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The thermal behavior of aluminum 5083 alloys deformed by equal channel angular pressing

Jong Cheon Lee[a], Sang Hyun Lee[b,*], Sok Won Kim[c], Duck Young Hwang[d], Dong Hyuk Shin[d], Seoung Won Lee[a]

[a] Department of Nanomaterial Engineering, Chungnam National University, Daejeon 305-764, Republic of Korea
[b] Division of Physical Metrology, Korea Research Institute of Standards and Science, 1 Doryong-Dong, Yuseong-Gu, Daejeon 305-340, Republic of Korea
[c] Department of Physics, University of Ulsan, Ulsan 680-749, Republic of Korea
[d] Department of Metallurgy and Materials Science, Hanyang University, Ansan 465-791, Republic of Korea

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ABSTRACT

Equal channel angular pressing (ECAP) has an advantage of being capable of achieving a submicron grain size. The 5083 Al alloy is a typical non-heat treatable Al–Mg based alloy that possesses many interesting characteristics as a structural material. In this study, a 5083 aluminum alloy was processed by equal channel angular pressing at 473 K to produce an ultra-fine grain size of about 200 nm. Aluminum 5083 was deformed by ECAP following route Bc up to eight passes at 473 K. The grain size was changed from about 200 μm to 300 nm by deformation. The specific heat capacity was increased monotonically with temperature rise, however, in the high temperature region at about 800 K, the trend is slightly different and this is thought to be caused by the re-crystallization of the ECAP process or precipitation, and therefore, the specific heat capacity was influenced by the amount of energy release.

1. Introduction

To improve the strength and toughness of materials and the mechanical properties such as super-plasticity, studies on the miniaturization of grain to below a submicron size are currently underway. The methods of miniaturizing the grains of existing materials include heat treatment, power metallurgy, rapid solidification, and deformation processing but all these methods are incapable of miniaturizing grain to below a submicron size and are limited in the deformation caused by cross section reduction from an increase in processed quantity. As a method to overcome such problems, there is severe plastic deformation (SPD) that miniaturizes grains of material by causing severe plastic deformation within the material. SPD methods include high pressure torsion (HPT), ECAP, equal channel angular rolling (ECAR), and accumulative roll bonding (ARB). Among these methods, ECAP is a method that miniaturizes grains of bulk materials relatively easily by applying 90° of flexion to a die with same channels and causing transverse shear deformation [1,2]. The changes in the grain structure occurring within the material at this time are processed in four stages [2]. Stage 1 is the work hardening stage where dislocation is formed and intertwined in the various slip systems, causing an increase of dislocation velocity. Stage 2 generates grain-shaped cells by forming the dislocation network with the continuous increase of dislocation velocity. During this stage, the greater the processed quantity, the greater the dislocation velocity, and thus the size of the cells decrease. Stage 3 is the subgrain stage where dynamic recovery caused by the interactions of high density dislocation occurs and dislocation forms a stable low-angle grain boundary. In Stage 4, the low-angle grain boundary is converted to the high-angle grain boundary with an increase in dislocation velocity and ultra-fine grains that has the orientation difference of over 15°. Likely, materials processed by ECAP can achieve improvements in strength and toughness, with no decrease in sectional area and no occurrence of air space regardless of the number of processing. It certainly is a method that can attain low temperature super-plasticity by accelerating the formation of high-angle grain boundary.

DSC, which is a very useful method in measuring phase transformation of material, latent heat, specific heat, glass transition temperature, re-crystallization temperature, fusion point, energy of activation and many more, have been applied to various metals, ceramics and high-polymers. “The analysis on the behavior of the precipitates from the aluminum supersaturated magnesium” researched by Hamana et al. [3,4] is used in Al–Mg alloy.

* Corresponding author. Tel.: +82 42 868 5199; fax: +82 42 868 5022.
E-mail address: leesh@kriss.re.kr (S.H. Lee).

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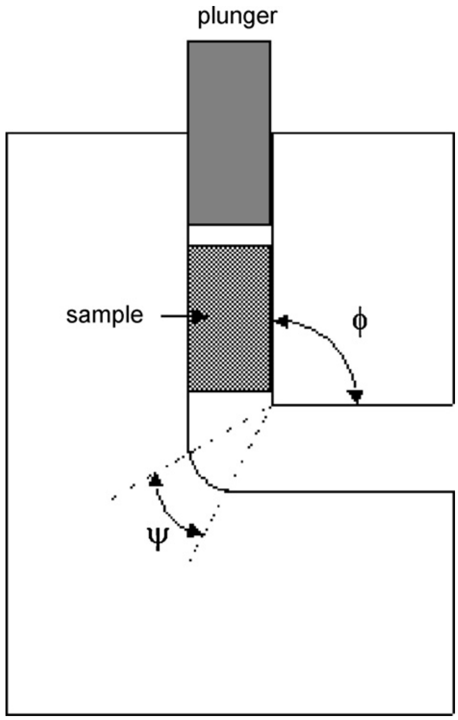

Fig. 1. Schematic illustration of ECA pressing die.

With the characteristics of low price, high strength, oxidation resistance, high formability, and low density, 5083 Al, which is a typical non-heat treatable alloy of the Al–Mg type, can replace the existing steel materials used in automobile planks and vessels, and thus it can have energy reduction effect by making them lighter. Since the studies conducted up until now on the commercial 5083 aluminum alloy processed by ECAP have focused only on the mechanical properties [5,6], this study has observed the energy accumulation within the structure of grain formation through the measurement of specific heat capacity, which is a typical characteristic of the physical properties within the materials processed by ECAP.

2. Experimental

The material used in this study is the commercial 5083 Al alloy extrudate and its chemical composition is 4.4 wt% Mg–0.7 wt% Mn–0.15 wt% Cr–Al. The extrudate is processed into a round bar specimen with a diameter of 17.5 mm and a length of 125 mm, and grain samples were obtained in the size of 200 μm by annealing the round bar specimen under 723 K for 1 h. Through the die having the channel angle (Φ) of 90° and the corner angle (Ψ) of 20° at the temperature of 473 K with the strain rate of 2 mm/s, the specimen was processed with one to eight passes via route Bc where the sample is rotated by 90° in the same sense between each pressing. To minimize the friction with the die, MoS_2 lubricant was applied to the specimen surface. The schematic illustration of the ECAP pressing die is shown in Fig. 1. Route A the sample is removed from the die and the pressing is repeated without any rotation of the sample. Route C the sample is rotated through 180° between each pressing. Route Bc repetitive pressing were performed on separate samples, under both A and C conditions, from an one to an eight pressings through the die [7]. According to the previous research results [8], it has been found that the greatest strain can be obtained at a single pressing when Φ is 90°.

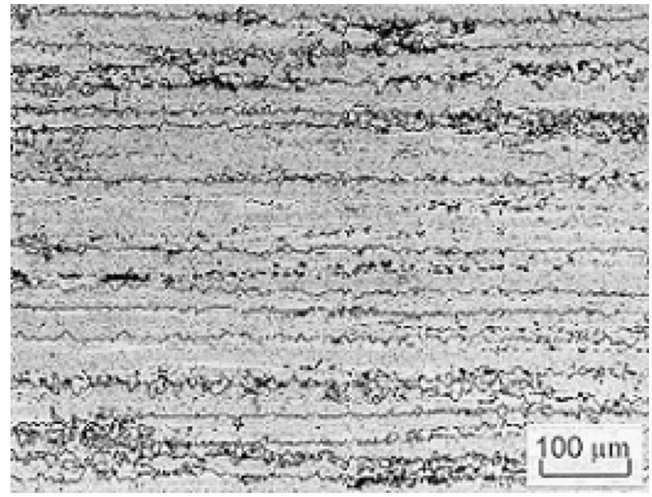

Fig. 2. Microstructure of as-received 5083 Al alloy by optical microscope.

In general, the effective strain (ε) accumulated in the specimen during the ECAP process is described in the following equation:

$$\varepsilon = N\left[\frac{2\cot[\Phi/2+\Psi/2]+\Psi\operatorname{cosec}[\Phi/2+\Psi/2]}{\sqrt{3}}\right] \tag{1}$$

Here, N is the number of passes through the die process of the specimen. The strain quantity based on die dimension can be easily obtained through Eq. (1), and when $\Phi = 90°$, the effective strain closest to around 1 can be obtained at one pass regardless of Ψ.

The specimen for the specific heat capacity measurement weighs about 100 mg with a diameter of 4 mm and a thickness of 2.5 mm. By using Perkin-Elmer (Pyris 1) for the low temperature range (100–298 K) and Netzsch (DSC 404C) for the high temperature range (298–823 K), the specific heat capacity was measured at the heating rate of 10 K/min with an argon flow of 50 L/min [9]. Also, JEOL 2010 was used as the transmission electron microscope (TEM) for the observation of the microstructure within the material and an acceleration voltage of 200 keV was used. For TEM observation, the specimen was put through jet polishing in nitric acid (25%) and methanol (75%) mixed solution under the voltage of 25 V. Also, the specimen was preprocessed using dilute Poulton's reagent for optical observation.

3. Results and discussion

3.1. Microstructure

Fig. 2 is an optical picture prior to the ECAP process and shows the grains in the size of 200 μm lining up in the tensile loading directions. In case of one pass as shown in Fig. 3(a), the subgrain bands parallel to the shear surface are formed and the size of the grain is 200 nm in width and 700 nm in length. After that, in two passes (Fig. 3(b)), the subgrain bands disappear and the equiaxed grain is formed in the size of about 500 nm. Looking at the SAED pattern, the diffraction pattern formed a ring pattern compared to one pass of the ECAP process. This shows that the angles between the grain boundaries form high-angle misorientation in polycrystals.

Accordingly, this signifies that the misorientation among grains has increased by the severe plastic deformation in the subgrains formed in two passes more than the subgrains formed in one pass, and also shows that the subgrains of low-angle grain boundary change to high-angle grain boundary with the increase in the number of passes.

In the case of four passes and eight passes shown in Fig. 3(c) and (d), the grain size was reduced to as small as 150 nm and have

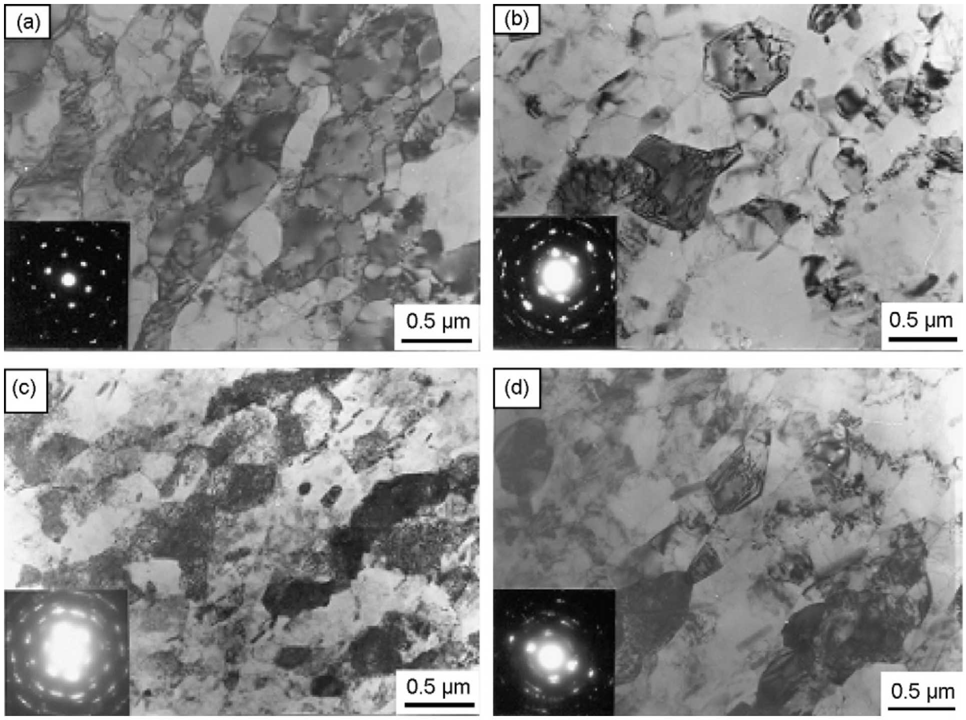

Fig. 3. TEM micrographs and corresponding SAED patterns of 5083 Al alloy pressed at 200, route Bc; one pass, (b) two passes, (c) four passes and (d) eight passes.

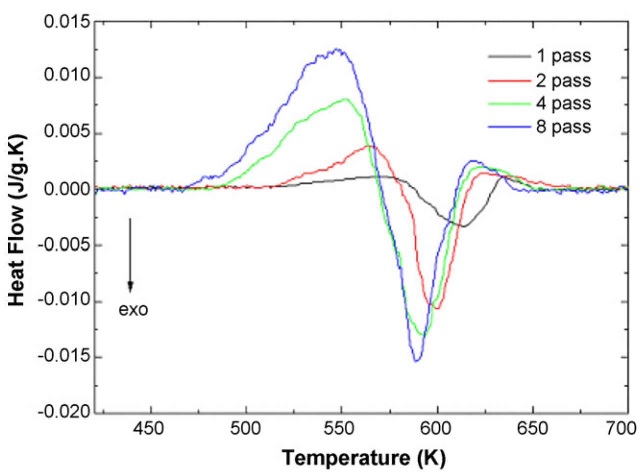

Fig. 4. Heat flow of ECAPed 5083 Al alloys.

obtained grains closer to the equiax than two passes. The SAED pattern in four passes and eight passes showed more vivid ring patterns compared to two passes, which signifies the formation of high-angle grain boundary with the increase in the number of passes. Nevertheless, the grain boundaries of four passes and eight passes, in comparison with one pass and two passes, are indefinite, which is caused by the lattice around the boundaries being severely twisted when the material receives severe plastic deformation. Also, these grain boundaries have extrinsic grain boundary dislocations of about 10^{15} m^{-2} from the doubling of grain boundary energy [10,11]. The non-equilibrium grain boundaries that have such high internal energy and long-range stress are known to have grain growth even at relatively low temperatures [10,11].

3.2. Thermal analysis

Fig. 4 shows the result of DSC analysis on the temperature range of 298–833 K in the heating rate of 10 K/min. It shows two endothermic peaks and one exothermic peak. The measurements of onset, peak temperature and heat of transition are shown in Table 1. There is no difference in peak temperature depending on the number of passes for two endothermic peaks and one exothermic peak but onset temperature and heat of transition showed big differences depending on the ECAP process. This is thought to be the formation of endothermic and exothermic reactions from crystallization and re-crystallization as grains are formed by the ECAP process. Al–Mg alloy is the precipitation sequence with GP zones (Guinier–Preston) → β″ → β′ → β [12]. β is equilib-

Table 1
Temperature and heat of fusion of the 5083 Al alloys by ECAP.

DSC scan	Number of ECAP	One pass	Two passes	Four passes	Eight passes
First endothermic	Onset temperature (K)	513.5	511.8	485.0	463.5
	Peak temperature (K)	567.7	564.3	551.8	544.8
	Heat of transition (J/g)	0.22	0.46	0.92	1.49
Second exothermic	Onset temperature (K)	598.8	578.3	571.0	553.4
	Peak temperature (K)	613.1	601.7	592.2	588.7
	Heat of transition (J/g)	−0.09	−0.30	−0.43	−0.61
Third endothermic	Onset temperature (K)	609.4	602.7	596.8	591.5
	Peak temperature (K)	633.4	625.6	621.7	615.3
	Heat of transition (J/g)	0.03	0.11	0.26	0.38

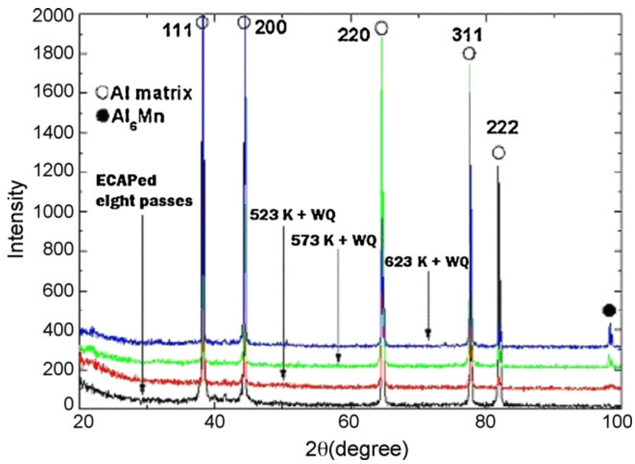

Fig. 5. XRD pattern of aluminum alloy by ECAPed depend on quenching condition.

rium phase (approximate composition Al_3Mg_2) having a complex f.c.c. structure. We are investigated temperature range from 450 to 640 K. This range has several endothermic and exothermic DSC peaks. Two exothermic of Al–Mg alloy are a 453–563 K of β′ formation, and 553–603 K of β′ → β. Two endothermic of Al–Mg alloy are a 473–573 K of β′ dissolution, and 603–703 K of β dissolution [13]. So our result of the first endothermic DSC peak was a β′ formation, the second peak was mixed of endothermic and exothermic because of β′ dissolution and transition of β′ → β. Third endothermic peak indicated β dissolution. We think the transition temperature decrease to by ECAP, but heat of fusion increase to ECAP passes. The 5083 Al alloy in the temperature range of from 523 to 723 K due to recovery and re-crystallization [14]. Fig. 5 shows the Al_6Mn precipitation appear to the heat treatment condition. So these temperature range of 5083 Al alloy were affected by precipitation of Al_3Mg, Al_6Mn, etc.

Each of the peaks in eight passes has been analyzed using TEM and XRD. First of all, looking at the XRD pattern in Fig. 5, the precipitation peak of Al_6Mn, which could not be found in the specimen of after as-ECAPed eight passes, are shown at 523, 572 and 650 K. This shows the precipitates of Al_6Mn phase at the first endothermic peak. Figs. 6 and 7 show the TEM picture of the specimen that is water-quenched at 523 K in the first endothermic peak interval. Fig. 6(a) is the picture of grains that have been unpolished into as great as 200 nm and shows relatively definite boundaries than the grain boundaries shown in Fig. 3(d). This is the result of reduction in boundary energy from the reduction in dislocation by thermal effect. Fig. 6(b) shows the precipitate with a diameter of 30 nm and a length of 100 nm and the EDS analysis result with the ratio of Al:Mn being 6:1. This proves that the precipitate of the peak analyzed using the XRD pattern in Fig. 5 is the precipitate of Al_6Mn, and such can also be seen in the previous research results [14,15].

Park et al. [14] were measured the micro-hardness for 5083 Al alloy with four passes ECAP, and it decreased rapid in temperature range of 473–523 K due to recovery and re-crystallization [14]. The first endothermic peak of four passes ECAPed sample was measured 485 K, so we think this affect by recovery and re-crystallization. The first endothermic peak of the specimen processed by eight passes ECAP is the result of repetitive reaction of recovery and precipitation. Fig. 7 shows Moiré and magnified fringes and this is closely related to the low-angle grain boundary. In general, it is below 4.0° but can be between 4.0° and 8.0° at time to time. Fig. 8 is the TEM result of the commercial 5083 aluminum alloy that has been water-quenched at 573 K. Fig. 8(a) shows the picture of grain that has grown to be as large as 500 nm and the grain in the size of 200 nm. Fig. 8(b) is the picture of the grain that has grown to be as large as 500 nm and it shows more precipitates than the precipitates shown in Fig. 5. This shows that precipitation occurs even at 573 K, which is the temperature in the range of exothermic peak. The opening temperature of exothermic peak is reduced from 598.8 to 543.3 K as the number of passes increased and ΔH increased. This is closely related to the strain energy accumulated within the material as the number of passes increased. The dislocation formed during the severe plastic deformation increases the strain energy and this can be expressed in the following equation [16,17]:

$$E_{\text{stored}} = Gb^2 \frac{\rho}{4\pi\kappa} \ln\left(b\sqrt{\rho}\right) \quad (2)$$

where G is shear modulus, b is Burgers vector, κ is arithmetic average of 1 and $(1-\upsilon)$, ρ is dislocation density and υ is Poisson ratio.

Eq. (2) is controlled by $\sqrt{\rho}$. Since the dislocation velocity increases as the number of passes increases, the strain energy

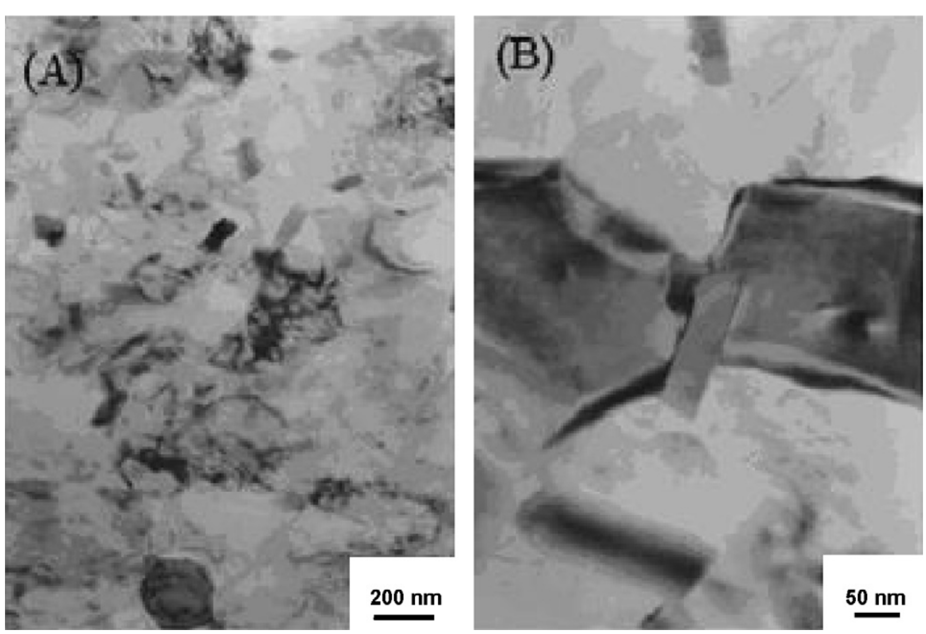

Fig. 6. Microstructure of ECAPed 5083 Al alloys at 523 K.

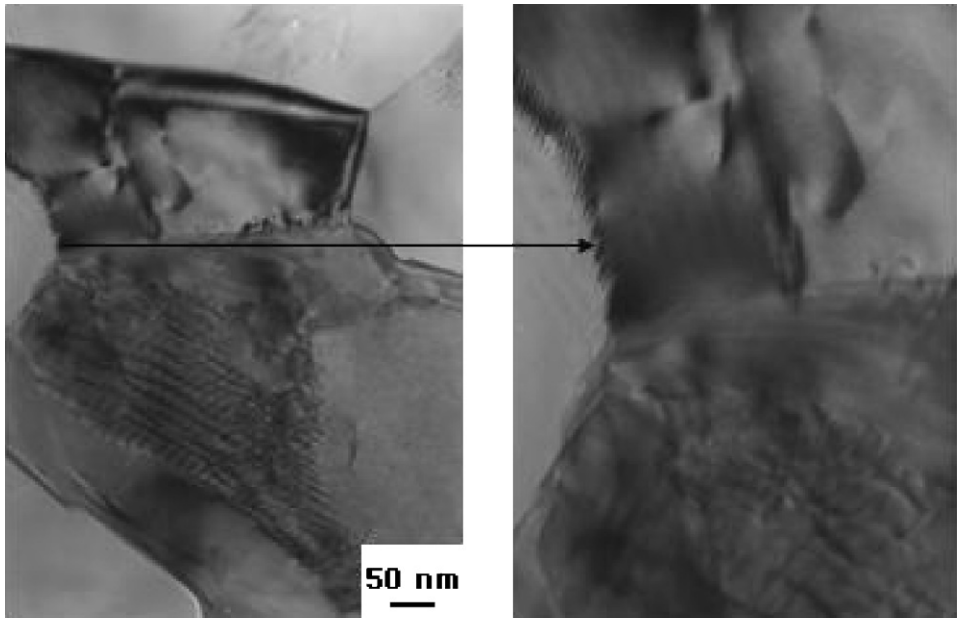

Fig. 7. Microstructure of ECAPed 5083 Al alloys at 523 K that indicates the Moiré and magnified fringes.

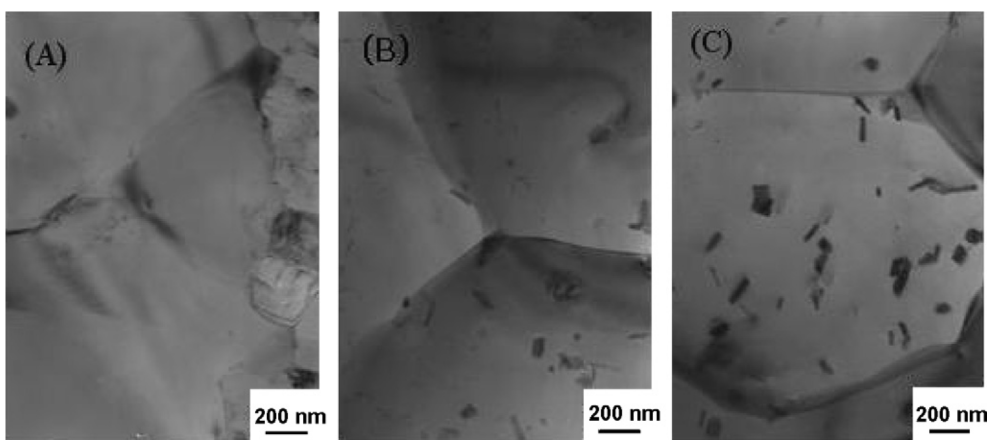

Fig. 8. Microstructure of ECAPed 5083 Al alloys at 573 K.

accumulated within the material is eight passes > four passes > two passes > one pass. Therefore, the heating value shown in eight passes from the thermal analysis result is 0.6144 J/g, which is greater than 0.0908 J/g of one pass.

As for the ECAPed commercial 5083 aluminum alloy, the endothermic peak and the exothermic peak move to low temperatures as the number of processing increases. This is the result of reduction in the temperature of recovery and re-crystallization when the quantity of dislocation generated during deformation is increased while increasing the driving force necessary in nucleation. The biggest problem in processing materials by ECAP process is the thermal instability. The dislocations that increased during the deformation reduce the thermal stability of materials and cause re-crystallization at a relatively low temperature. In the previous research results, Sc and Zr were added to complement such thermal instability to form fine precipitate particles. Sc and Zr combine with a matrix phase of aluminum and form nano-size Al_3Sc and Al_3Zr precipitates, and show coherent or semi-coherent relation with the α-Al matrix with an excellent high temperature safety. Also, Al_3Sc and Al_3Zr precipitates are distributed within the grains and restrain grain growth and prevent structure coarsening by hindering the movement of grain boundaries at high temperature [5,6].

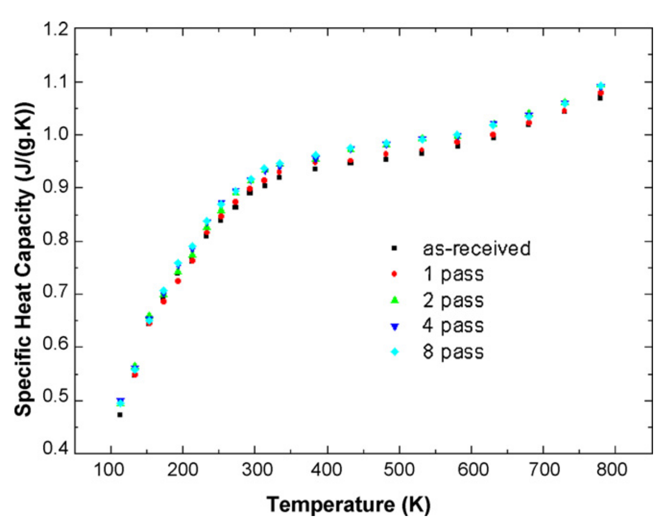

Fig. 9. Specific heat capacity of 5083 Al alloys fabricated by ECAP from 100 to 800 K.

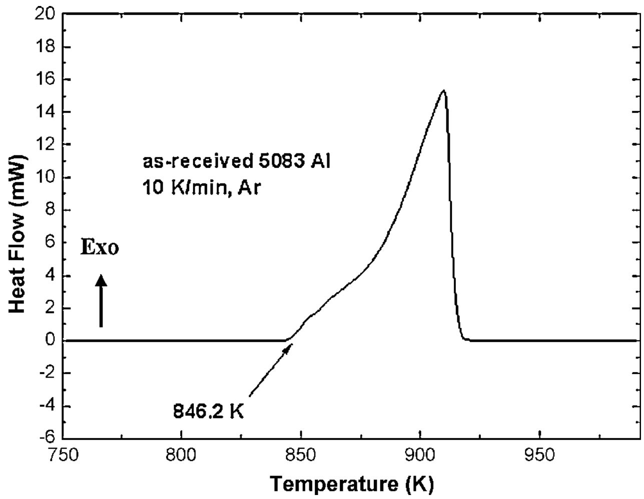

Fig. 10. The differential scanning calorimetry profile of the as-received 5083 Al alloy showing the occurrence of a partial melting at 846.2 K.

The specific heat capacity was measured at temperatures from 100 to 800 K as shown in Fig. 9. The specific heat capacity for all samples increased monotonically with temperature. The specific heat capacity of below 300 K region shows no difference by ECAP, but it increases with ECAP pass in the high temperature region. These differences are correlated with energy status of Fig. 4. Above 550 K, it was dependent on the crystallization and re-crystallization by the ECAP process.

Lee et al. [6] show the grain size as a function of annealing experiment on ECAPed Al–3Mg. When Sc is added, the grains remain ultra-fine without any grain growth up to 600 K but without the addition of Sc or Zr, grain growth occurs even at 500 K [6]. Such a result is clearly shown also in the melting point measurement of 5083 aluminum alloy using DSC. Fig. 10 is the opening temperature for the melting of 5083 aluminum alloy without Sc and is about 10 K lower than 857 K [14] measured by Park, thus can be concluded that Sc or Zr is an element that contributes to the securing of thermal stability in the Al–Mg type.

4. Conclusion

This study has observed the changes in the characteristics from grain atomization through the structural and thermal analysis on the ECAPed 5083 aluminum alloy. In accordance with the number of ECAP passes, 1st endothermic peak, 2nd exothermic peak and 3rd endothermic peak occurred, and the first endothermic peak was generated by the precipitates of Al_6Mn. The second exothermic peak also showed the process of precipitation. The scanning of the ultra-fine commercial 5083 aluminum alloy formed by ECAP process from 298 to 723 K showed a reduction in the formation temperature of recovery, re-crystallization and precipitation from the difference in deformation amount as the number of passes increased.

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