



## Effect of Thermally Formed Alumina on Density of AlMgSi Alloys Extrudate Recycled Via Solid State Technique

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### ABSTRACT

Solid state recycling of aluminium via hot extrusion is a sustainable technique. The process performed before hot extrusion plays a vital role on the extrudate properties. In this study, the effect of naturally and thermally formed in-situ alumina on the extrudate density were investigated. Four types of material identified as a solid as-received, non-treated recycle chip, 300 °C thermally treated recycle chip and 500 °C thermally treated recycle chip were prepared for the experiment. Prior to extrusion, the recycle chips were compacted into a chip-based billet, preheated and immediately extruded into a semi-finished product. The density test performed on the chip-based extrudate found that the type of chips influenced the density. The chip-based extrudate made of 300 °C and 500 °C thermally treated chips resulted in higher density than solid as-received and non-treated chips. Chip-based extrudate produced from 500 °C thermally treated chips resulted in density of 2724 kg/m<sup>3</sup>, which is the highest among the specimen. This density value was 0.7 % higher compared to the solid as-received extrudate. Microstructure examination on the cross-section revealed the alumina entrapped in the chip-based extrudate. The alumina entrapped in 500 °C thermally treated chips specimen was more prone than the non-treated and 300 °C thermally-treated chips. This finding explains the variation in the extrudate density.

## 1. Introduction

In recent years, there has been an increasing trend in research works that are focused on the issue related to sustainable manufacturing. The study includes recycling of aluminium chips using a solid-state technique, in which the process is performed in a melt less process [1–5]. The process used 95 % less energy than the conventional technique [6], which conserve natural resources and a safe environment due to less greenhouse gas emission. Without melting and casting, the process required less processing stage as shown in Figure 1. The process transforms the recycled chips into a

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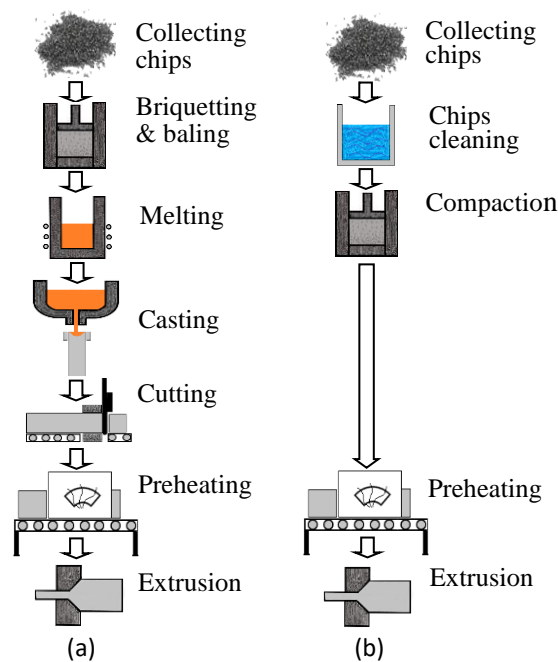
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semi-finished product directly through a severe deformation process. In comparison to the conventional technique, the process can transform the aluminium chips in a short cycle time, less workforce, and cost-saving.

Recycling aluminium via solid-state technique can be performed using various types of severe deformation processes. To this date, the deformation process such as forging [7–9], rolling [10,11] and extrusion [12–14] have been applied in previous research works. It has been claimed in a majority of research studies that the method is feasible in transforming recycled chips into semi-finished products. Moreover, the mechanical properties of the product made from recycling chips are also comparable to the as-cast billet extrudate.

The extrusion process transformed the recycling chips into a fixed cross-sectional profile extrudate. In this process, sufficient temperature and pressure are necessary for providing the required stress to deform the chips severely. The deformation causes the chip to shear, disrupts, and breaks the in-situ alumina layer that naturally forms on the chip surface into the ultrafine particle [15]. The fragmentation of the alumina layer removes the chip consolidation barriers, allowing contact between the fresh aluminium and consolidating the chip into the bulk. The entrapped ultrafine alumina reinforced the aluminium matrix, causing lattice distortion which impedes the dislocation motion and increases the strength of the material. With increasing the amount of alumina reinforcement in the matrix, the strength of the extrudate increased.



**Fig. 1.** Aluminium recycling technique (a) conventional (b) direct recycling [8]

Mixing the chips with other reinforcement material could enhance the aluminium alloys properties [16]. Lajis *et al.*, [4] reported that mixing the chip with alumina powder significantly affected the extrudate's strength. They found that adding alumina powder as much as 2 % into the chips has improved the extrudate's strength. However, too much alumina powder reduces the strength and ductility of the extrudate. Apart from weight fraction, the size of alumina powder and aluminium chips also affected the extrudate properties [17].

The usage of alumina powder as reinforcement material in direct recycling of aluminium chips has the drawback. The alumina powder tends to agglomerate, which weakened the chips

consolidation and thus worsened the extrudate properties. Furthermore, the powders and the chips have no bond, limiting the amount of powders sticks on the chip surface. Most of the powders will precipitate on the bottom side as it is small compared to the chip. It means that increasing the powder's weight fraction will not represent the real weight fraction entrapped in the extrudate. This problem is also affecting the uniformity of the distribution of the powder. The alumina and the chip may need to be bonded together before extruded in getting this problem solved.

Alternatively, the formation of alumina layer on the chip surface itself can be used as reinforced material. The reaction of aluminium with atmospheric oxygen will naturally generate thin film of alumina on the surface about 4 nanometers [18,19]. The alumina layer could be thermally enriched in-situ by employing thermal treatment. Depending on thermal treatment temperature, the treatment could result in alumina layer with a thickness of up to 50 nanometers. Besides, the treatment could also soften the chips, allowing the cold compaction to be performed at a much lower pressure than the compaction of non-treated chips

Since the hot extrusion on compacted billet can disperse the alumina layer into the fine particle, the effect of thermally form alumina on the chip is expected to have the same impact as the addition of alumina powder. However, until today, research investigating this technique is rarely reported. Therefore, in this work, the effect of the thermal-treated chips on the extrudate density was investigated. The variation in density will indicate the impact of the thermal treatment on the extrudate's physical properties.

## 2. Methodology

In this work, the machining chips were produced by face milling of AA6061-T650 aluminium alloys block using MAZAK CNC machine. Table 1 shows the composition of the aluminium alloys. The cutting was operated with side cut end mill using a cutting tool of 10 mm in diameter, 345 mm per second cutting speed, 1mm feed per tooth, and 1 mm depth of cut. The oil-based coolant was employed to control the machining temperature. The chips undergo acetone cleaning in an ultrasonic bath to remove the coolant and other contaminants. The chips were then dried in an oven at 100 °C for 1 hour to burnt out the acetone, water, and providing a clean chip surface.

**Table 1**

AA6061-T650 chemical composition

| Element | Si   | Fe   | Cu   | Mn   | Mg   | Cr   | Zn   | Ti   | Al   |
|---------|------|------|------|------|------|------|------|------|------|
| Wt (%)  | 0.67 | 0.52 | 0.28 | 0.02 | 1.10 | 0.19 | 0.01 | 0.02 | Rest |

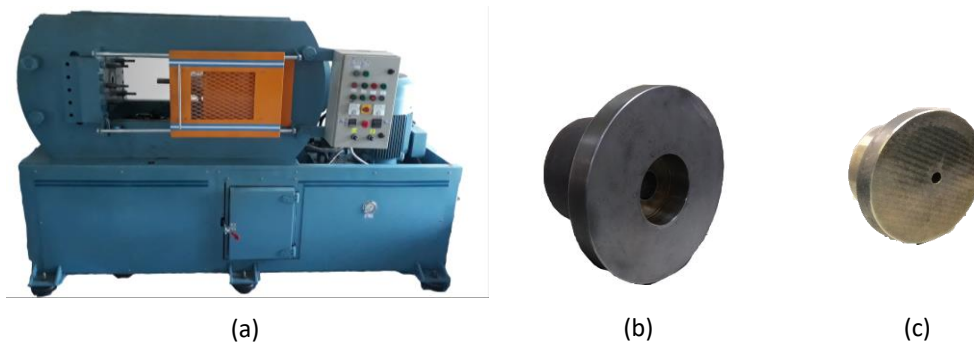
Four types of extrudate materials were prepared and labelled accordingly as shown in Table 2. The materials are solid as received, non-treated chip (NT), chips thermal treated at 300 °C (TT300) and chip treated at 500 °C (TT500). A solid as-received of AA6061-T650 was extruded for comparison with extrudate made from recycled chips. The TT300 and TT500 were thermally treated in the induction furnace for 1 hour. The chips were immediately cooled down naturally to ambient temperature after reaching the treatment time. The chips were then filled into the cylindrical cavity mould 30 mm in diameter and 100 mm in length. A press machine was used to compact the chips into a compacted chip billet for ease of handling. During the compaction process, the weight of the chips and compression force was controlled to produced a simmlar density for all billets of the compacted chip.

**Table 2**

Extrudate labels

| Extrudate material | As-received | Non-treated chip | Thermal-treated chip at 300 °C | Thermal-treated chip at 500 °C |
|--------------------|-------------|------------------|--------------------------------|--------------------------------|
| Specimen label     | AR          | NT               | TT300                          | TT500                          |

Prior to hot extrusion, the compacted-chip billets were preheated to 550 °C for 1 hour in the induction furnace. The billet is then immediately taken out from the furnace and extruded using a 300 tones horizontal press machine at a speed of 3 mm/s. The billet was driven by the plunger to pass through the flat face cylindrical orifice die with the extrusion ratio of 12 and die temperature of 350 °C. Figure 2 show the extrusion machine and the die used in this work.



**Fig. 2.** Extrusion equipment (a) Extrusion machine (b) Die container (c) Flat face die

The extrudate billet was cut off just after the extrusion was completed and naturally cool instantly to ambient temperature. A solid as-received of AA6061-T650 was also extruded for comparison. Figure 3 shows the chips, the billet of compacted chips and the extrudate of the recycled chip. Without additional heat treatment, the extrudate was cut into a sample parallel to the extrusion direction. The sample was ground by alumina sandpaper and polished by cloth associated with a various diamond polishers. The sample was etched by Baker reagent and examined using an optical microscope (OPM). The density of the extrudate samples was measured based on Archimedes's principles.



**Fig. 3.** Recycle chips and the products

### 3. Result

The density of the extrudates made from the different chip is depicted in Figure 4. The results showed that material resulted in different density value. For all extrudates, the density values were in the range of  $2704 \text{ kg/m}^3 - 2724 \text{ kg/m}^3$ . The extrudate of AR has a density of  $2704 \text{ kg/m}^3$ . All extrudate made from recycling chips has a density higher than the AR. The extrudate of TT500 has a density of  $2724 \text{ kg/m}^3$ , the highest among the sample. The density was about 0.7 % higher compared to the AR sample.

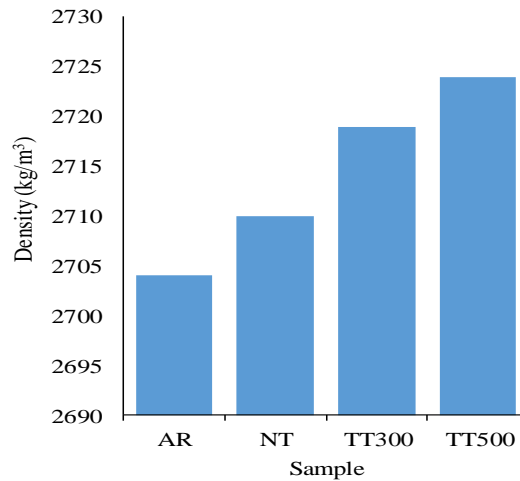


Fig. 4. Density of extrudate

Meanwhile, the extrudate density of the thermal-treated chip was higher than the extrudate made of non-treated chips. The NT sample has a density of  $2710 \text{ kg/m}^3$ . The density rose  $5 \text{ kg/m}^3$  to  $2715 \text{ kg/m}^3$  when the chips were thermal-treated at  $300 \text{ }^\circ\text{C}$ . Increasing the treatment temperature to  $500 \text{ }^\circ\text{C}$  caused the density gain further to  $2724 \text{ kg/m}^3$ . The density increment indicated that the density of the extrudate relied on the chip treatment temperature.

Microstructure examination on the extrudate cross-section of AR, NT and TT500 was depicted in Figure 5. For all samples, it can be observed that the fine precipitates of  $\text{Mg}_2\text{Si}$  and  $\text{FeAlSi}$  were the primary substance observed in the microstructure. The  $\text{Mg}_2\text{Si}$  and  $\text{FeAlSi}$  could be identified from the black and white colors, respectively. This finding was similar to the study by Voort *et al.*, [20]. The AR sample consists of coarse  $\text{Mg}_2\text{Si}$  and  $\text{FeAlSi}$  while the NT and TT500 consist of mixed coarse and fine  $\text{Mg}_2\text{Si}$  and  $\text{FeAlSi}$ .

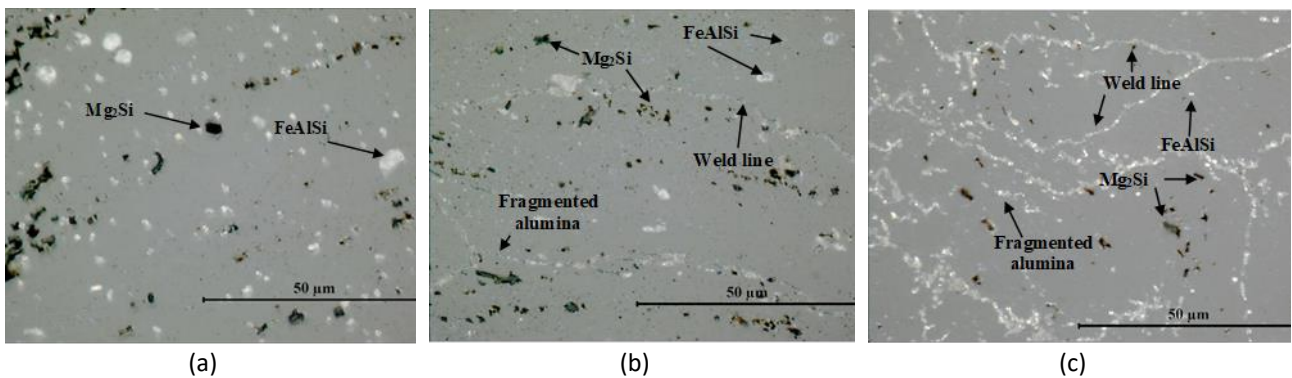


Fig. 5. Extrudate microstructure (a) AR (b) NT (c) TT500

The AR microstructure revealed no chip weld line since it was extruded from solid as-received. The NT and TT500 clearly showed chip weld lines in the microstructure. The chip weld line was formed due to chip consolidation. Besides, the chips consolidation in NT and TT500 were sound good, in which no micropore and crack existed in the microstructure.

Meanwhile, the fragmented alumina layer was also observed entrapped in the aluminium matrix of all chip-based extrudate. The NT observed that the alumina was mainly entrapped on the chip weld line. In contrast with NT, the presence of alumina in the microstructure of the TT500 was more prone than the NT. The TT500 showed the alumina scattered in the entire microstructure, although some were also observed in the weld line.

#### 4. Discussion

The density variation of AR, NT, TT300, and TT500 extrudate could be explained by the presence of alumina in the aluminium matrix. Since the density of alumina is two times higher compared to aluminium alloys, any increment of the in-situ alumina on the surface of the chip due to thermal treatment would gain the density of the chip-based extrudate. During the hot extrusion, the plunger forced the billet of compacted chips to go through the die cylindrical orifice. The difference in diameter of the billet and the orifice provide high shear stress and severely deformed the compacted billet. The alumina layer that forms either naturally or thermally on the chip surface was severely fragmented into fine particles and entrapped in the aluminium matrix. The higher the amount of in-situ alumina formed on the surface would result in higher alumina entrapped in the chip-based extrudate.

The NT sample in which the alumina was naturally formed has a density lower than the thermally formed alumina (TT300 and TT500). The results indicated that the chip treatment temperature played an important role when recycling thermally treated chips. The thermal treatment performed on the chips has enriched the in-situ alumina layer on the aluminium surface [21]. The enrichment of the alumina is depended on the chip treatment temperature. High treatment temperature produced thicker alumina layer. The density relies on the thickness of the in-situ alumina layer that forms on the surface. The thicker the alumina layer resulted in more alumina entrapped in the aluminium matrix. Therefore, the chips treated at high temperature resulted in higher extrudate density than the low-temperature treatment.

Entrapped alumina in the aluminium matrix could answer why the density of the extrudate made from recycling chip is higher than the original alloys. Theoretically, alumina has a density of  $3950 \text{ kg/m}^3$ , which is about one time higher than the aluminium. Therefore any small increment of the alumina in the matrix could significantly affect the extrudate density. Furthermore, the chips have also been cleaned by ultrasonic cleaning, which removed all contamination on the chips surface. Therefore, it is believed that the increment on the density of the TT300 and TT500 is solely due to alumina enrichment by the thermal treatment. The higher the amount of alumina enriched by the treatment resulted in higher extrudate density.

Instead of density, other researchers also claim that the presence of the alumina layer on the chip surface also affected the other properties. According to [10] and [22], the extrudate of recycling chip shows higher strength properties than the as-received. Therefore, entrapped alumina in the matrix is not a drawback, but it could be manipulated to improve the extrudate's mechanical properties.



#### 4. Conclusions

Direct hot extrusion is a sustainable technique in recycling aluminium chips. The density of the extrudate produced by the technique relies on the process before hot extrusion. In this experiment, the entrapped alumina was found to be the main reason affecting the variation of the density. Non-treated chips that have naturally form alumina was found to have extrudate density less than the thermally treated alumina layer. The alumina was naturally formed on the chip surface due to the reaction between the oxygen and aluminium. The thermal treatment performed on the chips has enriched the alumina. The higher the thermal treatment temperature, the thicker the alumina layer form on the surface, resulting in higher extrudate density. Although the thermal treatment resulted in alumina enrichment on the chips surface and increased the density of the extrudate, but it could be used to manipulate the strength of the alloys.

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