SURFACE FUNCTIONALIZED CERAMIC FOAM FILTER FOR THE FILTRATION OF ALUMINUM

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ABSTRACT

The recycling of aluminum provides high efficiency in terms of ecologic and economic aspects. To ensure high quality products for both secondary and primary aluminum, appropriate and effective procedures have to be applied for the removal of non-metallic inclusions. A simple, relatively cheap, and efficient process to reduce non-metallic inclusions is the usage of ceramic foam filters during the casting process.

The present study focuses on the effect of the filter surface chemistry on the filtration behavior. Five different oxide filter surface chemistries (alumina (Al_2O_3), spinel ($MgAl_2O_4$), mullite ($3Al_2O_3 \cdot 2SiO_2$), silica (SiO_2) and titanium dioxide (TiO_2)) were tested with the aluminum alloy AlSi7Mg with regard to their filtration behavior.

The filtration experiments were conducted on a laboratory scale with a filtration pilot plant at Constellium C-TEC (Voreppe, France), which allows appropriate filtration durations (40 to 76 min) using a 750 kg aluminum furnace. During these trials the amount and size of the non-metallic inclusions were quantified with LiMCA (Liquid metal cleanliness analyzer). The test set-up allows the simultaneous use of two LiMCAs (before and after the filter) from which the filtration efficiency can be calculated.

All filter natures were quite efficient in terms of inclusions removal (85 to 95%). The test results showed the coatings to rank in the following sequence (from highest to lowest): Al_2O_3, $MgAl_2O_4$, $3Al_2O_3 \cdot 2SiO_2$ and TiO_2. The analysis of the removal efficiency as function of the inclusion size shows that the Al_2O_3 and the $MgAl_2O_4$ filters are comparable (efficiency closed to 100% for inclusions larger than 90 µm) while the $3Al_2O_3 \cdot 2SiO_2$ filter possesses the lowest filtration efficiency for the larger inclusions (> 110 µm). In these trials, the $3Al_2O_3 \cdot 2SiO_2$ filter showed the highest filtration efficiencies for smaller inclusions (< 60 µm). The TiO_2 filter showed the lowest filtration efficiencies.

The spent filters were metallographically analyzed at the SEM in order to observe the interactions between the inclusions and the filter surface. Only limited interaction between inclusions and the four different coating natures were observed, regardless of the chemistry of the coating.

Keywords
Aluminum, Filtration, ceramic foam filter

INTRODUCTION

Melt cleanliness is essential for high quality aluminum production [1]. The main cleanliness criteria are dissolved hydrogen (potentially resulting in porosity) and non-metallic inclusions.

For cast parts, non-metallic inclusions may impair the castability, the mechanical properties, and the machinability of the components. In thin rolled products (e.g. aluminum cans and foil) inclusions can create pinholes, while in thick rolled products (used e.g. in transportation), inclusions may restrict fatigue life. These effects of inclusions illustrate the fact that the continuous improvement of melt cleanliness is essential to meet the ever-increasing quality requirements of today's final products. Inclusions also have different origins, e.g. from scrap melting, oxidation of the melt surface, and erosion of the refractory material. In aluminum castings, oxides (Al_2O_3, MgO, $MgAl_2O_4$, $3Al_2O_3 \cdot 2SiO_2$, SiO_2), carbides, and nitrides are the most common types of inclusion [2]. A simple, relatively cheap, and efficient way to meet the increasing requirements of aluminum castings relies on ceramic filters during the casting process. The ceramic foam filter (CFF) is the most common filter type for aluminum. A significant amount of work has been done to assess the efficiency of filters [3,4].

The filter is selected based on processing, thermo-mechanical, and economic criteria. It is known that inclusion capture in a CFF mainly occurs by hydrodynamic effects, which leads to instability risks during operation [4]. Creating a chemical interaction between the inclusion and a filter surface would be of interest with this respect. Furthermore an active filter surface may lead to a selective capture of inclusions.

There is only limited information, however, on the influence of the filter surface material on the filtration efficiency of liquid aluminum.

According to Görner et al., for example an active filter surface made of AlF_3 removes Na and Mg by forming NaF and MgF_2 [5]. According to Zhou et al. for example an active coating made of an enamel can capture nonmetallic inclusions and dissolve Al_2O_3 during the casting of aluminum. They find an increase in the elongation at fracture of aluminum samples filtered with enamel-coated filters compared with aluminum samples filtered with uncoated filters [6]. A second example for sticking active coatings are filter surfaces made of NaBr are tested by Luyten et al. [7] and Oosumi et al. [8] for the filtration of small particles and intermetallic particles. The third context of active filter coating is used by Emmel et al. [9] in the area of the steel filtration for filter coatings consisting of refractory materials with a coating compositions comparable to inclusions and different wetting behavior. Emmel et al. [9] improved the deposition of Al_2O_3 inclusions from a steel melt by applying an alumina coating on ceramic foam filters made of Al_2O_3-C.

Syvertsen et al. [10] compare the filtration of aluminum of Al_2O_3 and SiC foam filters, and find for the SiC filters. A possible explanation for the better filtration efficiency of SiC could be the better wetting of the SiC. The measured wetting angle at 1373 K (1100°C) for the SiC filter material is 39° and for the Al_2O_3 filters material 84° [10].

Inspired by positive results obtained during the active filtration of steel (which resulted in inclusion sintering on the filter surface) [11], Voigt et al. investigate the filtration efficiency (in filtering aluminum) of filters with different surface chemistries: Al_2O_3, $MgAl_2O_4$, 3 $Al_2O_3 \cdot 2SiO_2$ and TiO_2. Based on scanning electron microscopic observations, they find that Al_2O_3 and $MgAl_2O_4$ filter surfaces exhibit better filtration behavior than the other two materials [11].

The filtration efficiency of filters is most often characterized by LiMCA (Liquid Metal Cleanliness Analyzer) and/or PoDFA (Porous Disk Filtration Apparatus). LiMCA is considered the reference technology for quantifying inclusions in molten aluminum [12,13]. Based on the principle of the Coulter Counter, LiMCA facilitates the quantification of inclusion number and size in molten aluminum. This technique uses an orifice which is plunged in the liquid aluminum and is based on an increase of the effective electrical resistance when an inclusion passes through the orifice [14, 15].

In order to contribute to the understanding of the impact of surface chemistry on aluminum filtration, four different filter surfaces were produced: Al_2O_3 (alumina), $MgAl_2O_4$ (spinel), 3 $Al_2O_3 \cdot 2SiO_2$ (mullite), and TiO_2 (rutile). Experimental casts of

AlSi7Mg0.3 were performed under controlled test conditions, including a particle seeding process. The casts were monitored by LiMCA, and the spent filters were characterized by metallographic methods.

MATERIALS AND METHODS

The pilot filtration line used for the trials is shown in Figure 1 and consists of a 750 kg resistance tilting furnace, a launder system, a filter box, and a ladle to collect the filtered material. A metallic grid was put at the exit of the tilting furnace in order to retain the oxide films and prevent premature filter clogging.

The ingots (AlSi7Mg0.3, EN AC-42100) were melted in the furnace at 1023 K (750 °C). A rotary degassing treatment was applied in the furnace (rotor with 6 blades, Ar 500 Nl/h, 10% Cl_2, 20 min), and the metal was left to settle.

In order to guarantee the reproducibility of the trials, the casting line was equipped with thermocouples and a metal level indicator. The filter box was preheated and once a set point was reached the filter used for the trial (see 2.2) was set in and preheated up to 923 K (650°C) with a hot air burner.

In order to guarantee similar inclusion contents in each trial, it was decided to seed the metal before casting by using inclusion composite ingots. The following particle types were chosen: alumina (Al_2O_3), spinel ($MgAl_2O_4$), mullite ($3Al_2O_3{\cdot}2SiO_2$), and silicon carbide (SiC). The preparation of the inclusion composite ingots was carried out as follows:

1. Alumina, spinel, mullite and silicon carbide powders (at a ratio of 1 to 1 to 1 to 1) were introduced in aluminium melt with the help of a vortex created in a 10 kg crucible with a graphite rotor. This was carried out using a vibrating table.
2. Critical parameters were controlled during the fabrication of the inclusion composite ingot, i.e. the rotation velocity, metal temperature, and feeding rate.
3. The base metal was an industrial-purity alloy with 5% Mg (primary ingots + Mg addition), which improved the wetting behaviour.
4. The inclusions were kept in suspension by stirring during about 5 min before 4 times 2 kg were removed by spoon sampling for each ingot.
5. The homogeneity of the ingots was checked by optical metallography on cut ingots.

For each test, the metal was seeded with a piece of 2 kg inclusion composite ingot from the same inclusion composite ingot production run.

There was typically less than 10 min between the end of stirring and the beginning of the cast.

After the casting, the metal solidified in the filter: the tile was then removed from its seating and cut in order to perform metallographic observations of the inclusion distribution.

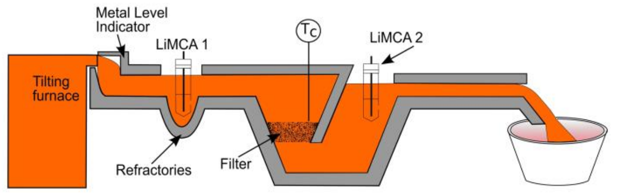

Fig. 1: Illustration of the filtration pilot line

In this study, 30 ppi (pores per inch) alumina filters were used as the starting material. Each filter pyramid had a truncated pyramid shape with a square section of 90 mm x 90 mm on the small side (melt outlet), 115 mm x 115 mm on the large section (melt entry) and a thickness of 50 mm. The initial alumina skeletons were coated using a modified replica technique, which uses a centrifuge for the removal of excess slurry coating and a two-step sintering process according to Voigt et al. [16]. This process generates filters with comparable pore size and varying surface properties. Also the alumina skeleton was coated with alumina slurry to reach a pore size comparable to the other filters which were coated with spinel, mullite and rutile. Every coating step increases the strut thickness and decreases the pore size of the filter.

In this work, the following surface chemistries were evaluated: Al_2O_3 (alumina), $MgAl_2O_4$ (spinel), $3Al_2O_3{\cdot}2SiO_2$ (mullite), and TiO_2 (titanium dioxide). A detailed description (compositions and characterization of the prepared filters) is given by Voigt et al. [16]. One filtration test with 750 kg aluminum alloy was carried out for every filter surface chemistry. The filters were installed in the filtration box and an expandable gasket was used to prevent metal leaks on the lateral sides.

In order to assess the inclusion content of the melt, two LiMCA II units (ABB Inc., Quebec, Canada) were installed on the casting line, one before and one after the filter. The LiMCA measurements are expressed in k/kg (thousands of inclusions per kg of molten aluminum). N20 and N40 are often used, representing the total amount of inclusions larger than 20 and 40 µm, respectively, expressed in k/kg. Similarly, N20-30 represents the number of inclusions in the size range 20-30 µm. These measurements also enabled the determination of the filtration efficiency E of the filters.

$$E = \frac{N20_0 - N20_i}{N20_0} \cdot 100 \qquad (1)$$

where $N20_0$ and $N20_i$ are the average number of inclusions larger than 20 µm before and after the filter, respectively [4]. In this calculation, the delay between the two LiMCA's is taken into account. About 0.5 kg of metal is analyzed by each LiMCA during one experiment.

A Philips XL 30 scanning electron microscope (SEM) was used to characterize the functionalized filters before and after the filtration trials. In the case of the spent filters, a more detailed investigation of the filtration behavior was carried out using 3 samples. An area of 6 mm x 4.6 mm was scanned in the back-scattered electron mode with a magnification of at least 200 x. The inclusions found were imaged with a suitable magnification and analyzed with EDX (EDAX Phoenix, Mahwah, USA).

RESULTS

The morphology of the experimental filters was assessed before being used for casting trials. The porosity, the pore, and struts sizes are comparable. The filters exhibited very similar structural properties. Considering that they were manufactured from the same substrate, this proves the reproducibility and comparability of the coating thickness independently of the viscosities and chemical composition of the slurries.

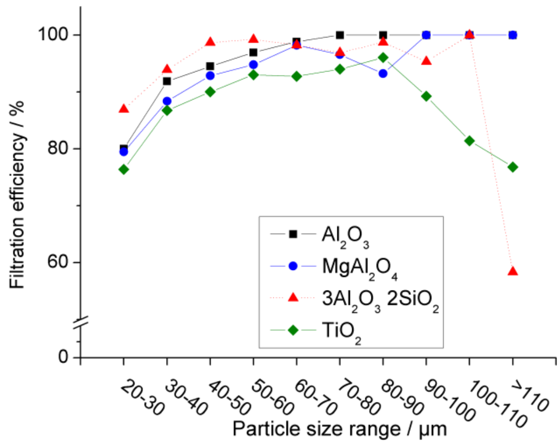

Fig. 2: Filtration efficiency of the four different coatings as a function of the particle size range (The typical error on the efficiency is around 5%)

The inclusion removal efficiency was calculated from the LiMCA data.

The overall efficiency was calculated from the counts of the whole cast. The efficiency was higher than 80% for all casts based on the N20 index, and higher than 90% for all casts for larger inclusions (N40 index).

A more detailed analysis can be done by focusing on size classes. Following this approach, the mean inclusion count of each size class (width 10 µm) was calculated before and after the filter, providing the mean efficiency for this specific class during casting. The results are presented in Figure 2. Differences were observed for the large inclusion sizes: while the spinel and alumina coatings have increasing efficiencies for larger inclusions, mullite and rutile coatings exhibited lower efficiencies for larger inclusions. This behavior was already observed in a previous work by Voigt et al. [11].

Before analyzing the inclusion capture mechanism on the filter surface, the stability of the coating had to be checked in order to identify any filter degradation during casting. This was done by combined SEM, EDX and EBSD.

The SEM analysis show that no damage had occurred to the coatings.

DISCUSSION

The methodology used in this study allowed a rigorous comparison of the behavior of four filter surfaces with different chemistries (Al_2O_3, $MgAl_2O_4$, $3Al_2O_3{\cdot}2SiO_2$ and TiO_2). The characterization of the initial filters showed that the porous structure was similar in each filter (in terms of mean porosity, strut thickness). During each test about 730 kg were cast in 40 to 75 min, which corresponds to filtration velocities ranging from 3 to 7 mm/s (calculated based on the ratio of the mean metal flow to the empty filter cross section surface and the metal density). These values are in the range of velocities recommended in the literature for ceramic foam filters (CFFs) for aluminum filtration [4]. Observation of the spent filters showed that the filters had a suitable stability for aluminum filtration: the filters were not damaged after removal (for instance, they were not cracked) and microscopic observations showed there was no deterioration of the coatings. In addition, there was no impact on the chemical composition of the melt when the functionalized filters were used.

In order to obtain stable inclusion levels at the entrance of the filter, a seeding procedure was applied. These added particles represented the majority of the inclusion population. Although an Ar/Cl treatment was applied to the melt before seeding, there were additional inclusions resulting from the addition of the seeding ingots, including oxide from the turbulence and other potential inclusions from the dissolution practice (refractory particles). This explains why inclusions larger than 40 µm were present in the melt. These larger inclusions may also have been clusters of small introduced particles.

All of these inclusions were taken into account in the analysis of the data.

The efficiency of the filtration was characterized in real time by LiMCA measurements. The assessed total filtration efficiencies (calculated on the basis of the N20 data) were above 85% for all four filters. These values are considered to be very high compared to industrial data, which would range typically from 40 to 60% [4]. However, it should be noted that there was no refiner used in the reported lab-scale trials. It has been shown that the use of refiner (TiB_2, TiC) prevents the formation of bridges inside the filter pores, resulting in a reduction in inclusion capture [17].

As already reported in the LiMCA results presentation, two filter surfaces appeared to be less efficient in inclusion capture: mullite and rutile. This was true in particular for large inclusion sizes (see Figure 2). The impact of the chemical composition of a filter surface chemistry on filtration efficiency has received little attention in the literature to date.

Based on the counting of the individual inclusions present in the filter, the selectivity of the inclusion capture was assessed. Figure 3 indicates the proportion of inclusions of a given nature that were counted on each surface. It can be seen that the inclusion capture of the seeded inclusions was not related to the nature of the chemical coating used. This was likely due to the low relative temperature of the oxides in aluminum filtration (e.g. for Al_2O_3: 0.34 corresponding to 973 K (700°C – testing temperature) / 2345 K (2072°C – melting point of Al_2O_3)). Aluminum filtration is, in this regard, less favorable than steel filtration, where the relative temperature is closer to 0.80. For aluminum, this further suggests that the hydrodynamic factors were of higher importance than the chemical ones, or at least for non-reactive coatings.

Additional research is required in this area to confirm the initial findings of this work.

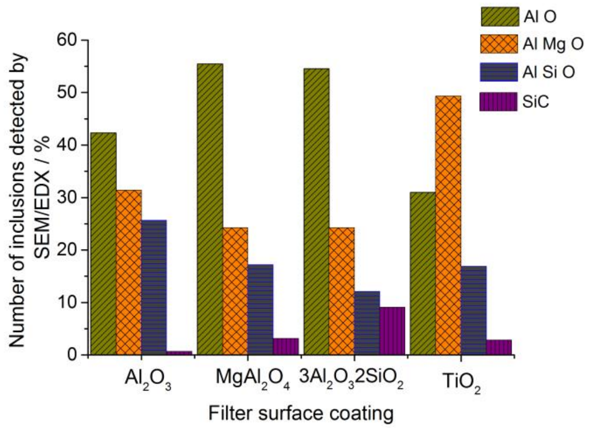

Fig. 3: Proportion of captured inclusions of a given nature as a function of the analyzed filter
coating

CONCLUSION

1. A rigorous lab-scale methodology was used to evaluate the behavior and the efficiency of ceramic foam filters for use in aluminum alloy production. Lab scale casts were performed which lasted for about 1.5 h, and LiMCA measurements were performed.
2. For the first time, filters with four different surfaces were evaluated with this methodology (alumina, mullite, spinel, and rutile). It was shown that the filters did not deteriorate during the tests. The surface deposits were not damaged. However, evolution of the chemical composition of the rutile coating surface occurred during the tests.
3. The filtration efficiency was determined by LiMCA. All the filter chemistries tested showed positive inclusion removal efficiencies by LiMCA. As a consequence, none of the filter compositions tested was considered inappropriate for CFF use based on the tests.
4. The interaction between the filter surfaces and the inclusions was observed in detail. During these observations, it was shown that inclusions were interacting with the surface of the filters.
5. More work is needed to understand the interaction between the particles and, in particular, the impact of the morphology and chemical interaction between the filter and the inclusion.

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