Removal of intermetallic precipitates from Al-melts by immersed centrifugation technology

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Abstract

For the refining of Aluminium alloys the precipitation of intermetallic particles is one possible route. However this route is always combined with the loss of aluminium melt in sedimentation residues or filter cakes. This paper presents an innovative procedure to remove intermetallic (and also ceramic) particles from aluminium melts with a low loss of aluminium melt. The removal technology is called filter centrifugation, a process that is widely known at room temperature for example in washing machines. The centrifuge used in this work consists of two discs forming a ring-space held together by an axis, that is also responsible for the power transmission. The upper centrifuge disc contains two holes, while the lower has a solid wall. When this centrifuge head is submerged in a melt and rotated, melt gets sucked in the space through the holes in the upper disc. Because of the radial acceleration of the melt it is pressed out between the outer contact area of the discs while the solids remain inside. The principle is similar to cake filtration, but there are some advantages: the filter medium can be repeatedly used, the melt passes through the discs more than once and because of the centrifugal acceleration the liquid removal is very good. After the head is filled, it is taken out of the melt and the remaining melt is spun out at high turning speeds.

Experiments were conducted in a water model and in a lab scale furnace. At first the feasibility was confirmed and challenges of the "high" temperature application were solved. Several parameters concerning particle size, centrifuge geometry and turning speed were investigated to achieve a fast removal of particles at a low loss of liquid in the filter cake.

The results showed that in the water model for particles with a grain size > 50 µm a residual liquid of < 5 % could be achieved, while the best result in the trials with a Al-melt was 25 %. The difference in the residual liquid lies in the different physical properties of the water silica suspension and freezing of the Al-melt during spinning.

1. Introduction

Aluminium has outstanding properties in the most important material characterising fields as high strength to weight ratio and high corrosion stability as well as good electric and heat conductivity. The high energy consumption during the primary production is faced by the low energy consumption during the recycling of this metal because in general only the melting energy is necessary. Problems can occur, in case, that during the recycling process impurities are brought in that form intermetallics with aluminium in the melt like iron and manganese. Such intermetallics can lead in wrought alloys to holes in sheets and in cast alloys to a reduction of the mechanical properties. Although aluminium alloys are often filtered before casting, the amount of intermetallics can lead to an inefficient precipitation and even blocking.

In this investigation an alternative technique for the removal of intermetallic particles is presented. A filter centrifuge is applied, that was developed for the copper dross removal from lead melts [1]. This kind of centrifuge was already used for refining of Aluminium, especially for the removal of Titanium and Iron from Aluminium-Silicon alloys [2]. Also oxide stripping of molten salts from the Aluminium industry was evaluated [3] and the separation of oxides and carbides from MMC scrap melts was investigated [4]. Sedimentation centrifuges were investigated for the separation of intermetallics from aluminium melts [5] and for the development of new Aluminium alloys [6]. In conventional filters a lot of residual liquid is present while in a centrifuge - due to the increased acceleration in comparison to gravitation - a higher separation efficiency can be achieved. Furthermore the filtrate passes through the centrifuge several times and increases he separation efficiency.

As a first step of the adaptation process of the centrifuge technique different shaped rotor heads were evaluated for the efficiency in solid liquid separation during spinning out of melt in a water model. Although water and Aluminium are not very similar in density, viscosity and surface tension and a prediction of results from the water model to the Aluminium is not trivial, only tendencies resulted showing the principal effect of parameters. The rotor design is the most important part of this process and is explained in the following in more detail.

2. Experimental setup

For the Experiments two different setups were used: A water model and a setup for trials in Aluminium alloy melts.

2.1. Rotor design:

The centrifuge consists of two discs that enclose an air space. In the upper disc two holes are placed close to the centre axis for a liquid inlet. The liquid is pressed out through the gap between the two centrifuging discs while the solids are held back. In practice at first an empty rotor head is lowered into the melt and rotated in general for a few minutes. The rotor head is taken out of the melt while rotating assuring that the solid liquid mixture does not flow out again. At a height above the melt but below the border of the crucible the elevation is stopped. The centrifuge is accelerated and the residual liquid is spun out. In industrial centrifuges the head is then emptied automatically and the cycle starts again. In laboratory scale the rotor head has to be unscrewed after each run.

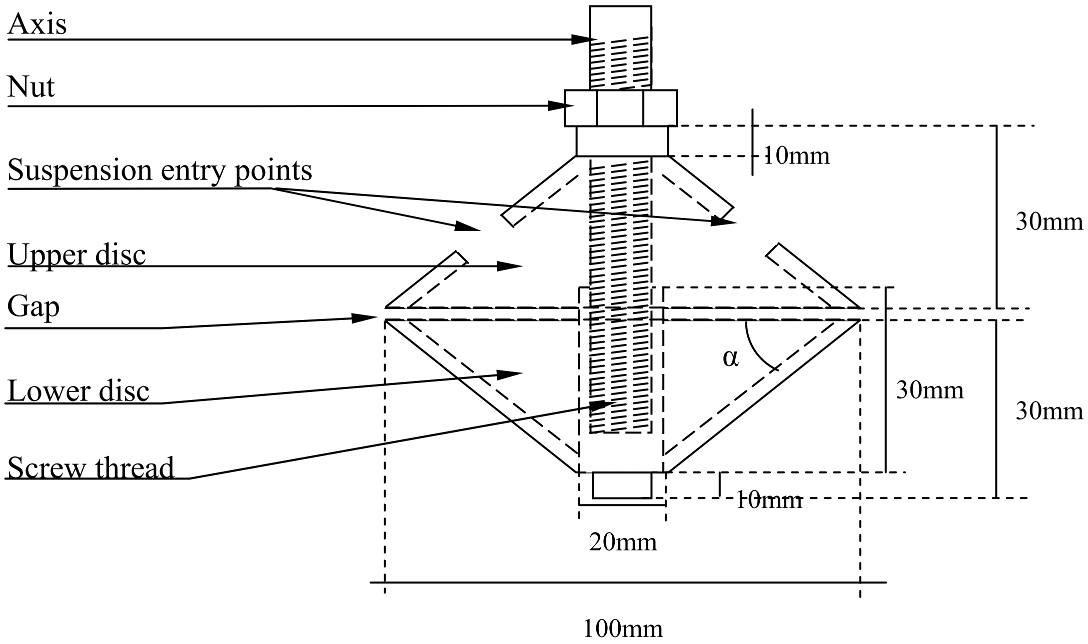

Figure 1: Principle of the lab scale centrifuge head

For the experiments three different disc angles α of the centrifuge head were investigated 30°, 45° and 60°. The rotor design is one of the major optimising tasks of this project. A higher rotor angle leads to a higher enclosed volume and therefore can take out more particles during a run, but the filter cake width increases and the total amount of suspension as well so that a efficient separation of liquid may be hindered.

2.2. Water model

The size of the water model was chosen similar to the size of the experimental equipment for Al-melts. The design is adaptable for different centrifuge diameters. As well different plastic cylinders can be placed in the holding container to simulate different crucible diameters. The water model consists of an Aluminium frame holding a Plexiglas water container. Two electric motors are installed; one motor lifts the centrifuge head, the second acts as the drive. Both motors are so called step motors and are controlled by a single control unit, allowing that certain patterns of lifting and centrifuging can be achieved. The step motors are used, because they have a very precise pattern of movement, so the trials can be repeated with high accuracy. The centrifuge discs are also made of Plexiglas and are transparent.

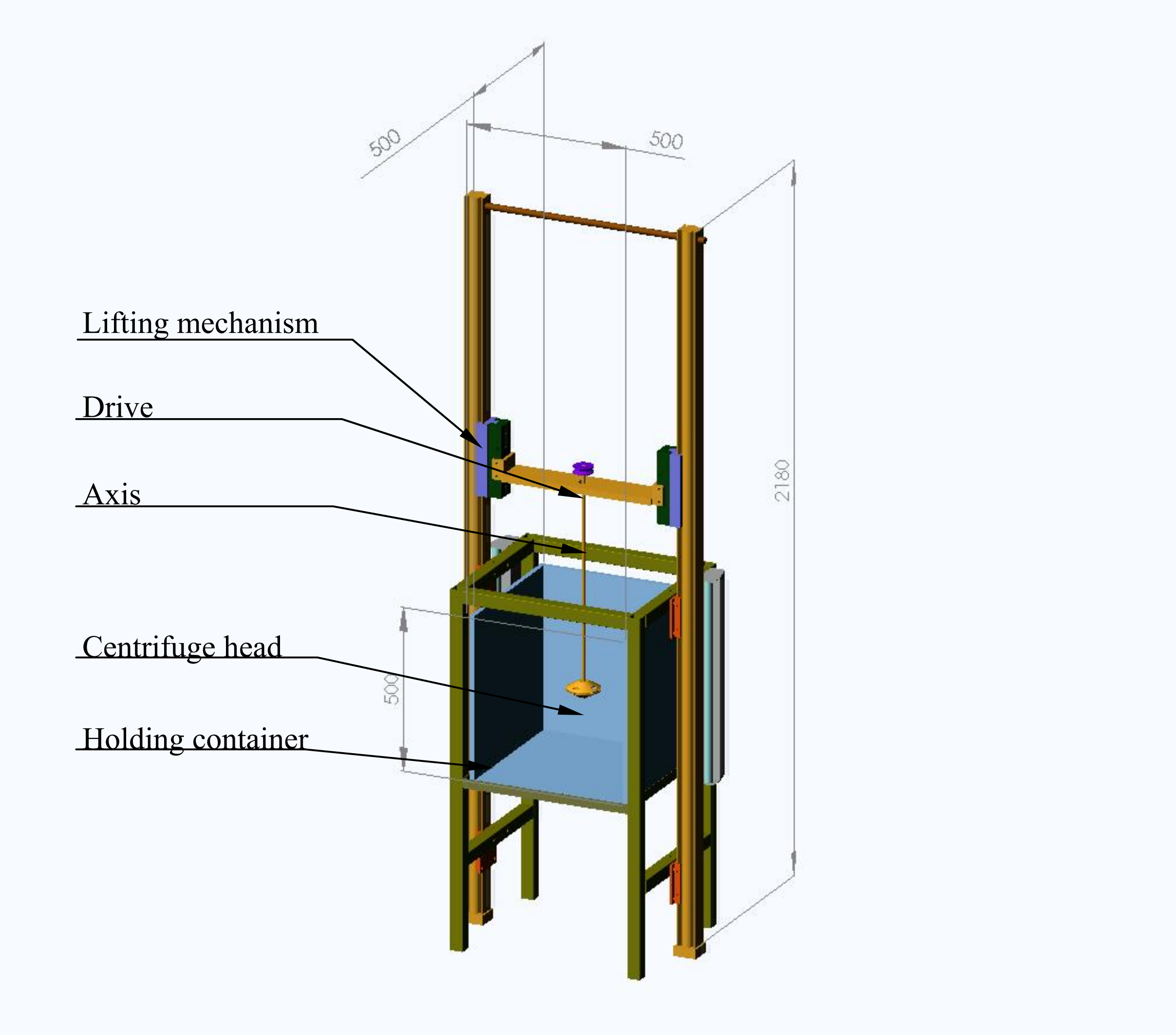

Figure 2: Sketch of the water model

As solid material in the water suspension glass beads with a spherical shape were used. The density of those particles was ~2.6 g/cm^3.

2.3. Lab-scale melt centrifuge:

The melt centrifuge operates in an indirect electric heated furnace. An Eurotherm controller that uses two thermocouples for the furnace control algorithm is used. One thermocouple is placed in the melt, a second close to the heating elements. With this control temperature homogeneity during stirring of 0.5 K could be reached. A sophisticated temperature control is very important, because the centrifuge works close to the melting point of the eutectic composition where freezing is imminent. The centrifuge is fixed to a standard electric motor, which has to be adjusted by hand. The speed is controlled by a handheld rpm-measurement device.

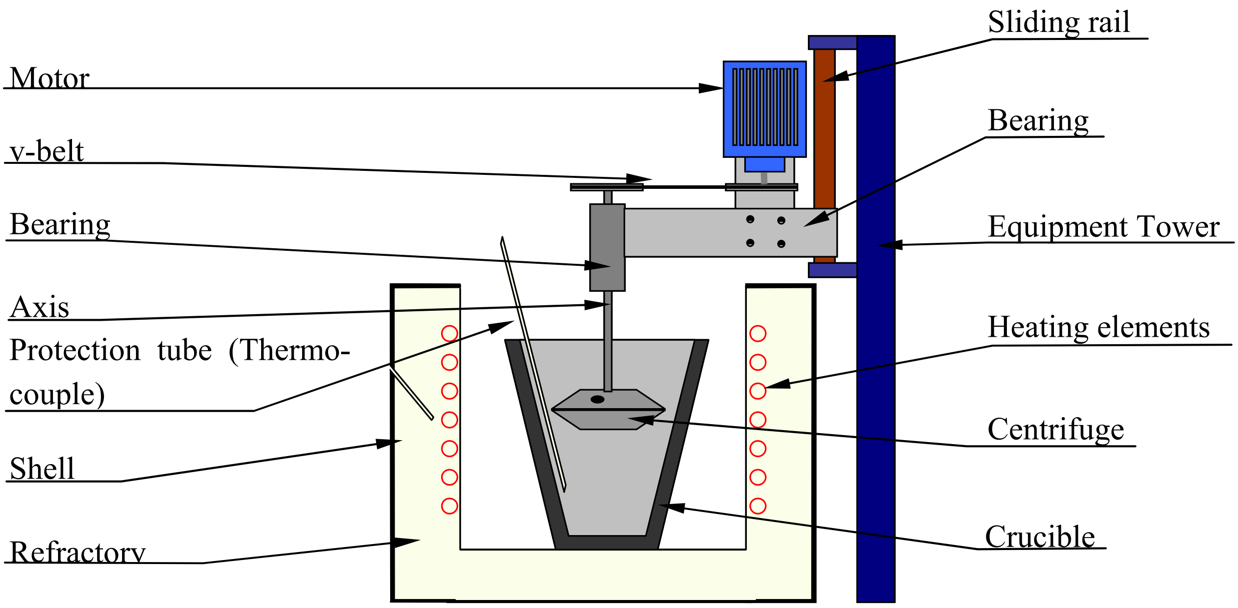

Figure 3: Sketch of the setup of centrifugation with melts.

As experimental medium an Aluminium alloy with 2.5 wt. % Fe and 1.5 wt. % Mn was used. For the trials the aluminium was molten and heated to 700°C. At his temperature the alloy elements were added and the melt homogenised by stirring for 30 min. The total amount of one batch of experiments was ~ 6 kg Al melt. The melt then was cooled to 680°C to precipitate intermetallics. The composition was selected, that the melt contains ~ 5 % of solid particles. The centrifuge head was made of high temperature resistant steel and was coated before every immersion into the Al-melt with a Boron-nitride coating (from am). The axis was made of high temperature steel as well.

3. Experiments in the water model

3.1. Trial programme

The aim of these experiments was the identification of process parameters leading to a reduction of the residual liquid that adheres between the solid particles. To investigate which parameters are the best for application of this technique, six different rotation speeds present as the acceleration at the outermost point of the centrifuge, three different centrifuge designs and five different particle sizes were investigated. The time of spinning was set to 5 min for all trials.

Table 1: Parameter variation of the water model trials

Acceleration[“g”]	Particle size [µm]	Centrifuge angle
13.5	< 50	30°
57.2	50 – 105	45°
125.0	105 - 210	60°
158.1	150 - 210	
227.3	250 - 420	
349.6		

All parameters were combined full factorial, which means each combination was tried out. For each trial the centrifuge head was filled with 20 g solids and 20 ml of desalinated water. The centrifuge was slowly rotated so that the suspension distributes evenly in the centrifuge. The centrifuge head was weighed before filling and after spinning and the residual water was calculated. In previous trials it was found that the solid loss due to particles passing through the gap is negligible. Before the full factorial trial set preliminary tests were conducted with 349.6 g and < 50 µm particle size resp. 250 – 420 µm particle band as borders of the experiments. The same parameters for this pre-evaluation was used to determine the reproducibility of this experimental setup.

3.2. Results

The trials for the repeatability showed, that for the experiments with a particle size of < 50 µm the standard error is 3.8 % and for the particle size of 250 – 420 % it is 0.3 %. So the experimental methodology can be accepted.

In Figure 4 the results of the centrifuge with 30° angle are shown. The diagram shows, that the desired amount of residual liquid (< 5 %) is achieved for all particle-size-fractions except 0 – 50 µm, where even at highest accelerations the residue liquid amounts to 10 – 15 %. For the biggest particle size an acceleration of 125 g is already sufficient to achieve the aimed residual liquid. Between these two extreme values the area of < 5 % follows a curved line.

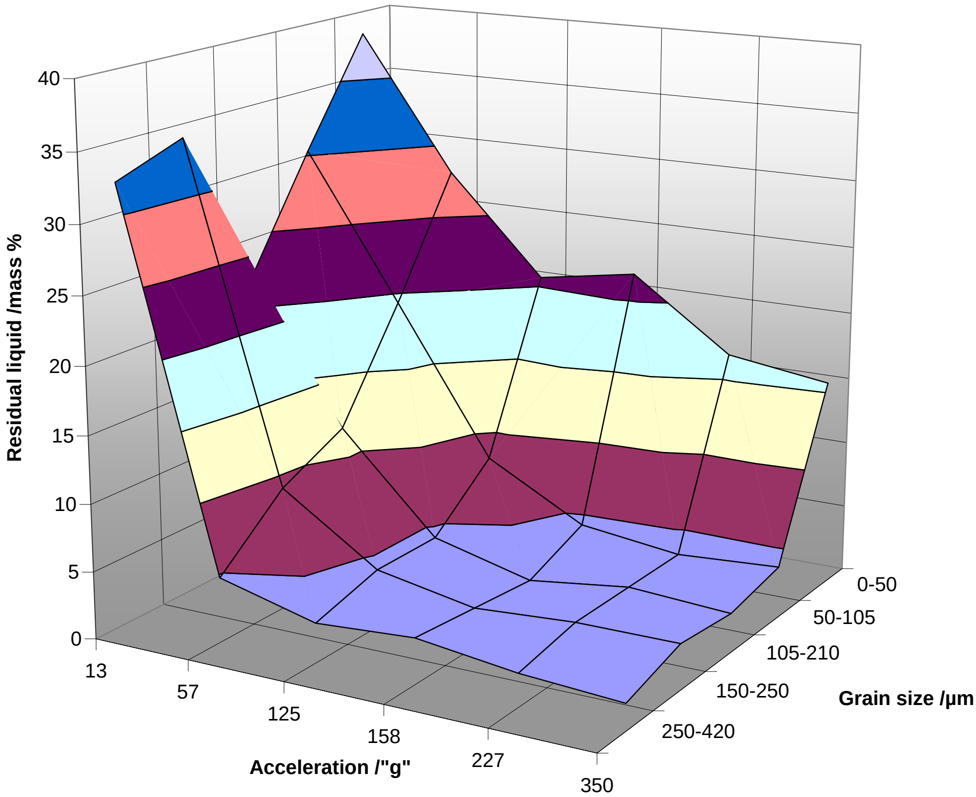

Figure 4: Residual Liquid for centrifuge head of 30°

The results of the centrifuge head with 45° are summarised in Figure 5 and show a similar behaviour of the results of the 30° angle, but the area of < 5 % residual liquid is smaller and the values are shifted to bigger particle size and higher acceleration

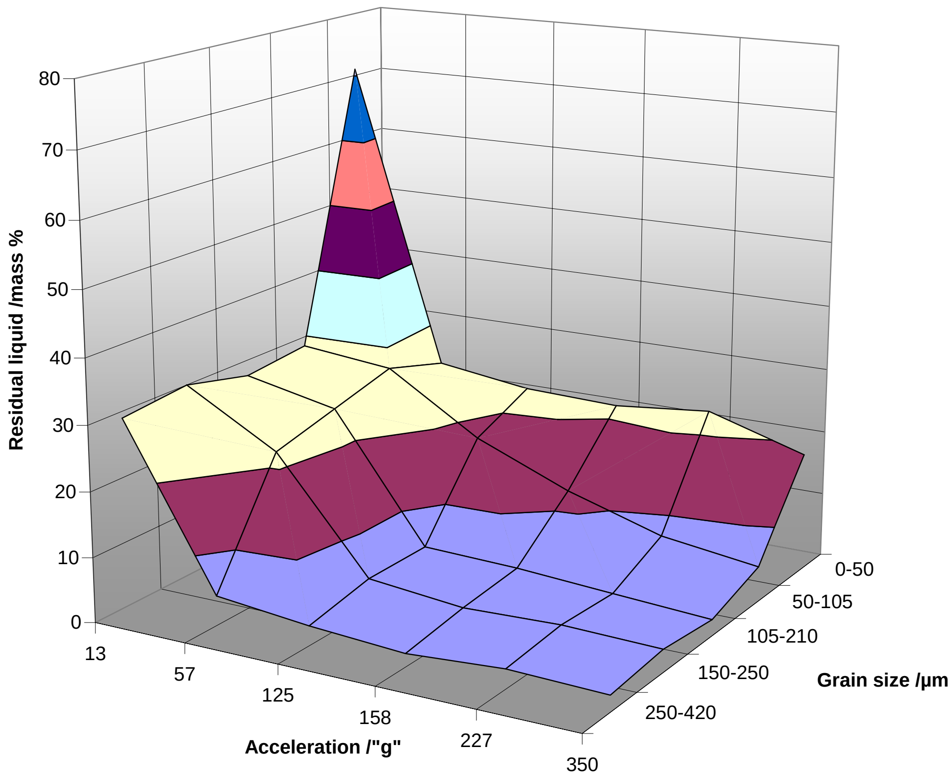

Figure 5: Residual liquid for centrifuge head of 45°

From the centrifugation trials with a 60° rotor head angle as shown in Figure 6 it is obvious, that a very high residual liquid amount remains at very low accelerations and a smaller area of a residual liquid of < 5 % is formed**.**

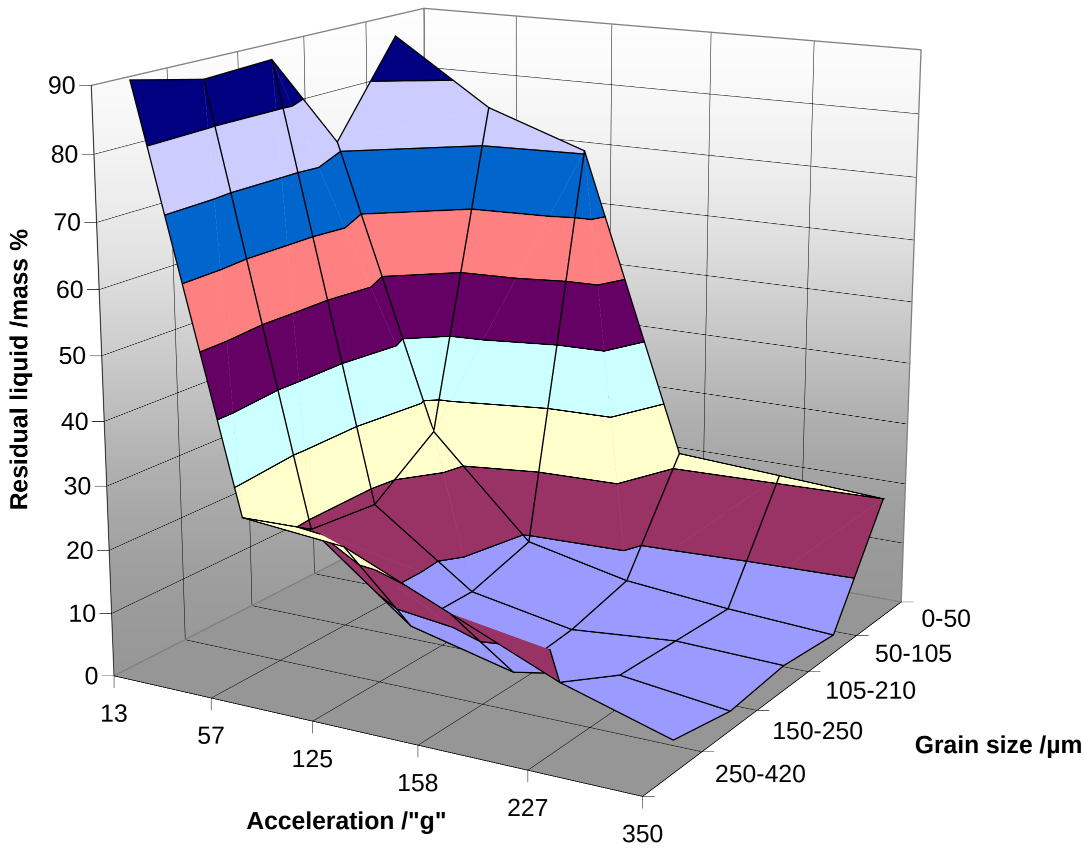

Figure 6: Residual liquid for centrifuge head of 60°

4. Experiments with the melt-centrifuge

4.1. Trial programme with an Aluminium alloy

The experiments of the water model show that a high turning speed always leads to a good separation of solids and liquid. So the spinning was undertaken at the highest possible spinning speed of the equipment that is ~2000 rpm (350 g). For the melt trials only the duration of the spinning period was varied from 10 to 300 seconds. In order to collect particles and to fill the rotor head it was lowered in the melt suspension and slowly turned for 1 min allowing, that almost the same amount of solids were introduced in each trial. The rotor had to be preheated externally before the immersion to avoid freezing of the melt on and in the rotor. For the trials with spinning times of > 30 s the rotor head was also heated externally during spinning. As heating device an acetylene burner was used. For statistical reasons each trial was repeated for 3 – 5 times and the mean value as well as the standard deviation was calculated.

4.2. Centrifugation results of Al-melts

For the evaluation of the residual (capillary) melt in the filter cake a chemical analysis of each centrifuged sample was taken. With the ICP-analysed concentration of Fe and Mn and the assumption that only the intermetallics $Al_{13}Fe_4$ and $Al_6(Mn, Fe)$ develop, the amount of the almost eutectic melt was calculated. One sample of the centrifuged rings is shown in Figure 7.

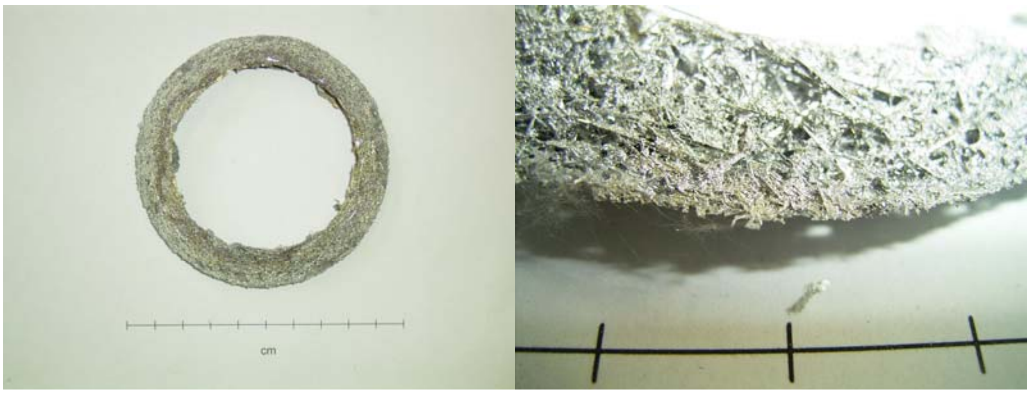

Figure 7: Centrifuged ring and macro picture

The results of the centrifuge rotor with 30°angle show a maximum of 72 % residual eutectic melt after 10 s of spinning, and only ~ 25 % at 5 min of spinning, here the mean value has a high standard error as shown in Figure 8. The results of the centrifuge with 45° angle also shows a lower content of residual liquid at high spinning times, but here the values form a plateau from 60 s to 5 min at ~ 60 %. The samples of the centrifuge with 60° angle did not show any difference of the composition of the filter cake in the centrifuge and the composition of the melt.

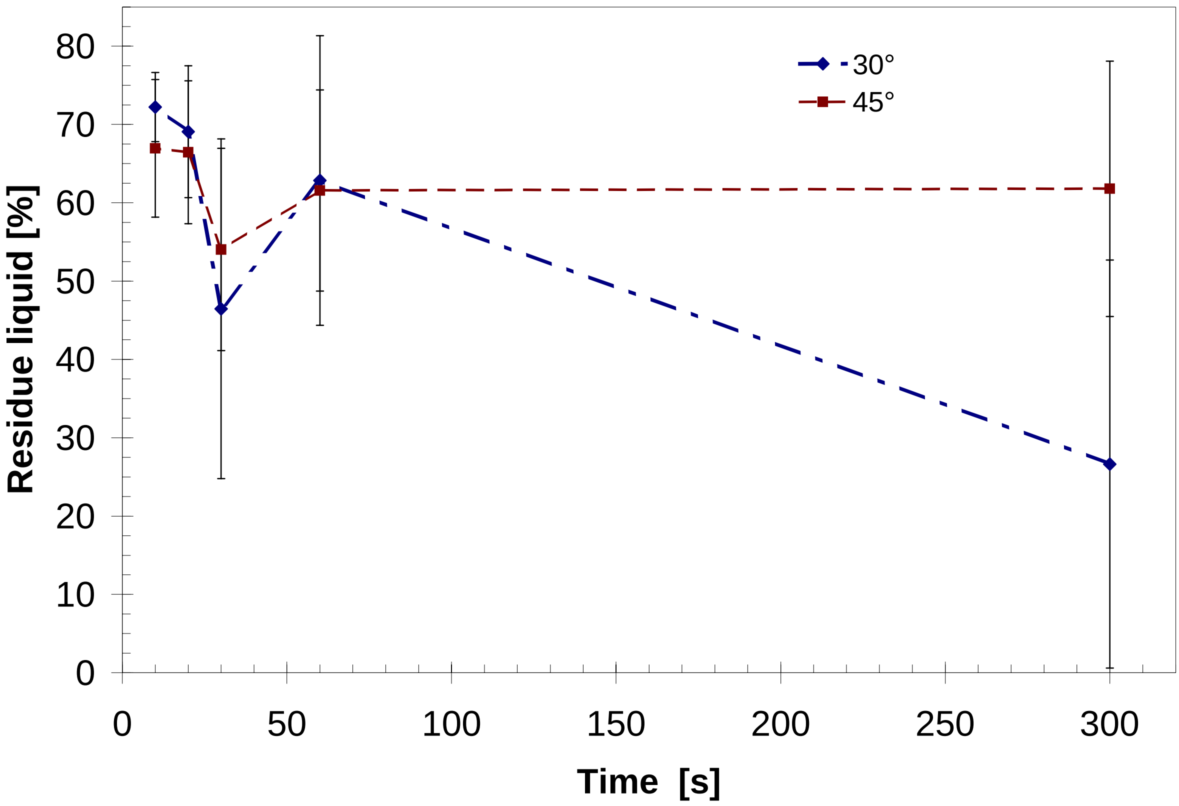

Figure 8: Al-melt trials of the rotor head with 30° and 45° angle

From the developing filter cake cross sections were cut and analysed by REM as shown in Figure 9.

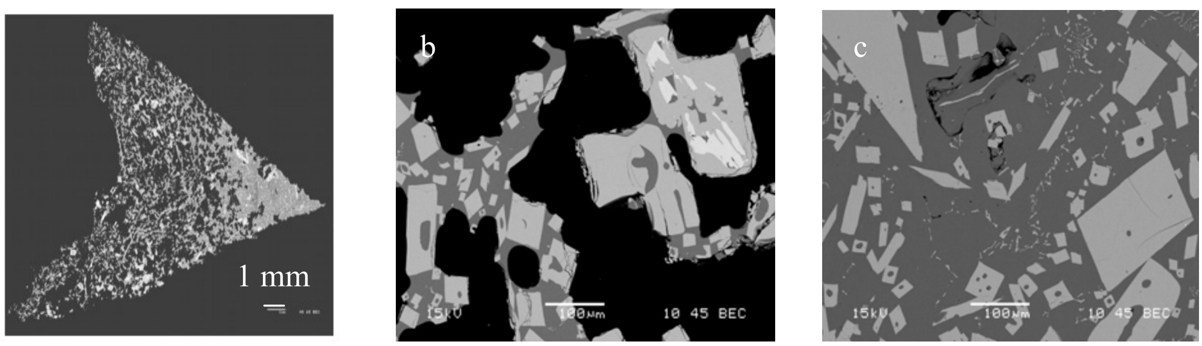

Figure 9: Cross section of the developing filter cake; a) total section, b) left of total, c) right of total.

With the help of the SEM three phases were identified. The white phase represents $Al_{13}Fe_4$ with a composition of 18 wt. % Fe and 6 wt. % Mn. The light grey phase contains 8 wt. % Fe and 6 wt. % Mn and is usually called $Al_6(Mn,Fe)$. The dark grey phase is eutectic melt and the black colour represents porosity. A scan of the cross section revealed, that the area close to the gap contains 65 wt. % eutectic phase, the middle part 60 wt. % and the area close to the rotor centre 25 wt. %. The intermetallics have a needle like shape and a mean diameter of 100 µm while the length exceeds several mm.

The centrifuge with 30° led to filter cakes with a mean weight of 10 g while with the 45° angle mean of 22 g could be realized per dip in the melt. The contents in the filter cake of Fe varied around a mean of 7.7 wt. %, while the amount of Mn had a mean value of 4.6 wt. %. After the trials the remaining melt contained 1.9 wt. % Fe and 0.9 wt. % Mn.

5. Discussion and conclusion

The water model showed a good separation efficiency of water from solid quartz particles of > 50 µm. At accelerations of ~ 350 "g" the residual liquid was lowered to < 5 % by spinning. The influence of the centrifuge head design was rather small but still there is a tendency that smaller angles lead to smaller amounts of residue liquid and so to higher separation yields.

The results of the water model could not be transferred to trials with molten Aluminium with suspended intermetallic particles $Al_{13}Fe_4$ and Al_6 (Mn, Fe). The lowest achieved amount of residual "eutectic" Al-melt in the filter cake was 25 % for a centrifuge with 30° angle. This can be assigned to differences between the water-silica mixture and the Al-melt. The particles in the water model are spherical while the particles in the melt are of needle like shape. It is common knowledge that needles do not have very high volume packing and are actually easier to filter than spheres because they contain less capillary liquid. The surface tension and wetting behaviour between water and silica particles is different compared to liquid Al and intermetallic precipitations. But most likely the high residual liquid results from freezing of the melt inside the rotor head although the head was heated before each trial. Especially the centrifuge head with 60° angle and a very high inner volume showed this effect. It is possible that the plateau of the residual melt > 120 s of the 45° centrifuge is also due to the freezing effect, so that during long spinning times the melt cannot be removed further.

The experimental results show that in order to remove 5 % of intermetallic particles (300 g) from a 6 kg melt with a 30° centrifuge 30 dips would be necessary in comparison to about 15 dips with a 45° centrifuge. This data are valid in case of pure intermetallics $Al_{13}Fe_4$ containing ~43 wt. % Fe and Al_6(Mn;Fe) with 25 wt. % Fe + Mn were present in the filter cake. From the SEM picture it becomes clear that the major part of the particles develops as Al_6(Mn;Fe). The residue calculations show, that the major part of the filter cakes contain about 60 % residue melt, that means including residual liquid the amount of solids is ~ 500 g. That means the number of dips have to be multiplied by 1.6 that leads to 48 dips for the 30° centrifuge and 24 for the 45° centrifuge.

The initial composition of 2.5 wt. % Fe and 1.5 wt. % Mn was reduced to 1.9 wt. % Fe and 0.9 wt. % Mn that is close to the three phase eutectic at 1.7 wt. Fe and 0.7 wt. % [7]. Because the trials were performed 25°C above the eutectic where the composition of the melt shifts to higher values the cleaning efficiency is almost theoretical. The removal of residual melt from the intermetallic filter cake is with an average of 60 % still to high for a good Aluminium efficiency. But one promising trial did show that 25 % residual melt is possible. This motivates future research and a heating of the centrifuge head during spinning will be improved as well as a salt as a washing liquid will be applied. The molten salt should displace the remaining

Al-liquid. Afterwards the salt can be easy separated from the filtered intermetallics by dissolving in water.

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