diameters of drawn tubes also exhibited the similar tendency with the final diameters close to the ideal dimensions for stents as indicated by the dash lines in Fig. 9.

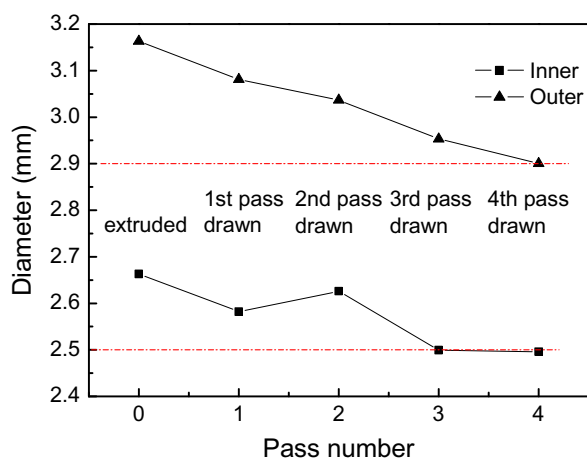


Fig. 9. Changes of the inside and outside diameters of tubes along with multi-pass drawing.

### 4.3. Microstructure evolution

To fulfill the requirements of the mechanical performance of vascular stents, represented by strength, ductility and fatigue resistance, the control of the microstructure along the material processing route holds the key.

The as-deformed microstructures of the tubes after indirect extrusion and cold drawing at selected passes are shown in Fig. 10. The as-extruded material had the typical necklace-like structure with small and equiaxial grains distributed around the former larger grains (Fig. 10a), indicating that partial dynamic recrystallization (DRX) took place in the pre-extruded microstructure. Throughout four-pass drawing, the average grain sizes of the tubes remained almost unchanged. The average size of grains was about 10  $\mu\text{m}$  with variations over wide ranges. The sizes of large un-recrystallized grains were around 20  $\mu\text{m}$ , while recrystallized ones were as fine as 2  $\mu\text{m}$ . Over a wall thickness of 200  $\mu\text{m}$ , the number of grains across the strut of a typical stent was between 15 and 30, being far larger than the minimum value of three, as reported in Ref. [17].

The poor plasticity of magnesium due to the absence of sufficient slip systems in the HCP-structured materials promoted the generation of twins during cold drawing. However, little evidence of twinning was observed after the first and second passes of cold drawing, probably the amount of plastic deformation was yet too moderate to trigger the formation of twins (Fig. 10b). Twinning became visible only at the third pass of cold drawing (Fig. 10c). At this pass, the wall thickness uniformity became improved and large plastic deformation was exerted at the surface layers of the tubes where twinning occurred.

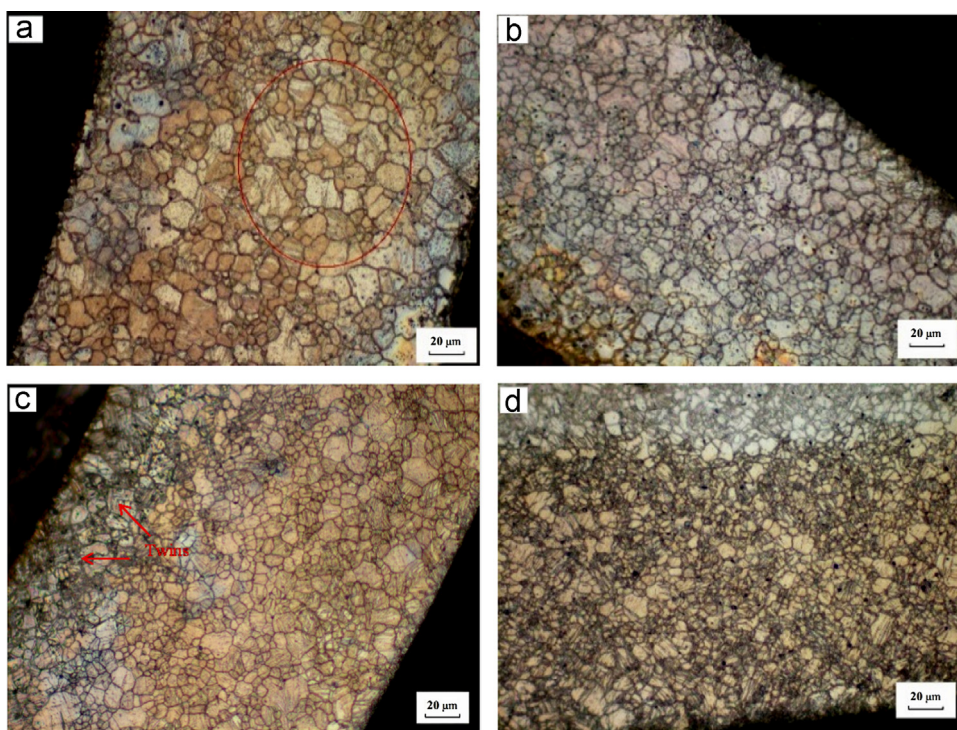


Fig. 10. Cross-section microstructures of microtubes after: (a) hot indirect extrusion; (b) two-pass drawing; (c) three-pass drawing and (d) four-pass drawing.

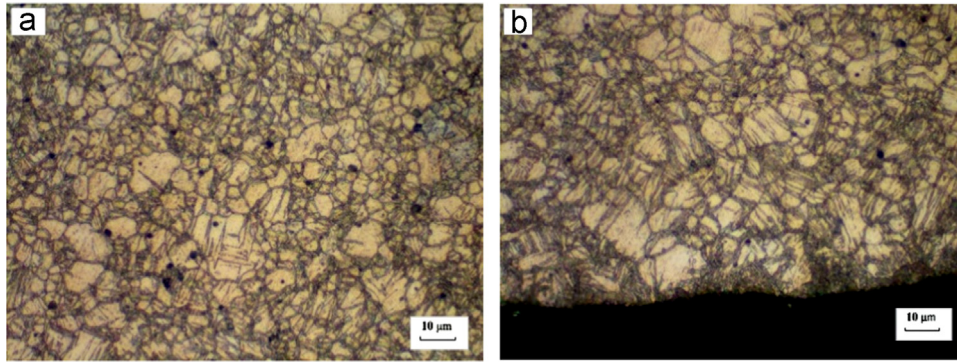


Fig. 11. Microstructures of microtubes after four-pass drawing: (a) in the central part and (b) near the outer surface.

As compared with our previous work on the ZM21 magnesium alloy [15], the drawn tubes in the present research had a much smaller number of twins. Accordingly, the drawing force was lower, probably due to the higher extrusion temperature used, which would lead to the formation of a more complete recrystallized microstructure during indirect extrusion. Better mechanical properties could therefore be expected.

The accumulation of work hardening in the form of generating twins and dislocations increased the difficulties for further deformation. As can be seen in Fig. 6, the drawing force at the fourth pass almost doubled the value at the third pass. Grains near the outer surface as well as in the central part of the tube contained a very large number of twins, as shown in Fig. 10d.

More detailed microstructures after cold drawing at the fourth pass are shown in Fig. 11. Twins emerged within the grains at the central part of the tube, especially within the larger ones. Moreover, the larger grains near the outer surface were divided into small grains by twins, while the small grains were full of twins and could hardly be distinguished. It implies that the forming force would have become unacceptably high and fracture might have occurred, if an additional drawing pass had been applied. Annealing, as an effective way to annihilate tangled dislocations and generated twins and to extend the forming limit, would have to be applied, if further drawing is needed, as demonstrated in the case of the ZM21 magnesium alloy [15].

## 5. Conclusions

A simple forming process route without involving a severe plastic deformation process was proposed to fabricate magnesium alloy seamless microtubes for vascular stents. The lab-scale hot extrusion press and cold drawing tooling were designed and manufactured for the proof of the concept. The ZM21 magnesium alloy was used as a representative material for biodegradable stents and subjected to hot and cold forming, including indirect extrusion and drawing to the dimensions of microtubes for stents. The dimensional accuracies and

microstructure of microtubes along the forming process route were examined. The following conclusions could be drawn:

- (1) Microtubes of the AZ21 magnesium alloy with an outside diameter of 2.9 mm and a wall thickness of 0.2 mm were successfully fabricated after hot indirect extrusion and four-pass cold drawing without inter-pass annealing.
- (2) Hot indirect extrusion was effective in refining the grain structure with DRX occurring during extrusion. However, the forming force was very high and the wall thickness uniformity of the extruded tubes was difficult to control.
- (3) Multi-pass cold drawing was an effective way to improve the dimensional accuracies and wall thickness uniformity of microtubes. However, the generation and accumulation of twins during the multi-pass drawing imposed limits to further drawing. Inter-pass annealing of the drawn tube after four-pass drawing would be necessary.

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