Compaction of aluminium foil and its effect on oxidation and recycling yield

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Abstract One of the problems when recycling aluminium is its oxidation and consequent metal loss. This is especially critical for the thin sheet/foil materials used for food packaging applications. Compacting the scrap into briquettes may partly reduce such losses in addition to facilitate transport and storage. Shredded aluminium materials of different thicknesses (15-300 μm) were compacted into cylindrical briquettes of 4 cm diameter, each weighting 20 g by uniaxial pressure or moderate-pressure-torsion. A sub-set of briquettes and chips was subsequently oxidized at 650 °C, while a sub-set was left untreated. Finally, all samples were re-melted under molten protective salt-flux. Compacting reduced the specific oxidation during the heat-treatment and promoted the coalescence and yield for the heat-treated materials. Both effects were most significant for the thinnest foil in the study (15 μm). The material thickness influenced the porosity and surface roughness of the resultant briquette, as well as the pressure required to reach a given bulk density.

Keywords Aluminium foil · Recycling · Compaction · Thickness

1 Introduction

About one-third of the aluminium (Al) currently produced is coming from recycling of either industrial or post-consumer waste, which brings forth substantial economic and environmental benefits [1]. Primary aluminium production is very energy intensive [2] [3] and of the overall energy that is utilized by the aluminium industry annually (7.6 Exajoules), the electrolysis process is by far the most energy intensive, using more than ten times the energy that is used to re-melt scrap (5.0 vs 0.4 Exajoules) [4]. One of the challenges during recycling is the scrap oxidation and consequent metal loss. The yield and quality of the metal obtained greatly depend on

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scrap properties, re-melting and pre-treatment methods. Xiao and Reuter showed in several studies the relevance of scrap type, size, surface conditions and cleanliness on the re-melting losses [5, 6]. One of the results especially relevant for this study was the observation of increasing re-melting losses with decreasing scrap size [7]. Rossel [8] investigated the effect of material thickness on re-melting, and showed that the dross formation was higher for the thinnest material (2 mm), especially for the alloys with high Mg content. Scrap with higher Mg content is generally more susceptible to oxidation [9, 10]. Despite the high reactivity of pure Al with oxygen, aluminium products present excellent corrosion resistance, thanks to the passivating effect of the oxide layer in most environments [11]. This, together with good barrier properties against gases, moisture and light, excellent malleability and formability, flexibility, surface resilience, and aesthetic properties makes it a popular choice for food packaging [12]. Of the total Al consumption in 2019 (almost 90 Mt), 8 % was for packaging applications [13]. Due to its short life-time, the recyclability of packaging is particularly relevant from a circular economy perspective. For example, assuming a Used Beverage Can (UBC) lifetime of 6 months and recovery and recycling rates of 97 %, the cumulative material losses would be 84 % after 10 years [14]. In order to overcome yield losses for thin scrap, compacting the scrap into denser pieces such as briquettes or bales, may be a solution. This is already an established practice for pre-consumed scrap (e.g. chips from extrusion and machining), since it facilitates storage and transport and prevents the small chips from floating when added into the molten Al pool [15]. Several authors have studied scrap compaction either as a re-melting pre-treatment step [16, 17] or for solid-state-recycling, which would allow using the compacted pieces directly without re-melting [18, 19, 20]. The aim of this study was to have a closer look at the compaction properties of thin Al materials, and to evaluate the role of briquetting in oxidation and re-melting losses for the different foil thicknesses. The materials studied were non-coated aluminium foil/sheet of 5 different thicknesses (15, 30, 100, 200 and 300 μm), and the compaction methods uniaxial, moderate-pressure torsion (MPT) and moderate-pressure torsion at 450 °C (Hot MPT). The results in this study were part of a Master Thesis carried out at NTNU by Philipson [22].

2 Experimental Materials and Procedure

Table 1 summarizes the thickness and chemical composition of the materials. Norsk Hydro provided the aluminium sheet alloy AA8006 in gauges of 100, 200 and 300 μm. The 15 μm foil is regular household foil, and the 30 μm a laboratory foil. Their compositions were measured with a portable XRF analyser with and error of $\pm 0.4^{*}$.

2.1 Shredding and briquetting

The foils were shredded using a Getecha RS 1600-A1.1.1 shredding machine, which operates with three rotary blades at a rotor revolution of 240 rpm. Depending on

Table 1 Composition and thickness of the materials studied

Foil gauge	Al	Fe	Si	Mg	Mn	Cu	Zn	Remain.
15μm	98.6±0.4*	0.73	0.53	<0.16				<0.05
30μm	98.9±0.4*	0.8	0.11	<0.17				<0.15
100, 200, 300 μm	95.9-98.5	1.2-2	0.4	0.10	0.3-1	0.3	0.10	<0.15

the thickness and stiffness of the material, the resulting chips differed in size and level of deformation. Two sieves (square mesh 5 and 2 mm^2) were used to unify the size, keeping only the chips that fell within this range, hence discarding 10-20 % wt. of the material. The average mass per chip after sieving was measured to be 6, 12.3, 12.4, 18.2 and 21.8 mg for the 15, 30, 100, 200 and 300 μm thick material respectively. The chips were subsequently compressed into cylindrical briquettes of 4 cm diameter, each weighting 20 g, using a hydraulic press MTS 311. Since the loose chips occupy more volume than the mould, they were added in several batches and pressed manually until the 20 g was able to fit into the mould space. The chips were compacted by uniaxial pressure, MPT and Hot MPT at 450 °C. The goal was to obtain briquettes pressed to a wide range of bulk densities, to evaluate both the material's compaction behaviour and the influence of the resultant bulk density, porosity and surface roughness on the re-melting yield. The internal porosity was characterized with a computed-tomography scan (CT), and the surface roughness with an optical 3D-microscope (Alicona 3D-microscope).

2.2 Thermal pre-treatment and re-melting

The aluminium was melted under protective salt-flux, a common procedure in industry for dealing with highly contaminated and oxidized scrap. The salt-flux protects the molten metal from further oxidation, captures the oxides and impurities and promotes the coalescence of the metal drops. [23] A subset of the chips and briquettes were directly re-melted, while another subset was first heat-treated at 650 °C for 1 hour in a Nabertherm electric furnace featured with air circulation, to promote oxidation, simulating a thermal de-coating pre-treatment. This process is sometimes applied industrially when recycling scrap containing organic coatings or contaminants [24]. The oxidation temperature was chosen based on the work by Capuzzi et al [23] that showed that by pre-treating the scrap at 600 °C, a complete de-coating was achieved that increased the coalescence of the metal droplets. The weight gain during heat-treatment was measured to evaluate whether oxidation is dependent on foil thickness and degree of compaction. The re-melting experiments were performed in ceramic crucibles (Al_2O_3-SiO_2) placed into a muffle furnace. The furnace was pre-heated, and the crucibles filled with the mixed salts (68.6 % $NaCl$, 29.4 % KCl and 2 % CaF_2) were introduced when the furnace temperature reached 800 °C. The melting point of a mixture of 30 % KCl and 70 % $NaCl$ is approximately 690 °C, and the small additions of CaF_2 increase it by 20-30 °C [25]. Although the furnace temperature was 800 °C when the crucibles were put inside, it took around 30 minutes for the salt to melt due to its low thermal conductivity, and some of the salt evaporated. Once
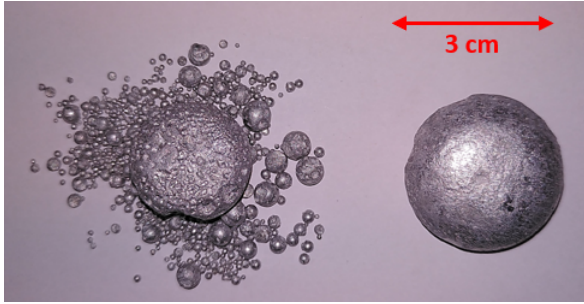
the salt melted, the briquettes were dropped into the crucibles using steel tongs. The briquettes were re-melted using 80 g of salt-flux (4:1 salt/metal ratio), whereas 150 g of salt-flux was used when re-melting the loose chips. Although this salt/metal ratios are far higher than the industrial, they were chosen so that the salt could completely cover the chips. The present re-melting laboratory set-up is static, without stirring, in contrast to the industrial rotary furnaces. The density of the melted salt-flux was approximately 1.5 g/cm^3 and it was observed that briquettes with lower bulk densities floated just below the surface, whereas the denser briquettes quickly sank into the crucible. After inserting the samples, the muffle furnace was closed and held at 800 °C for 10 minutes, and the crucibles were then taken out and cooled down at room temperature. The aluminium was recovered from the crucible by crushing and dissolving the solidified salt with water against an 800 μm mesh-size sieve. The Al-pieces with a smaller diameter than 800 μm were considered losses and not counted into the weight of the recovered material. During re-melting, typically most of the material coalesced into a main rounded-shaped piece and few small pieces (Figure 1). The percentage of input material that successfully melted into one piece was defined as coalesced recovery and it is as crucial as the metal yield in the industrial recycling operations, where the small pieces tend to remain trapped into the salt slag. The material yield and coalescence recovery were calculated using Equation 1 and 2:

$$Material\ Yield\ [\%] = (m_r/m_i) * 100 \tag{1}$$

$$Coalescence\ [\%] = (m_c/m_i) * 100 \tag{2}$$

Where m_r is the sum of the masses of the pieces (of diameter >800 μm) recovered, m_c is the mass of the biggest piece, and m_i is the initial mass of the briquette or batch of chips.
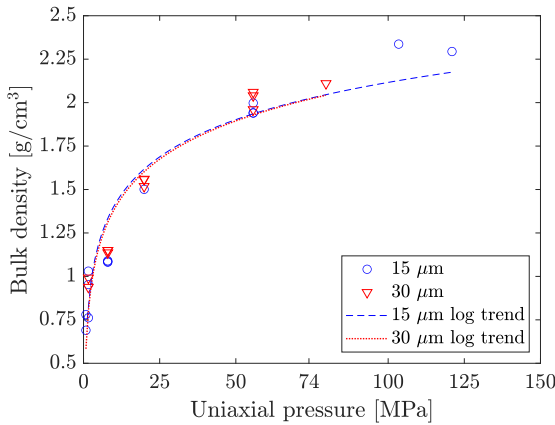
3 Results and Discussion

3.1 Compaction

Figure 2 compares the stress needed to compact the materials of different thickness to three density ranges. The material thickness influenced the compaction behaviour,
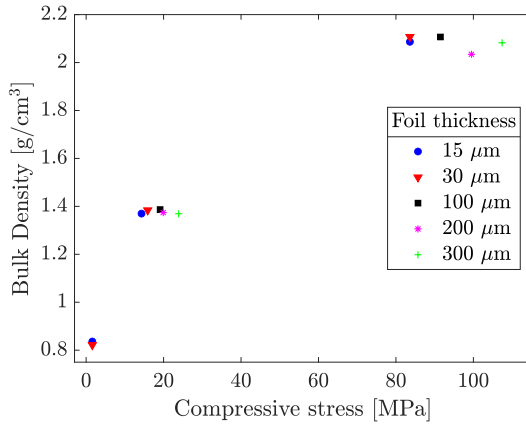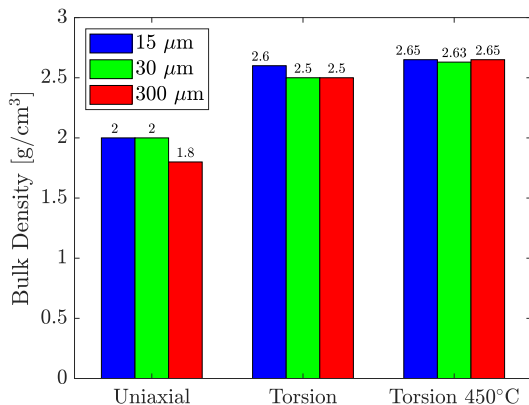
Fig. 2 Uniaxial stress vs. briquette bulk density for shredded chips of the materials of different thickness.

Fig. 3 Compressibility curve for the 15 and 30 μm gauge foils, showing a logarithmic behaviour in the uniaxial pressure vs. bulk density.

and the chips of thin foil were more easily compacted, while thicker materials demanded higher pressure. The lowest and highest achievable bulk densities also differed between materials. In Figure 3, compressibility curves for the 15 and 30 μm foils are shown. For these materials, it was possible to obtain briquettes with bulk densities as low as 0.8-0.9 g/cm^3 by applying 1.6 MPa (2 kN force). For increasing pressures, the bulk densities increase logarithmically until stabilizing at a plateau around 2.2-2.3 g/cm^3, just below the density of liquid aluminium. A similar plateau was observed in previous investigations [17][21]. For the 100, 200 and 300 μm sheets it was not possible to obtain densities lower than 1.35-1.4 g/cm^3 since the chips would not hold together. The comparison between the three compaction methods is plotted in Figure 4. Combining uniaxial pressure with torsion (MPT) proved to be a very effective compaction method, which allowed reaching densities above the values of the uniaxial plateau at relatively low uniaxial pressures. For example, just by turning the die 180°after applying 200 kN of force, a higher density was reached (2.69 g/cm^3) than for 500 kN uniaxially (2.58 g/cm^3) for the 300 μm material. Turning the

Fig. 4 Mean briquette bulk densities for the 15, 30 and 300 μm gauge materials obtained when compacted with uniaxial pressure (56 MPa), MPT (56 MPa + 4 turns) and Hot MPT (56 MPa + 4 turnings at 450 °C).
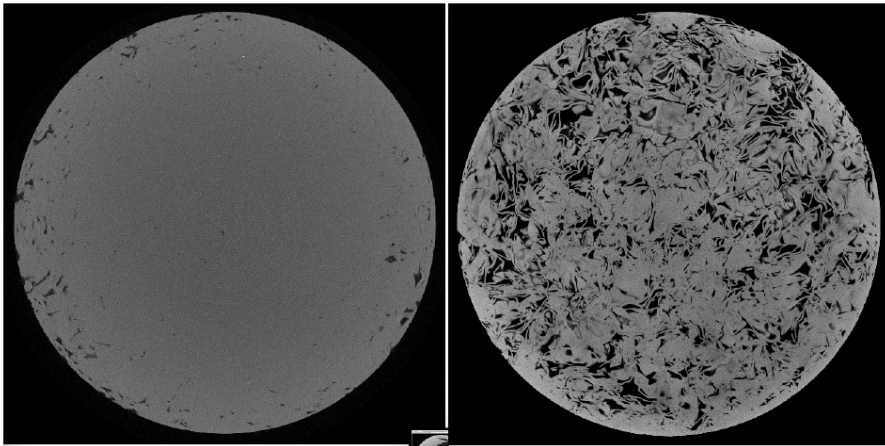
die 360°four times while compacting uniaxially at 56 MPa (70 kN), increased the bulk density 29, 24 and 37 % for the 15, 30 and 300 μm foils respectively, reaching densities around 2.5 g/cm^3. Finally, the Hot MPT method (450 °C) increased the bulk density by 33, 32 and 47 % for the 15, 30 and 300 μm materials, giving bulk densities above 2.6 g/cm^3, just below the density of solid aluminium (2.7 g/cm^3). The Hot MTP method aimed to simulate solid-state-recycling technologies, such as the screw-extrusion process, which bonds shredded scrap into a solid piece of aluminium without the need of re-melting. It was seen that for a given foil thickness, the bulk density increased significantly by adding torsion, while heating to 450 °C on top of that only increased the compaction slightly.

3.2 Porosity and surface roughness

The porosities of several briquettes were analysed by CT and are summarized in Table 2. The relationship between porosity and bulk density was found to be approximately linear; higher bulk densities have lower internal porosity. For briquettes of similar densities, the lowest porosity was achieved for the thinnest foil, but the porosity differences between materials of different thickness significantly decreased for the highly compacted briquettes. For instance, in the column to the right (Table 2), which corresponds to samples compacted by MPT, the difference in porosity between samples is less than 2 %. Figure 5 shows how the MPT method successfully reduced the internal porosity.

Table 2 Average internal porosity (%) measured by CT for different material thickness and briquette bulk density ranges

	0.8-0.9 g/cm^3	1.1-1.2 g/cm^3	2.0-2.1 g/cm^3	2.4-2.5 g/cm^3
15 μm	57	29	10	4
30 μm	57	45	15	
100 μm			17	6
300 μm			18	4

Fig. 5 Briquettes of 100 μm foil. Left: MPT sample with bulk density 2.48 g/cm^3 compacted with 90 kN and 4 torsion turns. Right: Sample compacted with 115 kN (91.5 MPa) uniaxially to bulk density 2.09 g/cm^3.

3.3 Oxidation

After heat-treatment in air at 650 °C for 1 h, the weight of the briquettes and loose chips increased due to oxidation. Each briquette and batch of chips weighted 20 g initially, and Figure 6 shows the mean percentages of weight increase. The results show a correlation between the degree of oxidation and material thickness. Chips and briquettes of thinner material oxidize more, since they display a higher surface to mass ratio. For the 5 studied materials in order of increasing thickness, the area of foil/sheet in 20 g is approximately 494, 247, 74, 37 and 25 cm^2 (values calculated assuming all materials have the density of pure Al 2.7 g/cm^3). The oxidation differences between materials are reduced when compacting the chips uniaxially, likely due to more similar values of surface area exposed when compacted. For the MPT briquettes, the oxidation differences between materials are almost negligible. Compacting the chips reduced the % wt. gains by 50 % or more for all the materials. However, the degree to which they were compacted did not seem to have a significant effect, and all the briquettes compacted uniaxially (bulk density range 0.8-2.1 g/cm^3) resulted in similar weight increases, despite the CT analysis revealing large porosity variations within the briquettes and the optical microscopy analysis showing reductions of the surface
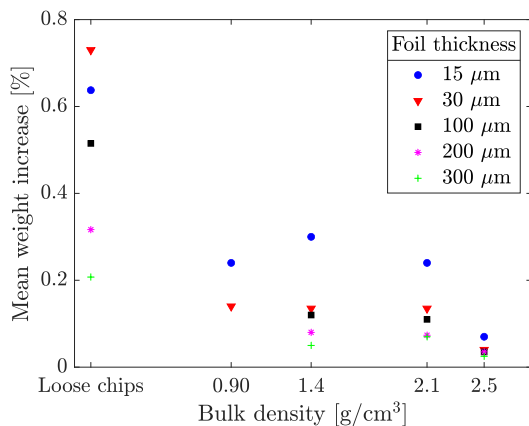
Fig. 6 Mean percentage of weight increase after heat-treating 20 g samples of the 5 different materials in 5 bulk densities. The bulk density of the loose chips is considered as 0.

roughness after high compactions. A further reduction in the % wt. gain did occur for the briquettes compacted by MPT to 2.5 g/cm^3. A hypothesis is that the MPT method achieves a certain level of solid-state bonding which inhibits the oxygen from penetrating through the briquette. On the contrary, the briquettes compacted uniaxially were more porous and the oxygen would still partially oxidize some of the surfaces between the chips.

3.4 Re-melting

Figure 7 shows the coalescence differences between heat-treated loose chips and briquettes for the 30-300 μm materials, and Figure 8 the coalescence and yield vs. bulk density for the 15 μm foil. The coalescence differences between loose chips and briquettes is 15.6, 2.7, 1.3 and 0.8 % for the, 30, 100, 200 and 300 μm materials respectively. The results showed that compacting the chips improved the coalescence, but this effect decreased for increasing material thickness. The variations in briquette bulk density for the material gauges 30, 100, 200 and 300 μm did not have a tangible effect in re-melting thus all the results were averaged. For the 15 μm foil, the coalescence did vary for different values of bulk density and therefore the results are plotted separately in Figure 8. The reduction when compacting 15 μm foil chips to the lowest density briquettes (0.9 g/cm^3) was 36.1 g/cm^3.

The reasons why coalescence improved with increasing bulk density for the thinnest foil might be related to several factors such as the breakage of the oxide thickness, decrease in the specific surface area or increase in thermal conductivity. This hypothesis is discussed in more detail in [22]. The understanding of these mechanisms at a more fundamental level will be the focus of future investigations. For the non-heat-treated samples, the material yield ranged between 98 and 100 % for all thicknesses and degrees of compaction. Thus, the pre-existing oxide layer (before re-melting) appears in this case to be more relevant than the oxidation developed during re-melting. It is important to remember that the experimental set-up consisted of adding clean scrap directly into a crucible at 800 °C filled with a bath of molten salt-flux. The salt-flux
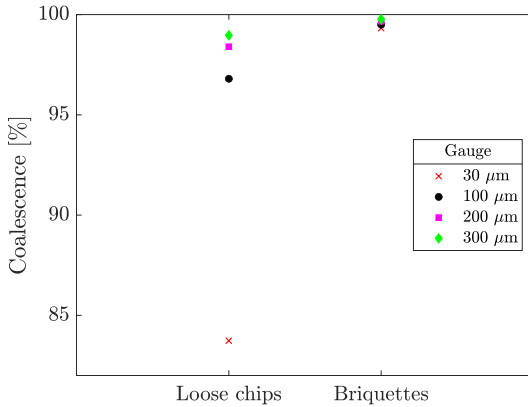
Fig. 7 Coalesced recovery after re-melting loose chips and the briquettes for the 30-300 μm material.
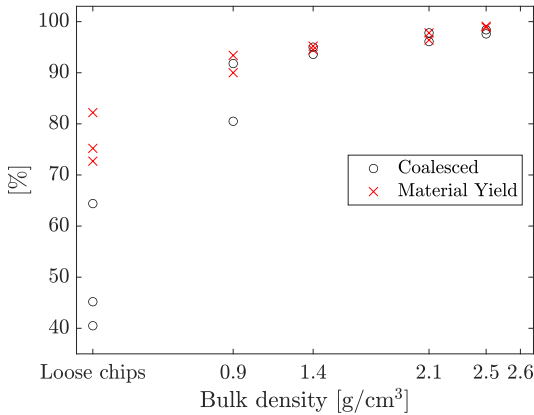
Fig. 8 Material yield and coalesced recovery for 15 μm foil compacted to different densities. Unit is % wt. of the input material, which weighted 20 g.

method effectively protected the scrap from oxidation, but this may not be the case for other re-melting processes or scrap properties, where thickness and compaction could influence the yield and coalescence differently.

4 Conclusions

The aim of the present work was to study the compaction of thin Al (15-300 μm) foil as a recycling pre-treatment. The relationship between foil thickness, briquette bulk density, oxidation during heat-treatment, and recycling yield and coalescence was investigated. The following conclusions were drawn from the results of the study:

Compaction

- Material thickness plays an essential role in the compaction step. Thicker materials required higher pressures to reach a given bulk density, and higher bulk densities for the briquettes to hold together.

- For equivalent bulk densities, the internal porosity was higher for thicker materials. This dependency on thickness decreased for higher bulk densities. Higher compaction reduced the briquette's porosity and surface roughness.
- The MPT method allowed reaching densities close to the bulk density of aluminium at relatively low uniaxial pressures, and the Hot MPT method gave slightly higher densities: 2.5 and 2.6 g/cm^3 respectively.

Oxidation

- Compacting foils into briquettes reduces the specific oxidation during heat-treatment significantly, and this is more explicit the thinner the material is.
- The degree of oxidation did not vary for different bulk densities within the range 0.86-2.12 g/cm^3, which corresponded with the uniaxial compaction method. The briquettes compacted with MPT to bulk densities 2.4-2.6 g/cm^3 were the least oxidized after the heat-treatment.

Re-melting

- Compacting the aluminium chips before the heat-treatment promoted their coalescence and material yield, and this effect increased for thinner foil gauges.
- For the thinnest foil (15 μm), the degree of compaction of the briquettes influenced coalescence and material yield. The lowest bulk density achieving yields higher than 95 % was 2 g/cm^3. For the thicker materials (30-300 μm) the difference between degree of compaction was negligible and all achieved yields higher than 98 %.
- Re-melting the non-heat-treated chips and briquettes resulted in material yields above 98 % for all the materials.

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