ARCHIVES
of
FOUNDRY ENGINEERING
10.24425/afe.2025.153791
Published quarterly as the organ of the Foundry Commission of the Polish Academy of Sciences

ISSN (2299-2944)
Volume 2025
Issue 2/2025
35 – 40
4/2

Behaviour of High-density Aluminium Briquette During Melting in Laboratory Conditions

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Received 02.07.2024; accepted in revised form 13.01.2025; available online 17.04.2025

Abstract

The aluminium industry is one of the most energy-intensive industrial fields and is associated with severe environmental impact mainly due to GHG emissions. Aluminium recycling is one of the best ways to eliminate this impact and to achieve better economic viability of aluminium production. Piece size and contamination of aluminium scrap are two of the most important factors that affect recyclability of aluminium and its alloys. Scrap with large piece size is relatively easy to recycle because it is associated with lower metal loss and fewer undesired inclusions introduced into the molten metal during remelting. Unfortunately, a large portion of scrap generated by industrial sector and by end-users is of small piece size. Despite its high importance, recycling of these types of scrap is often overlooked by contemporary literature. The aim of this work is to describe melting behaviour of high-density aluminium briquette prepared from thin aluminium foils and to provide metallographic observation of inclusions introduced into the molten metal during melting of the briquette. Melting is performed in laboratory conditions. The inclusions are analysed using optical microscopy in combination with SEM and EDX analysis. The results indicates that despite fast immersion of aluminium briquette, high portion of oxide films were introduced into the melt. Carbide like particles were also observed in microstructure, probably as a result of burning of organic contamination of the briquette. However, melting process in real industrial conditions differs from laboratory experiment which is a topic for further study.

Keywords: Aluminium, Recycling, High-density briquette, Oxide films

1. Introduction

Extracting aluminium from ores is an extremely energy demanding process and is associated with high economic expenses and large environmental impact. [1–2] It is estimated that the aluminium industry is responsible for up to 3 % of human-related GHG emissions [1]. On the other hand, aluminium recycling is considered a much more environmentally friendly approach and also more commercially viable option. [1–2] It is reported that by aluminium recycling up to 95 % energy savings can be achieved. [3–4]

Aluminium is often considered an infinitely recyclable material. This is supported by figures often quoted in the literature. For example, it is generally stated that about 75 % of all aluminium produced in the world history is still in use, or that up to 90 % of aluminium and its alloys from the transportation industry and the civil engineering industry are recycled. [2] However, above figures need to be taken into account with respect to some other realities.

Firstly, aluminium and its alloys are often recycled into less valuable materials which is referred to by the term "downcycling".

That means, they are recycled into materials with lower properties or lower chemical purity. [5–6] Secondly, there are several industries with much lower recycling rates such as packaging industry. [7–8]

Several factors influence the recyclability of aluminium and its alloys. The tree main factors are the chemical composition (uniformity of chemical composition) of the scrap, the scrap piece size and shape (especially surface-to-volume ratio) and the scrap contamination with organic residues. [3–5, 8–11] The first factor can be controlled by a sorting system and by narrowing portfolio of alloys used for the application. [3, 6, 11–12] However, the other two factors cannot be bypassed in a similar fashion and can cause significant problems during the recycling process.

The scrap piece size significantly affects the metal recovery. Smaller scrap piece size means higher metal loss during remelting due to oxidation. Scrap types as thin foils are very problematic and usually cannot be remelted directly. They must be compacted before remelting to avoid metal loss to slags/dross. [13–15] Briquetting is a very common form of compacting for thin aluminium foil scrap but also for other alloying additives (metallic powders, fluxes, etc.) used in aluminium metallurgy [16–17]. Recently, briquetting was also introduced as a method in solid-state aluminium recycling chain [18–19]. The bulk density depends mainly on pressure during briquetting and the thickness of the foil. Higher pressure means higher briquette density for the same foil thickness which is usually better for achieving higher metal recovery. [4]

Ideally, the density of the briquette is higher than that of molten aluminium which is approximately 2,33 g/cm^3 at 800 °C [20]. This condition means that the briquette can immediately immerse when added into the melt. In order to prevent oxidation and impurities, protective salts are used in the remelting of compacted scrap under real industrial conditions. [4, 21]

Current environmental pressures highly accelerate research in the field of processing low piece size and contaminated aluminium scrap. There are several papers that address this issue, e. g.: [14–15]. Unfortunately, despite the EU legislation plan to recycle 75 % of all aluminium from packaging applications by 2030 [7], recycling of thin foil aluminium scrap is often overlooked with exceptions, e. g.: [15, 22]. This work describes the behaviour of the high-density thin-foil aluminium briquette when remelted under laboratory conditions without a refining process. It aims to investigate the types of inclusions that are released into molten aluminium by optical microscopy, SEM and EDX analysis. The process without refining was chosen to map the natural processes in the melt and to highlight the role of proper metal treatment. However, the real behaviour of the compacted aluminium scrap will need to be investigated in real industrial process in a future study.

2. Material and methods

High-density aluminium briquette (Fig. 1) was prepared in cooperation with an external partner. It consisted of aluminium foils of a wide range of chemical composition and thickness from 0.135 mm to 0.007 mm. The exact process of its production is unknown. The density of the briquette ($\rho_{briquette}$) was calculated from its mass and volume according to Eq. (1):

$$\rho_{briquette} = \frac{m_{briquette}}{V_{briquette}}, \tag{1}$$

where $m_{briquette}$ is the mass of the briquette and $V_{briquette}$ is the volume of the briquette.

The mass of the briquette was measured directly using standard laboratory scales. Briquette volume was measured indirectly as a volume of water displaced by immersion of the briquette.

Fig. 1. The high-density briquette

The melt was prepared from 1.668 kg of high purity aluminium scrap (EC wire with a min. 99.7 % aluminium content) in a laboratory furnace. The crucible containing the scrap was heated at 770 °C for 2 hours until melting. Prior to the start of the experiment, slag was removed from the surface of the molten metal by stirring.

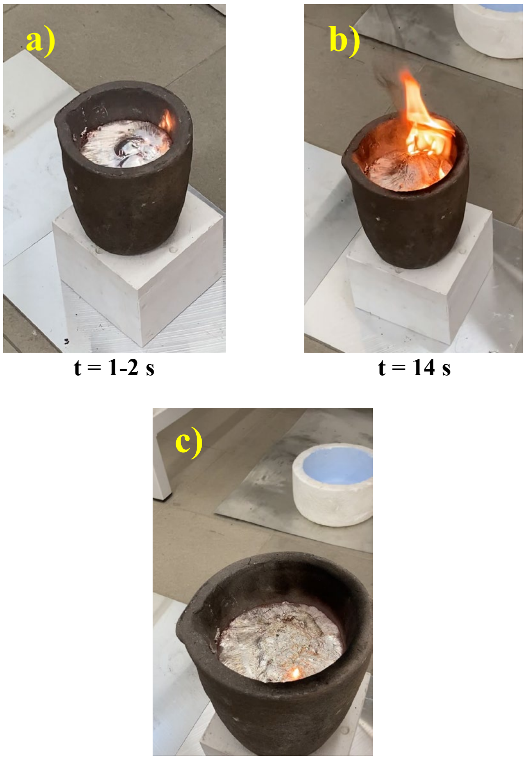
After the melt was prepared, the briquette was slowly laid at the surface of the molten metal. Its behaviour was observed by a camera. Then the crucible with the melt was returned to the laboratory furnace (without stirring) for 6 hours. After the holding time, the crucible was slowly cooled in the furnace.

After cooling, the metal casting was removed from the crucible and samples were prepared for metallographic observation. Optical microscopy was performed on an OLYMPUS GX 53 optical microscope. SEM and EDX analysis were performed on a Thermo Scientific electron microscope. Samples were analysed in unetched state. The chemical composition of the casting was measured on the iSPARK quantometer. It was measured in two different sample areas.

3. Results

3.1. Behaviour of the briquette after immersion

Fig. 2 shows the time sequence during the briquette immersion experiment. After placing the briquette at the molten metal surface, the briquette immediately sank below the surface. About 1–2 seconds after the immersion (Fig 2a), a burning reaction with the development of gas bubbles appeared at the surface of the molten aluminium. The intensity of the reaction can be seen in Fig 2b. The reaction lasted about 26–28 seconds then stopped spontaneously (Fig 2c).



t = 1-2 s **t = 14 s**

t = 28 s

Fig. 2. Behaviour of the briquette – time sequence

3.2. Observation of the sample and chemical composition of the casting

Fig. 3 a) shows a macroscopic observation of the casting after removal from the crucible. Shrinkage was observed in the middle of the casting from the bottom to the middle. On the top side of the casting (below the surface), briquette residue (oxide formation) was observed.

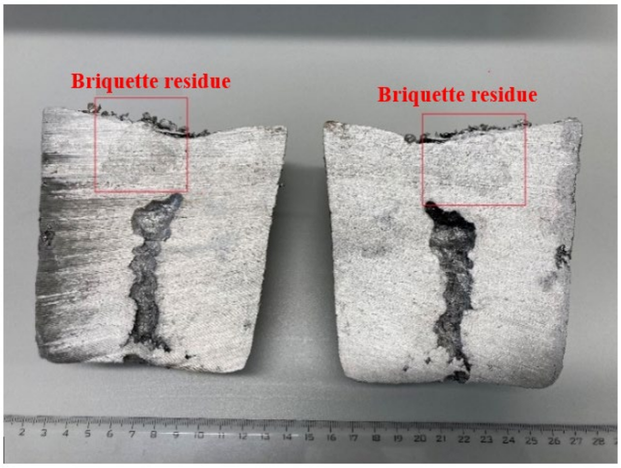


Fig. 3. Macroscopic observation of the casting

Fig. 4 shows the areas from which the samples were prepared for optical microscopy. Samples were prepared from Area 1 (near the surface), Area 2 (briquette residue), Area 4 (near shrinkage site) and Area 5 (compact metal).

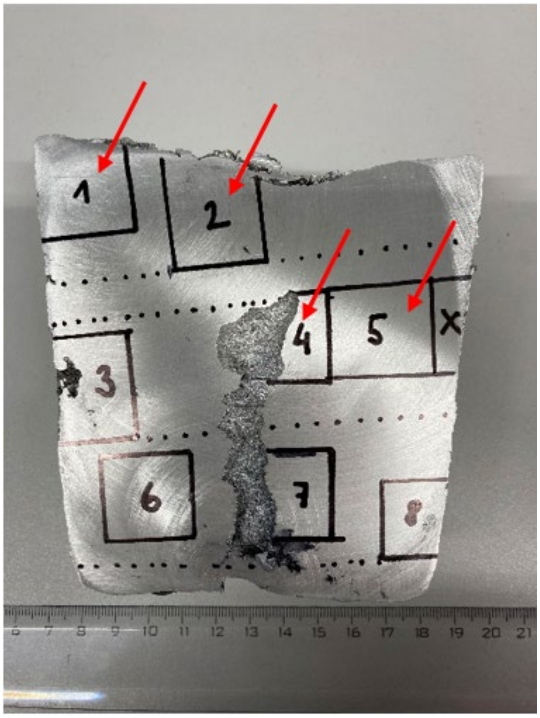
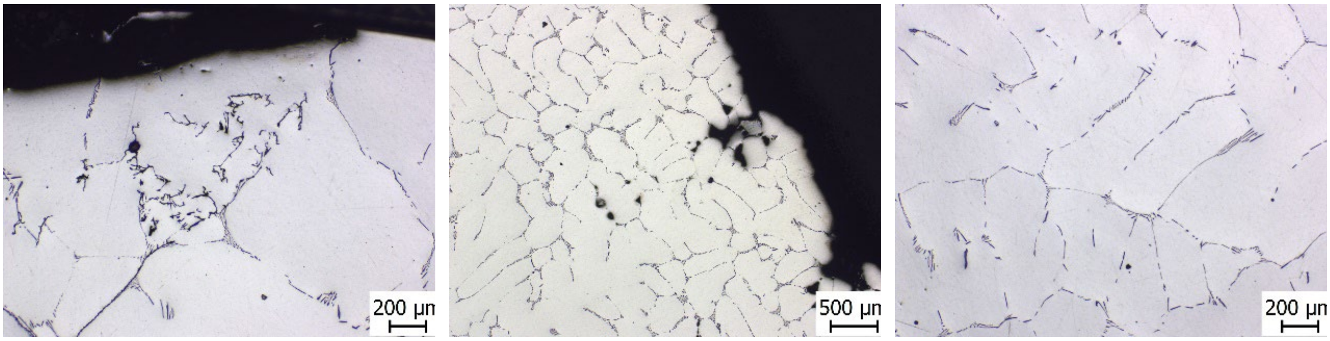


Fig. 4. Macroscopic observation of the casting

In Area 1, Area 4 and Area 5, no significant inclusion content was found which is demonstrated by the Fig. 5.

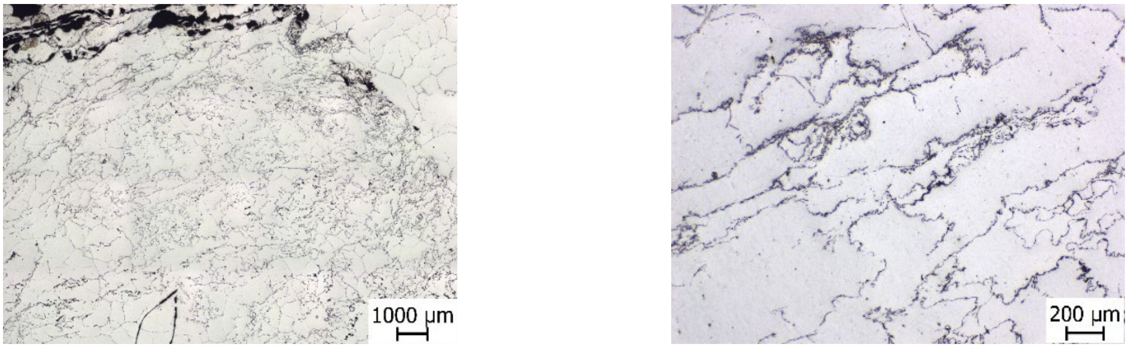
An overall view of the microstructure of Area 2 is represented by Fig. 6 a). Fig. 6 b) demonstrates that the structure contains a large number of oxide films. Carbide particles can also be seen in the microstructure, as shown in Fig. 6 c) and Fig. d).

The chemical composition of the resulting casting was evaluated in two different sample areas. One area was Area 5 (compact metal) and the other area was Area 2 (briquette residue). Results are shown in Table 1 in wt. %. As can be seen from the Table the chemical composition did not differ significantly between the two areas. The only minor exceptions were the iron content, which was approximately 0.1 wt. % higher in case of the compact metal area (Area 5), and the titanium content which was approximately 0.071 wt. % higher in the case of the briquette residue.

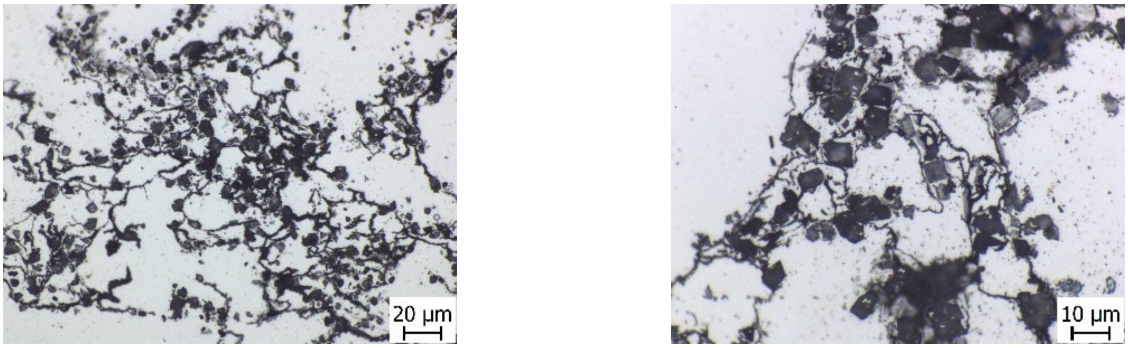


Area 1 Area 4 Area 5

Fig. 5. Microstructure of the different parts of the casting



a) general view b) oxide films



c) carbides d) carbides (detail)

Fig. 6. Microstructure of Area 2

Table 1.
Chemical composition of different samples areas

		Al [%]	Fe [%]	Si [%]	Cu [%]	Mg [%]	Mn [%]	Cr [%]	Zn [%]	Ti [%]	V [%]	Zr [%]	Others [%]
Compact metal	**Area 5**	99.676	0.211	0.075	0.002	0.001	0.003	0.002	0.012	0.002	0.002	0.001	0.014
Briquette residue	**Area 2**	99.636	0.103	0.091	0.003	0.005	0.003	0.003	0.054	0.073	0.005	0.004	0.021

The morphology and the structure of inclusions and casting defects can be also seen in Fig. 7 in BSE. Fig. 7 shows the void with elongated inclusions (dark contrast) and intermetallic phases (bright contrast) around. The morphology of the inclusions is typical of oxide films.

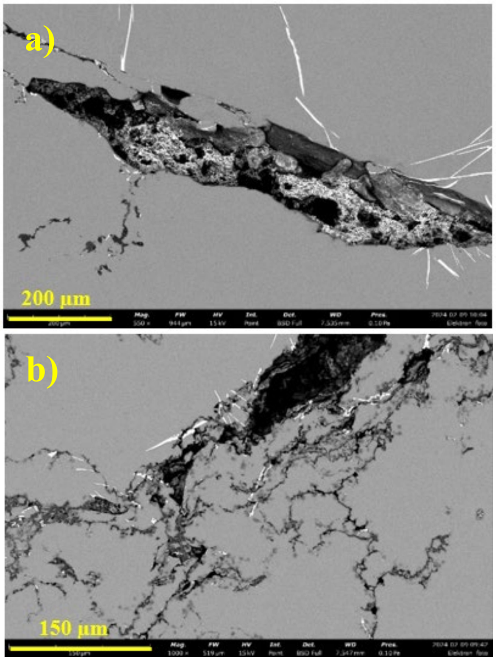
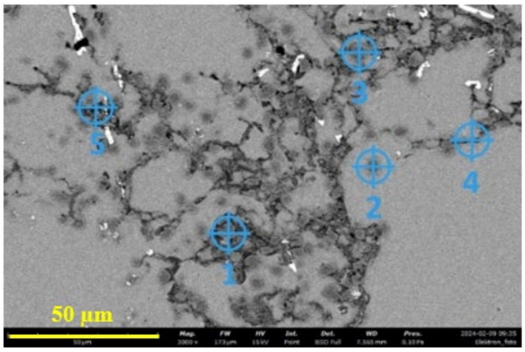


Fig. 7. Morphology and structure of the inclusions and the voids

Fig. 8 shows the results of the EDX analysis of the inclusions in which aluminium and oxygen were identified. The carbon content is problematic because it is a common contamination that is always present in EDX analysis of aluminium alloys.



Point	Al [wt. %]	O [wt. %]	C [wt. %]
1	39,5	38,0	22,5
2	40,9	38,2	20,9
3	42,9	38,9	18,2
4	46,1	30,6	23,3
5	40,3	42,4	17,3

Fig. 8. Results of EDX analysis

4. Discussion

Immediate immersion of the briquette into molten metal indicates that the density of the briquette was higher than that of molten metal [20], which is positive for the recycling process in terms of industrial conditions. On the other hand, the strong burning reaction (Fig. 2b) that appeared on the molten metal surface can be attributed to some organic residues (probably organic contamination) in the briquette. [21] This can be potential source of undesired non-metallic inclusions [15, 22].

Despite the very fast immersion of the briquette in the molten metal, Fig. 2 shows that briquette did not sink on the bottom of the crucible, nor did it disperse throughout the volume of molten metal. Instead, the briquette remained near the surface at the point of immersion and formed the briquette residue which can be clearly identified by eye.

By the optical microscopy (Fig. 6), SEM and EDX analysis (Fig. 7 and 8), it was proved that briquette residue was formed from oxide films. The density of the oxide films in the briquette residue was very high in comparison with the rest of the molten metal where there were almost no oxide films (except where it was in contact with the crucible). The oxide films in the briquette residue formed cluster with a diameter of about 30 mm. The cluster has bonded to the slag at the surface.

The aforementioned behaviour of the briquette could be problematic during the industrial process because on an industrial scale, the briquette can be withdrawn with the slag [15]. This means high metal loss. It is therefore necessary to ensure proper melt stirring. [14] In addition, it must be found how many briquettes can be added into melt in real industrial conditions. This is an important point for future study.

Except oxide films, carbide particles were also captured during metallographic observation of the briquette residue (Fig. 6). The particles probably appeared as a result of a burning reaction at the surface [15]. However, the composition of these particles cannot be verified by EDX analysis because carbon is often found on the aluminium sample as a standard contamination. All non-metallic inclusions are detrimental to the material properties which means that a sufficient refining process should be applied to avoid quality problems which is in accordance with literature. [5]

5. Conclusions

The high-density briquette made of aluminium foils was immersed into molten metal under laboratory conditions. During the experiment, the behaviour of the briquette was observed. After the end of the experiment, a metallographic observation was performed to analyse inclusions inserted in the melt. The following statements can be concluded:

- The briquette sank almost immediately after the immersion which means it has higher density than the molten metal. However, the briquette did not sink to the bottom of the crucible, nor disperse in the volume of the molten metal. Instead, it formed briquette residue near the surface of the molten metal. The briquette interacted with the surface slag. This could be problematic in terms of industrial conditions.

- Metallographic investigation proved that the briquette residue was formed from oxide films. Carbide particles were also detected in the residue. For this reason, achieving the proper refining process is very important in real industrial conditions to avoid quality issues.
- The behaviour of briquettes under real industrial conditions is an important step in future study.

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