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Thixoforming raw material development by means of optimized design of experiments (DoE)

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Abstract

Thixocasting and -forging are modern "soft forming" processes in which semi solid slurries with a stiffness of "butter" can be formed at substantially lower pressure and temperature than those, used for conventional diecasting processes.

Thixoforming gains an increasing interest. The automotive industry is focused on process technologies which combines complex parts with improved mechanical properties. A significant potential of energy- and costs- savings can be expected.

Although this process was already developed in 1972 by MIT (Massachusetts Institute of Technology, Cambridge) [1] the required improvement of process stability and the introduction of suitable quality management systems are still to be enhanced and has to start with raw material development.

First this paper gives results of a stock taking and an introduction into various Thixoforming processing routes and each typical process steps. The most important technologies of raw material preparation is presented with special aspects on grain refinement.

On the second hand the development of "thixo-specific" raw materials produced by chemical grain refinement and determination of suitable DC-casting process parameters is discussed. A mathematical experimental planning method is used to optimize scope and type of experiments. A comparison will be given to conventional grain refinement processing.

Introduction

Thixoforming is a process, which uses the behaviour of globular, semi solid suspensions, which is characterised by a decrease of viscosity under the load of shear stress (Fig. 1).

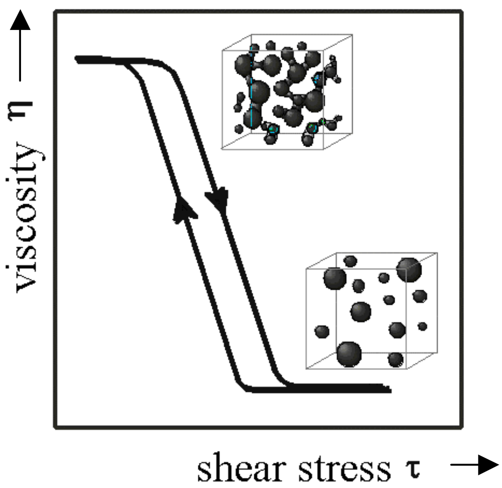


Figure 1: Thixotropic behavior of globular suspensions [(2)]

Compared to diecasting and forging Thixoforming has some important advantages, which are connected to the semi solid status of the material and its typical globular structure. Advantages of Thixoforming compared to diecasting:

- lower casting temperature (for aluminum alloys at least 120 °K lower than for conventional diecasting)
- improved quality of products (less gas content, less porosity, less hot cracks), better weldable
- longer mould life (fewer thermal shocks, less corrosion)
- energy conservation (as a function of the total production cycle)
- higher cycle time

Advantages of Thixoforming compared to forging:

- liberty in shape of components (undercuts are possible)
- not castable alloys can fill complex moldshapes

However besides to these advantages some challenges exist for Thixoforming improvement:

- upgrading of process control and product quality (e.g. decrease of chemical segregation)
- recommendation and clear definition of application possibilities
- predictableness of process parameters intervals for each respective application case
- workability of different materials (and development of thixo-specific developed materials)

Theoretically all types of alloys are applicable for Thixoforming processing, if they indicate at least an expanded melting range of 50°K. The most common type of alloy for Thixoforming is still A356 (AlSi7Mg0.3). The typical work area for Thixoforming is at a temperature range, with 40-60 % liquid fraction (**Fig.** 2).

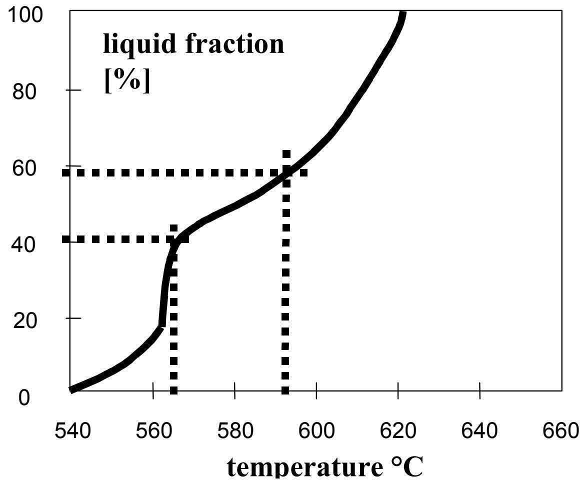


Figure 2: temperature dependence of liquid fraction (A356 – AlSi7Mg0.3)

Today two major Thixoforming processing routes exists, which differ significantly in time- temperature control (Fig. 3).

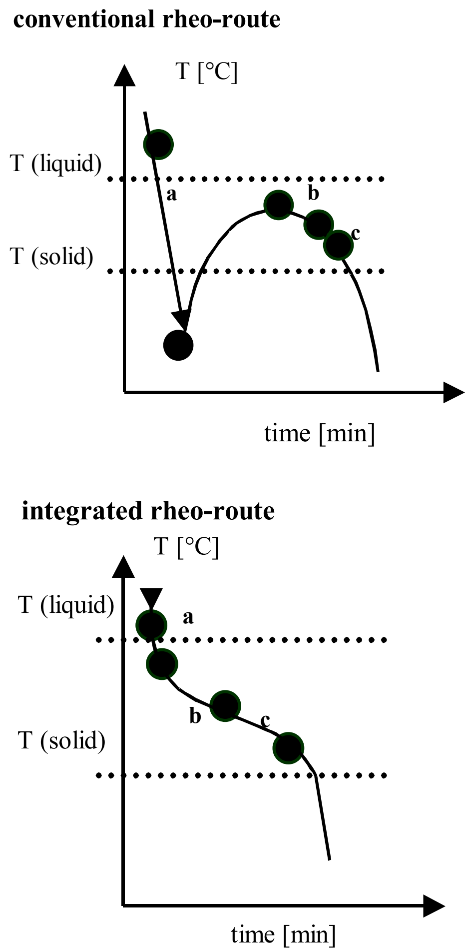


Figure 3: Time- temperature control for different Thixoforming routes

The conventional route is a multi-step process. First an ingot is casted by industrial established continuous casting. During this raw material production step grain refining and avoidance of dendrite growth are achieved by means of a magnetic hydrodynamic field within the sump area of the ingot. The melt is strongly agitated, so that the dendrites break and act as new nuclei of crystals. In the following process step the solid bar is cut into bilettes and warmed up into the semi solid state (**Fig. 3** – conventional process - marker a). This raw material has a remaining stiffness, which permit the feed in the pouring- or shaping machine by a gripper **(Fig. 3** – conventional process - marker b) [(3)]. The alternative process idea follows the target to shorten the process chain. The process steps continuous casting and reheating are replaced by using one integrated process step. UBE Industries, Japan, has developed a innovative process concept, which is according to this idea. From a melting and dosing furnace a slightly superheated melt is poured into a steel crucibel. During filling the crucible the melt temperature is reduced, and the melt quickly moves into the semi-solid state (**Fig. 3** – integrated rheocasting process - marker a). When further cooled the nuclei grow to globulitic, interconnected solid phase, which serves the required stability for the transfer into the casting machine (**Fig. 3** – integrated rheocasting process - marker b) [(4)]. A second integrated rheo-route concept- was developed by Ritter Aluminium Gießerei, Germany. Granulated primary crystals are mixed with melt in a handling chamber, which is coupled with the diecasting machine. By means of a melt launder and a further casting chamber the suspension is directly transported into the diecasting machine [(5)]. All process routes lead to the final forming step, in a diecasting or forging machine (**Fig. 3** – conventional route and integrated rheocasting process - marker c).

For the implementation of the Thixoforming technology into existing casting plants, the new processes offer some operational advantages. Forging plants will probably prefer the conventional processing route, as they can rely on existing technology based on solid raw material.

All processes need to make raw material available, which is particularly suitable for Thixoforming or -casting of semi solid slurries. The most important characteristics of "thixo-specific" raw material are:

- fine grain size (<100µm)
- particle size distribution within a small range and homogeneous distribution
- fine and homogenous distributed eutectic phase
- expanded solidifying temperature range (>50°K)
- low temperature sensitivity of the phase formation
- solid-/ liquid distribution 40-60%
- large shape factor = globular structure in the semi-solid state
- low level of network structure of the globulares (low contiguity number)
- low content of oxides and other impurities
- low sticking inclination of the raw material vs. tool combination

For Thixo-raw material production different grain refining procedures can be applied; they are differed according to the significant nucleation mechanisms:

- chemical grain refinement (addition of grain refiner e.g. AlTi5B1)
- physical grain refinement e.g. by means of mechanical agitation, magnetic-hydrodynamic stirring, strain induced melt activation, undercooling of the melt, ultrasonic vibration)

A comparison of the intense demand of process engineering indicates, that chemical grain refining offers thereby some advantages compared to physical grain refinement methods:

- easy to integrate into conventional processing routes
- no significant modification on equipment required

Therfore the investigations at IME - Process Metallurgy and Metal Recycling, Department and Chair of RWTH Aachen, are focused on thixo-specific raw material development by means of chemical grain refinement methods.

The theoretical mechanisms of chemical grain refining are still discussed [(6, 7 ,8)]. Currently different theories exists to describe how grain refiners work. The "peritectic" model e.g. proposes that the Al-Ti peritectic composition is depressed by the presence of B such that $AlTi_3$ particles, which are known to be potential nuclei, are preserved at low Ti concentration

$$L + TiAl_3 \rightarrow \alpha\text{ - Al (solid solution)} \qquad (1)$$

In this status the nuclei occur slowly as aluminide and dissolve so that performance fades with holding time. As a refinement to the model it has been proposed that Boron in solution may combine at the surface of aluminides to form a protective TiB_2 which preserves aluminides. It is already observed, that the grain refining effectiveness is strongly influenced by alloying elements [(9, 10, 11)].

Own practical chemical grain refining and DC-casting tests were done with the most common Thixoforming alloy A356 – AlSi7Mg0.3. The intend of these tests was, to prove the mechanism also for Thixo-alloys and to develop a thixo-designed raw material with an optimized grain size, ideal globular structure and fine eutectic phase. The effect of alloying elements on grain refinement and alloy development must be worked out. To identify the nucleants, grain center investigations were carried out. In the second part of these studies an adaptation of the continuous casting parameters has been done with respect to homogeneity and surface quality of casting bars.

Experimental

The purpose of the experimental work was to determine the main factors impacting on the grain size, shape factor of the primary dendrites, modification of the eutectic phase and to quantify their effects and interactions. Controlled parameters were:

- AlTi5B1-addition, (0;0.1;0.2;0.25;0.3;0.4 Ti %)
- type of used modifying elements (0; 200 ppm Na and/ or 0; 200 ppm Sr)
- addition of surface-active elements (0; 0.2 % Pb)
- addition of other elements (0; 0.25 % Mn)
- reheating time into the semi-solid state (0; 5; 10; 15; 20; 30 min)

An optimized design of experiments was generated to conduct and plan the experiments in order to extract the maximum amount of information from the collected data in the presence of noise, in the fewest number of experimental runs. The basic idea is to vary all relevant factors simultaneously over a set of planned experiments and then connect the results by means of a mathematical model.

$$y_n = c_0 + b_1 Ti[\%] + b_2 Ti[\%]^2 + b_3 Pb[\%] + ... + b_n Ti[\%] Sr[\%] \quad (2)$$

y_n: response (e.g. grain size, shape factor)
c_0, b_1, b_2,…: coefficients of the effects

For the determined results (e.g. grain size, shape factor) the individual effects (coefficients) and interaction coefficients had been calculated computer aided. This model has been used for interpretation, predictions and optimisation. During this investigation one needs answers to the following questions:

- Which factors have a significant influence on the responses (results)?
- Which factors have significant interactions (synergy's or antagonism)?
- What are the best settings of the factors to achieve optimal conditions for best performance of a process, a system or a product?

An optimised design (D-optimal) of experiments for that mathematical model was applied (**Tab.** I). The run-order has been randomised and includes three repetition tests.

Exp No	Ti [%]	Pb [%]	Mn [%]	Na [ppm]	Sr [ppm]	t [min]
1	0,4	0	0,3	200	200	5
2	0	0,2	0,3	0	0	30
3	0,4	0	0	0	0	0
4	0	0	0	200	200	30
5	0	0	0,3	200	0	30
6	0,4	0,2	0,3	200	200	30
7	0	0,2	0	0	200	30
8	0	0,2	0	200	0	30
9	0,4	0	0	200	0	30
10	0	0	0,3	0	200	30
11	0,4	0,2	0,3	200	200	30
12	0,1	0,2	0,3	0	200	10
13	0	0,2	0	0	0	0
14	0	0,2	0	200	200	5
15	0,4	0,2	0,3	200	200	30
16	0,1	0,2	0	200	0	0
17	0,3	0	0	200	200	0
18	0	0	0,3	0	0	0
19	0,3	0	0,3	200	0	0
20	0	0	0	0	200	0
21	0	0	0	200	0	0
22	0,4	0,2	0	200	200	0
23	0,4	0	0	0	200	30
24	0,3	0	0,3	0	200	0
25	0,4	0,2	0,3	200	200	30
26	0	0,2	0,3	200	200	0
27	0,4	0,2	0	0	0	30
28	0,4	0	0,3	0	0	30
29	0,4	0,2	0,3	200	0	0
30	0,4	0,2	0,3	0	200	0
31	0,2	0,2	0	200	200	30
32	0	0	0	0	0	30
33	0,3	0,2	0	0	0	5
34	0	0,2	0,3	200	0	20
35	0	0	0	0	0	0
36	0,1	0	0	0	0	0

Table I: Experimental design matrix

The alloying of the elements was carried out in an induction furnace with a 2 kg crucible. The melt was heated up to 750°C and the alloying additions were charged -as required- in the following series: lead (commercial pure), manganese (AlMn80), strontium (AlSr10), sodium (technical) and finally grain refiner AlTi5B1. By addition of the grain refiner the desired (**Tab. I**) titanium concentrations were adjusted. The sequence of the addition of alloying elements has been kept constant for all tests. After addition of all alloying elements a preservation time was kept of 5 min with 750°C. Afterwards the melt was poured off into a water-cooled steel mould (∅ = 75-85 mm, h = 200 mm). The produced billets were cut into disks. Samples were taken and chemically analysed. For metallographic investigations the centre of the middle disk has been prepared, too. Samples were embedded in plastic and polished (sandpaper: 220, 320, 500, 1200 granulation, (3µm diamond)) and finally macro etched. All procedures have been according to ASTM Standards E-4 Metallography [12].

Grain size and shape factor are determined with computer aided image analysis. The eutectic phase was analysed, and the morphology was examined, too. In the second part of the practical work DC-Casting parameters (e.g. casting temperature, casting speed, cooling rate) were examined. The heating energy of the furnace is ~ 9 kW, the furnace temperature can be controlled between 400 - 850°C. The capacity is ~ 50 kg aluminium and melt treatment by alloy adjustment, gas treatment, and filtration is applied. The grain center investigations were carried out, with so produced and optimized raw material.

Results

Effect and interaction of alloying elements on:

- grain size:

It can be shown that titanium has a substantial influence on the grain size, as expected (**Fig.** 4). For factors (e.g. Ti, Ti*Sr, Ti*Ti) the values of the effects are plotted sorted (in absolute value) in descending order. The ± 95% confidence interval is drawn as error bars. The effect represent the change in the response (e.g. grain size) when a component varies over its range and all other factors are kept in the same proportion.

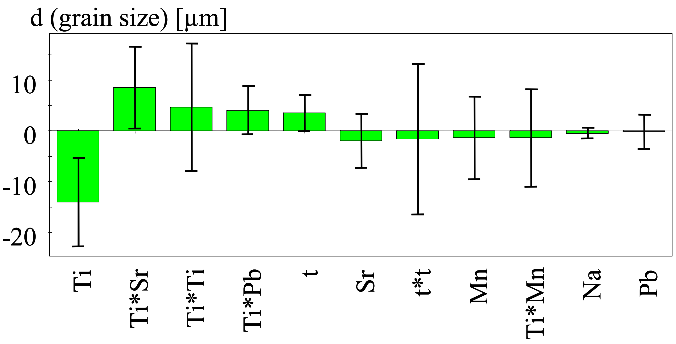


Figure 4: Statistically calculated effects and interactions of measured variables

With increasing titanium addition the grain size of α-aluminum is minimised. The coefficients of the interaction and square terms of the mathematical model additionally considered that the grain size is influenced by the interaction of titanium and strontium. For discussion of the significant effects on grain size therefore the measured variables became in the following detailed analysed. Titanium, strontium content and reheating time have to be considered (**Fig.** 5).

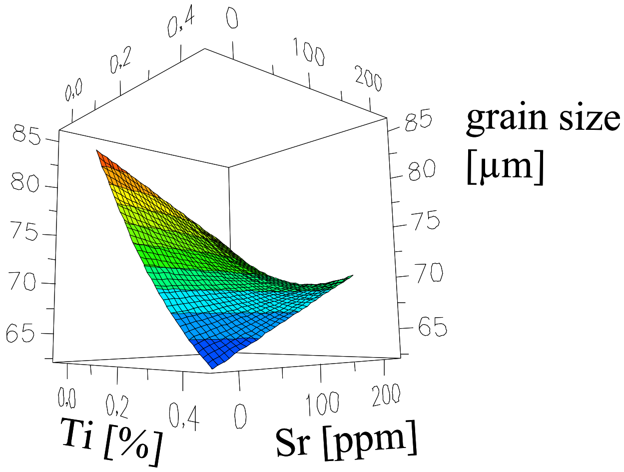


Figure 5: Three-dimensional chart of grain size as function of the most important effects

Grain refining with titanium and additional modification with strontium showed, that modification is counteracting to grain refining for this type of alloy.

<u>- shape factor:</u>

The main significant effect on the shape factor of α-aluminium was the strontium addition. For the description of the influenceableness of the shape factor additionally interactions between titanium and manganese and square terms for the reheating-time had to be considered. Other interactions had to be neglected however. Manganese had an additional effect on the globularisation of the dendrites while reheating.

<u>- modification of the eutectic phase:</u>

Main purpose of the modification is to transform the sharp-edged eutectic silicon into a fine-grained modification. The refinement improves the melt quality, the cast ability, mechanical behaviour, reduces shrinkage-porosity and hot cracking of the raw material [13].

Most important effect on the modification level was strontium. Besides titanium, manganese and lead had a small direct influence, but worked in interactions. The remaining measured variables had negligible influence, in this case.

A "thixo-designed" raw material with 0.25% Ti, 0.3% Mn, 200ppm Sr had been developed. This raw material had the required fine grain size, globular structure during the reheating step and good shaping/ casting characteristics.

<u>- DC-casting of chemical grain refined raw material:</u>

As expected the casting tests have shown, that casting parameters have a direct influence on the quality of the casting bars. **Fig.** 6 shows important reasons for the limits of continuous casting parameters.

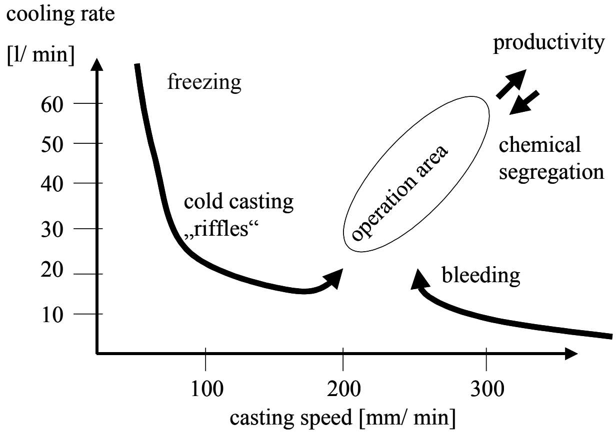


Figure 6: Limits of continuous casting parameters

With high cooling rate and low casting speed the melt starts to freeze in the casting-system. With the same cooling rate and increasing of casting speed this risk for casting breakdown becomes smaller. A further increase of casting speed transfers this region into a region of bad ingot quality, which is characterized by cold casting and "riffles" on the cast bar surface. On the other side high casting speeds and low cooling rates can lead to bleeding of the cast bar. Both reasons are responsible for the demand to optimise the system. With higher casting speed a suitable higher cooling rate must be defined, thereby the productivity can be increased by higher casting speed. Unfortunately the casting quality limits this due to chemical segregation; so limits for the operation area are predetermined.

In addition to these factors further influences on the position and limits of the operation area have to be defined. Contact length of melt in the casting mould, casting temperature, vibration and mechanical agitation (both for support of the grain refining), have to be studied.

<u>- Comparison to MHD grain refinement methods:</u>

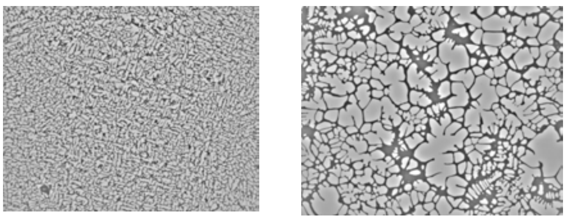
To compare chemical grain refined with conventional (MHD) raw material different commercial starting materials were checked in regard to chemical homogeneity. Samples of different manufacturers were taken out of the centre, 10 mm and 15 mm out of the centre and from the surface of the casting bars. In each case 10 samples were taken and the silicon content had been analysed. The resulting data's have been studied by means of statistical mathematical methods. With this method significant differences can be differed from statistical noise (**Tab.** II).

	Average Silicon contents in raw material			chemical homogeneity
producer	I	II	III	
centre	6,15	6,12	5,98	similar
surface	8,54	6,91	6,79	variance

Table II: Silicon content in raw material of different manufacturers

The enrichment of alloying elements at the surface of the cast bars is well-known as inverse segregation [14]. The inhomogeneity of the raw material was not limited on the silicon content. Most further alloying elements (e.g. Cu, Mg, Mn, Fe, Zn) enriched themselves according to the silicon concentration profile. Titanium and tin showed an opposite tendency. This effect can be explained by thermodynamic phase diagrams.

The chemical grain refined A356 (AlSi7Mg0.3) material shows an increased homogenous chemical composition and structure, compared to conventional MHD material. Chemical grain refined raw material had the same average grain size (dm<20µm) and shape factor (f~0.5) at the surface and the center of the bar. The average grain size of chemical grain refined raw material is nearly the half of conventional raw material (**Fig.** 7).



d_m=11µm, f=0.46 / (0.3%Ti)
chemical grain refined
V=200

d_m= 26µm, f=0.68 (-% Ti)
megneto-hydro-dynamic
V=200

Figure 7: Microstructure of chemical grain refined- and MHD-raw material

This is affected by different nucleation mechanisms, which are examined in detail.

<u>- nucleation mechanisms:</u>

For chemical grain refined- and MHD- raw material $AlTi_3$ and TiB_2 nucleation centers had be identified, as expected. Surprisingly particularly in chemical grain refined raw material Al_4C_3 particles (**Fig.** 8) have been identified as active nucleation centers.

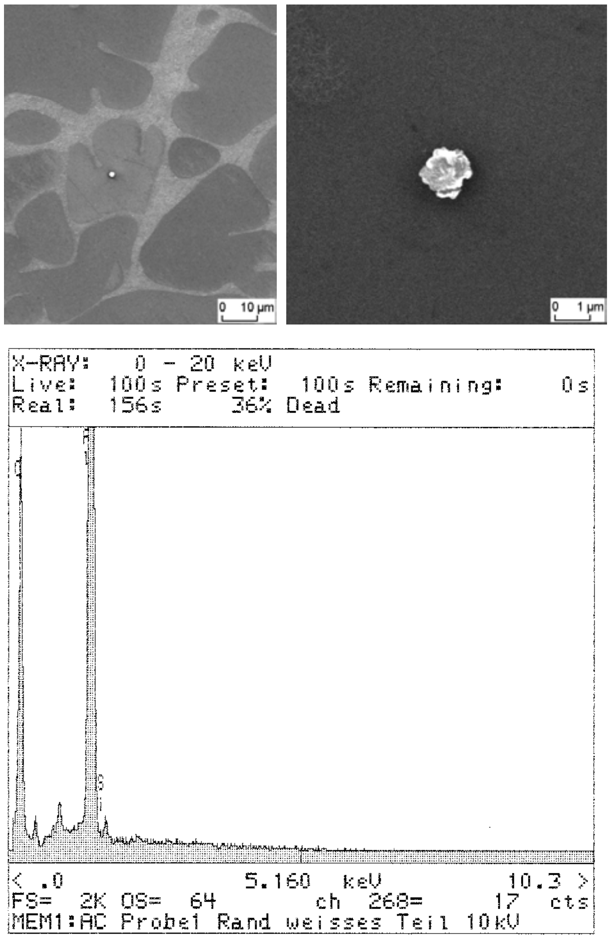


Figure 8: TiC nucleation centers in chemical grain refined raw material

A new potential is to produce chemical grain refined raw material by addition of TiC.

<u>Discussions and next steps:</u>

For Thixo-raw material production chemical grain refinement (addition of AlTi5B1) offers the advantages to be integrated easily into conventional processing routes and to require no significant modification on equipment compared to other raw material production strategies.

Titanium addition has a significant influence on the grain size, as expected. There exists a significant antagonism between grain refinement and modification of the eutectic morphology. To optimise grain size, morphology of the eutectic phase and the globularisation affinity of the Thixo-alloy a compromise has to be defined. The best chemical composition of a chemical grain refined thixo-raw-material based on A356 (AlSi7Mg0.3) is achieved by 0.25% Ti, 0.3% Mn and 200ppm Sr addition. Further alloy development studies are going to be conducted, by extending criteria's e.g. effect of alloying elements on solidification range and rheological behavior in order to design a "Thixo-alloy". According to this also chemical grain refined raw material with TiC addition has to be developed.

The purpose of further continuous casting evaluations is to define the optimum settings of casting parameters and to define position and limits of a suitable operation area for the designed Thixo-alloys.

"Benchmark" studies have started, in order to evaluate the different manufacturing routes. An alteration of the industrial thixo-processing steps (reheating strategy and casting/- forming parameters) will assess the process in regard to stability, overhead demand, expenditure and costs.

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