Tag-and-Trace Method of $\alpha$-Al Crystals Applied to Study Solidification and Casting of Aluminum Alloys

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In this study, a new tag-and-trace method of $\alpha$-Al crystals was developed and used to study the dissolution of a rotating Al-3Si-0.15Ti cylinder immersed into a superheated commercial purity aluminum melt. The developed tag-and-trace method consists of tagging the primary crystals of an alloy with the microsegregation of a peritectic forming solute element, e.g., titanium in aluminum alloys. During solidification, the primary crystals form with a high concentration of the peritectic forming solute, decreasing in the adjacent growth regions of the same crystal. After solidification, the solute microsegregation tag in the interior of the primary crystals can be revealed by color etching. In this work, an Al-3Si-0.15Ti cylinder with all the primary $\alpha$-Al crystals tagged with titanium was immersed into a superheated titanium-free aluminum alloy. The superheat was varied, and all samples were quenched 10s after immersion of the cylinder. The tagged $\alpha$-Al crystals from the original cylinder could be distinguished from the non-tagged $\alpha$-Al crystals formed in the thermally undercooled region surrounding the cylinder and during quenching. Indications of liquid penetration were observed in some $\alpha$-Al crystals, which indicates that disintegration of $\alpha$-Al crystals may occur during stirring of the alloy cylinder into a superheated alloy.

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I. INTRODUCTION

The study of aluminum alloy melting and solidification has experienced significant advances due to new techniques that allow for in-situ studies. For example, synchrotron X-ray tomography has played a vital role in the study of solidification phenomena in aluminum alloys, such as nucleation, dendritic growth, the transition from cellular to dendrite growth, granular deformation, and fragmentation of primary $\alpha$-Al grains. These studies are performed using relatively small samples and can be difficult for some alloy systems to obtain a high absorption contrast between the solute enriched liquid and solid. Additionally, the tomograms’ acquisition times require that the microstructure changes occur over time scales of some seconds to minutes. Therefore, X-ray tomography is suitable for studies of solidification phenomena occurring at low cooling rates or later stages of solidification, i.e., coalescence. In-situ X-ray radiography techniques allow real-time observation of solidification phenomena such as nucleation and growth of primary silicon, $\alpha$-Al, eutectic, and dendrite fragmentation. This technique is used to track individual crystals or particles and in real-time observation of nucleation, growth, and morphological changes of individual crystals. However, for the direct observation of individual crystals from two-dimensions radiography images, the formation of multiple crystals along the direction projected by the incident X-ray beam must be avoided. Therefore, very thin samples, typically <300μm thick, are used in X-ray radiography experiments. New methods to complement the existing techniques to study different phenomena occurring during solidification and at different length scales are necessary.

Color etching is often used for microstructural characterization of materials, e.g., cast iron and aluminum alloys. Weck’s reagent is a color etching technique that forms a manganese oxide film on the surface of aluminum alloys. The interface roughness formed between the manganese oxide film and the aluminum alloy surface and the film thickness determines the different color brightnesses observed in the interior of the $\alpha$-Al phase in the optical microscope. The segregation of elements in the $\alpha$-Al phase’s interior, particularly titanium, affects the interface roughness and
the thickness of the oxide film. This manganese oxide film starts to grow preferentially in the titanium-rich regions in the α-Al phase. For longer contact times, the oxide film also starts to grow in the areas with less titanium concentration in the α-Al phase’s interior. For a specific etching time, the film thickness and interface roughness are different in the interior of an α-Al crystal affected by the local titanium concentration. Therefore, Weck’s reagent can reveal the titanium microsegregation in the interior of the primary α-Al phase. Weck’s reagent also shows silicon and magnesium microsegregation in the primary α-Al phase interior but with less color contrast than titanium.

Aluminum wrought and foundry alloys typically contain solute titanium to control the grain size. Titanium has a low diffusion coefficient in the interior of the α-Al phase, and therefore, the initial titanium microsegregation profile in the interior of the primary α-Al phase is retained for some time at high temperatures. Color etching using Weck’s reagent has been used to evaluate solid fractions of SemiSolid Metal (SSM)-processed aluminum alloys. The Weck’s reagent reveals the microsegregation in the interior of primary α-Al crystals, and, for this reason, the growth layer formed during quenching of SSM castings can be identified. Gao et al. determined the solid fractions of isothermally held A356 aluminum alloys at different temperatures within the semisolid range. The solid fractions were determined from micrographs of isothermally held samples as polished and etched with Weck’s reagent. The results showed that the solid fractions were more accurately determined for the samples etched with Weck’s reagent in comparison to the as-polished samples due to the exclusion of the growth layer formed during quenching of the samples.

Weck’s reagent can also reveal High-Angle Grain Boundaries (HAGB) in the interior of deformed α-Al, as shown in different studies during partial remelting of aluminum alloys.

The titanium microsegregation in the interior of the α-Al phase of aluminum alloys containing solute titanium can also be revealed after anodizing and studied under polarized light in an optical microscope. Titanium microsegregation in the α-Al phase, resulting from the addition of solute titanium, is used in this study to print a tag in each α-Al crystal of an aluminum alloy cylinder. This aluminum alloy cylinder with tagged α-Al crystals was immersed into a superheated titanium-free aluminum alloy. The undissolved α-Al crystals tagged with titanium (revealed by the Weck’s reagent) from the original aluminum alloy cylinder immersed into the superheated alloy are later traced in the solidified samples. This novel tag-and-trace method is used to study the dissolution of a rotating aluminum alloy cylinder immersed into an aluminum alloy at different superheats, procedure comparable to the Rheometal™ process. The Rheometal™ is one of the many rheocasting processes used to produce SSM castings. In this process the superheated alloy is stirred by a lower enthalpy solid alloy, so-called Enthalpy Exchange Material (EEM) to produce the slurry that subsequently is poured in the shot sleeve and injected into the die-cavity.

II. EXPERIMENTAL

A. Materials and Methods

Commercial purity aluminum and silicon were added in a graphite-bonded silicon carbide crucible to prepare about 1kg of Al-3Si-0.15Ti alloy. An Al-10Ti master alloy was used to obtain the intended titanium level and thereby develop a titanium microsegregation profile inside the α-Al crystals. This titanium microsegregation in the α-Al crystals’ interior was used as a “tag” in this study. All individual α-Al crystals formed in the Al-3Si-0.15Ti alloy are tagged with titanium microsegregation. The mixture was melted at 750 °C in an electrical resistance furnace and held at the same temperature for 30 minutes for homogenization. The chemical composition of the Al-3Si-0.15Ti alloy after melting is shown in Table I. After preparation, the Al-3Si-0.15Ti alloy was poured into steel dies to produce Ø30mm cylinders.

The preparation of the alloy and the holding of the crucibles was transferred from the muffle furnace to the experimental setup, as shown in Figure 1(c). During the transfer, the top of the crucibles was covered by an insulation board to minimize heat losses by radiation. The insulation board was removed from the moment that the crucible was placed on the plate below the stirring device, as shown in Figure 1(c). The temperature of the superheated commercial purity aluminum alloy was monitored before the immersion of the cylinder using a type-K thermocouple calibrated against commercial purity aluminum. The Al-3Si-0.15Ti cylinder at room temperature, containing the tagged α-Al crystals,
was immersed while rotating at 900rpm into the commercial purity aluminum melt when the desired superheat was reached. The cylinder was immersed entirely and held at about 20mm from the commercial purity aluminum alloy surface during experimentation. All the samples were quenched 10s after the immersion of the cylinder by lifting the water bath at room temperature until the samples become entirely submerged.

B. Characterization

After casting, the samples were sectioned vertically, and one of the longitudinal cross sections of the samples was etched using Tucker’s reagent at room temperature to reveal the macrostructure. For microscopy, samples were sectioned at the location of the cylinder and about 15mm from the bottom of the longitudinal cross section. After sectioning, the samples were prepared using standard metallographic techniques, with the last polishing step completed using Struers OP-U suspension. After polishing, the samples were etched with Weck’s reagent (4g KMnO₄, 1g NaOH, and 100ml distilled water) with a contact time of 12s to reveal the titanium microsegregation tag inside the α-Al crystals from the original Al-3Si-0.15Ti cylinder. The fraction of α-Al crystals was determined using the manual point count method from ASTM 562-11[28] on micrographs obtained from polished surfaces etched with Weck’s reagent. The fraction of α-Al was determined at about 15mm from the bottom of the longitudinal cross section. A total of fifteen regions were analyzed for each quenched sample. The undissolved α-Al crystals from the original Al-3Si-0.15Ti cylinder were distinguished from the other crystal populations by the titanium microsegregation tag observed in their interior. For the determination of the α-Al crystals fraction in the Al-3Si-0.15Ti cylinder before immersion, a total of 10 different regions were used. The objectives used to determine the α-Al crystals fraction were 5x for the quenched samples and 10x for the Al-3Si-0.15Ti cylinder before immersion. A JEOL JSM-7001F Scanning Electron Microscope (SEM) equipped with Energy-Dispersive X-ray Spectroscopy (EDS) was used to detect the titanium microsegregation in the interior of the α-Al crystals and correlate with the coloring contrasts obtained after etching with Weck’s reagent. A fixed acceleration voltage of 15kV was used during the EDS mapping.

III. RESULTS AND DISCUSSION

A. Macrostructures

Figure 2 shows the macrostructures obtained after immersion of the Al-3Si-0.15Ti cylinder into the commercial purity aluminum alloy at different superheats and quenching. Columnar α-Al grains growing from the surface of the cylinder are observed in the sample obtained after immersion of the Al-3Si-0.15Ti cylinder into the commercial purity aluminum at 670 °C (10 °C superheat), as shown in Figure 2(a). Similar observations were reported in previous studies in which a layer of columnar α-Al dendrites formed on the surface of the cylinder a short time after immersion, denoted freeze-on layer.[27,30] It seems that the dissolution of the Al-3Si-0.15Ti cylinder was minimal 10s after immersion.

| Alloy                  | Si    | Fe | Cu    | Mg | Ti   | Al  |
|------------------------|-------|----|-------|----|------|-----|
| Commercial Purity Aluminum | < 0.80 | 0.13 | 0.002 | 0.001 | < 0.001 | bal. |
| Al-3Si-0.15Ti          | 3.13  | 0.10 | 0.002 | 0.003 | 0.14  | bal. |

Compositions in wt pct.
into the commercial purity aluminum at 670 °C, as shown in Figure 2(a).

A region with equiaxed α-Al grains is observed at the bottom of all samples (Figure 2). Figure 2(b) shows the sample’s macrostructure after immersion of the cylinder into the commercial purity aluminum at 700 °C (40 °C superheat) and quenching. There is no evidence of the original cylinder shape in this sample, which suggests that the cylinder’s disintegration and dissolution have occurred. Consequently, the undissolved, tagged α-Al crystals from the original aluminum alloy cylinder are likely dispersed throughout the sample. These tagged α-Al crystals can be traced after etching the sample with Weck’s reagent. The sample’s macrostructure consists of mostly equiaxed α-Al grains, as shown in Figure 2(b). Columnar α-Al grains, most likely formed during quenching, are also observed, similar to in the columnar α-Al grains observed in Figure 2(a). The increase in the superheat of the commercial purity aluminum alloy likely reduces the number of crystals nucleated on/near cylinder walls and the survival rate of crystals carried into the bulk liquid. However, it is expected that the disintegration of the cylinder contributes with additional α-Al crystals (tagged with titanium microsegregation) and fragments into the bulk liquid.

A fully equiaxed α-Al macrostructure was obtained after immersion of the cylinder into the commercial purity aluminum at 720 °C (60 °C superheat) and quenching, shown in Figure 2(c). No evidence of the original cylinder or formation of columnar α-Al grains was detected in this sample.

A temperature increase of the commercial purity aluminum from 720 (60 °C superheat) to 730 °C (70 °C superheat) produced no significant differences in the macrostructure, as shown in Figures 2(c) and (d), respectively. However, when the temperature of the commercial purity aluminum was increased from 730 °C to 750 °C (90 °C superheat), it seems that more columnar α-Al grains formed near the top of the sample, most likely during quenching, as shown in Figure 2(e). Fewer...
α-Al crystals nucleated on or near the cylinder walls and a decrease of the survival rate of α-Al crystals in the bulk liquid with the increase in superheat may explain the observation of the larger grains in Figure 2(e).

B. Microstructure of the Al-3Si-0.15Ti Cylinder Before Immersion

Figure 3 shows representative microstructures of the Al-3Si-0.15Ti cylinder after polishing and etching with Weck’s reagent.
Weck’s reagent and the respective EDS elemental maps of titanium and silicon. A bright dendritic structure is observed in the interior of the primary α-Al phase. The partition coefficient of titanium in aluminum alloys is larger than one.\textsuperscript{[33]} Therefore, during the solidification of an aluminum alloy containing solute titanium, the first solid formed has a higher titanium concentration than the adjacent growth regions formed at lower temperatures.\textsuperscript{[22]} For this reason, the early dendritic structure formed at higher temperatures (richer in titanium) appears brighter in the interior of the α-Al crystals that grow later with lower titanium concentration and a dark brown color in the microstructure, as shown in Figure 3.

There is a good correlation between the titanium microsegregation in the interior of the α-Al phase in the EDS maps and the different color brightnesses revealed by the Weck’s reagent, as shown in Figure 3. In Figures 3(b) and (c), the brighter areas in the α-Al reveal the early α-Al dendrites formed at high temperatures, richer in titanium compared to the adjacent growth regions.\textsuperscript{[13]} Therefore, the titanium microsegregation tag in the α-Al crystals of the cylinder is revealed after etching with Weck’s reagent, as shown in Figure 3. All the α-Al crystals in the cylinder are tagged with this titanium microsegregation, as shown in Figure 3(a).

C. Microstructures Obtained After Immersion of the Al-3Si-0.15Ti Cylinder into the Commercial Purity Aluminum Alloy

Figure 4(a) shows the cylinder’s microstructure after immersion into the commercial purity aluminum at 670 °C (10 °C superheat) and quenching. The color brightness in the interior of the α-Al phase of the cylinder after immersion is different compared to that observed before immersion, as shown in Figure 3(a). The α-Al phase appears brighter at the periphery near the eutectic regions compared to the core. Additionally, brighter regions can also be observed in the α-Al phase’s interior, generally surrounded by a darker region. The darker regions in the α-Al phase are richer in titanium compared to the brighter regions at the periphery, as shown in Figure 4(a) (see also electronic supplementary Figure S-1). The etching coloring is very sensitive to etching time and temperature.\textsuperscript{[18]} Therefore, the analysis of the etched samples is focused on the different color contrasts observed in the interior of the α-Al phase (revealing the titanium microsegregation) than the comparison of colors between different samples. In the eutectic regions, the morphology and size of eutectic silicon changed due to the increase in the cylinder’s temperature during immersion into the superheated commercial purity aluminum in this sample.

Figure 4(b) shows a representative microstructure of the region containing equiaxed α-Al crystals in the sample of 10°C superheat [see Figure 2(a)]. The microstructure consists of primary α-Al globules, quenched liquid, and a minor fraction of iron-rich intermetallics in the interdendritic regions. The primary α-Al globules were most likely nucleated in the thermally undercooled liquid adjacent to the cylinder wall.\textsuperscript{[34]} When the cylinder at room temperature is immersed into the superheated liquid, a steep thermal gradient is established between the cylinder and the liquid resulting in a thermally undercooled region in the immediate liquid surrounding the cylinder. Therefore, a large number of α-Al crystals nucleate in contact with or near the cylinder surface.\textsuperscript{[35]} The free crystals, not attached to the cylinder surface, were likely transported to the bottom of the crucible. The transport of the free crystals was supported by the flow caused by the rotating cylinder. Fragmentation of the α-Al grains from the freeze-on layer may also occur, and additional α-Al crystal fragments can be mixed into the bulk liquid and carried to the equiaxed α-Al grains region observed in the bottom of the sample.\textsuperscript{[27]} as shown in Figure 2(a). The rapid removal of the superheat and the thermal and compositional homogenization in the liquid promoted by the continuous stirring increased the crystals’ survival rate.\textsuperscript{[36]} Another region consisting of large columnar α-Al grains is observed in Figure 2(a), most likely formed during quenching of the sample. No evidence of tagged α-Al crystals was observed in the region with equiaxed α-Al crystals in this sample. The dissolution of the cylinder was minimal in this sample, as shown in Figure 2(a). Consequently, the fraction of tagged α-Al crystals from the cylinder dispersed in the sample was likely too low to be detectable.

Figure 4(c) shows a large cluster of α-Al crystals showing traces of the early dendrites formed at higher temperatures (richer in titanium) (see electronic supplementary Figure S-2). Therefore, these α-Al crystals tagged with titanium microsegregation were from the original cylinder and were traced in the solidified sample.
after etching with Weck’s reagent. Eutectic silicon is not observed in the interdendritic regions of these large clusters of tagged α-Al crystals, as shown in Figure 4(c). Therefore, the Al-Si eutectic regions of the original Al-3Si-0.15Ti cylinder melted completely in the sample of 40 °C superheat. It seems that a layer of oxides was located at the periphery of the cluster of tagged α-Al crystals (see electronic supplementary Figure S-3). This possible oxide layer promotes a weak bonding between the dislodged piece containing tagged α-Al crystals and the surroundings. A weak bonding between a cylinder surface and the freeze-on layer due to the presence of oxides was reported previously. This layer of oxides most likely results from the oxide layer present on the cylinder surface before immersion and the oxide layer from the commercial purity aluminum top surface wrapped around the cylinder surface during immersion.

In addition to the large dislodged piece from the cylinder containing tagged α-Al crystals [Figure 4(c)], isolated tagged α-Al crystals were also found dispersed in the sample of 40 °C superheat, as shown in Figure 4(d). After melting of the eutectic regions of the cylinder, the long-range bonding that previously existed between α-Al crystals vanished, and the less interlocked crystals (smaller and more globular) can become loose and dislodge into the melt. The disintegration of the cylinder after eutectic melting is determined by the size, fraction, and shape of the tagged α-Al crystals.

The titanium microsegregation tag in the α-Al crystals is still easily traced in the sample of 60 °C superheat, as shown in Figure 5(a). In this sample, the tagged α-Al crystals appeared more uniformly distributed throughout the sample compared to the sample of 40 °C superheat, as shown in Figure 4(c).

Figure 5(b) and (c) shows representative microstructures of the samples of 70 °C and 90 °C superheat, respectively. The titanium microsegregation tag in the α-Al crystals was barely traceable in these samples. The increase of temperature results in a higher diffusion rate of titanium in the α-Al phase, and consequently, the homogenization of titanium concentration in the interior of the α-Al phase occurs more quickly. The color contrasts revealed by Weck’s reagent in the interior of the α-Al are small. However, traces of the original titanium microsegregation tag in the α-Al phase’s interior can still be observed, as shown in Figure 5(b) and (c). Therefore, some undissolved α-Al crystals from the original cylinder remain dispersed in the bulk liquid after immersion of the cylinder into the commercial purity aluminum at temperatures higher than 720 °C, as shown in Figure 5(b) and (c). It seems that coalescence of α-Al crystals starts to occur for superheats higher than 60 °C, as shown in Figure 5(b).

D. Fractions of Tagged and Untagged α-Al Crystals

Figure 6 shows the fraction of tagged and untagged α-Al crystals determined after immersion of the cylinder into the superheated commercial purity aluminum alloy at different superheats and quenching. No tagged α-Al crystals were detected in the sample of 10 °C superheat, as shown in Figure 6. The increase in temperature from 670 to 700 (10 °C to 40 °C superheat) resulted in the dispersion of large dislodged pieces and individual tagged α-Al crystals from the original cylinder throughout the sample, as shown in Figure 4(c) and (d). Therefore, a fraction of 0.07±0.02 of tagged α-Al crystals was determined for the sample of 40 °C superheat, as shown in Figure 6. The increase in temperature results in a higher dissolution rate of the tagged α-Al crystals from the original cylinder dispersed in the liquid. Therefore, the fraction of tagged α-Al crystals
decreased with the increase of superheat from 40 to 60 °C, as shown in Figure 6. No significant differences were found for the fraction of tagged \( \alpha \)-Al crystals for temperatures higher than 720 °C (60 °C superheat).

It would be expected that with the increase of temperature, the fraction of \( \alpha \)-Al crystals formed in the thermal undercooled region surrounding the cylinder would decrease while the fraction of the \( \alpha \)-Al crystals formed during quenching would increase. However, the increase of temperature of the commercial purity aluminum alloy did not result in a clear trend for the fraction of \( \alpha \)-Al crystals formed in the thermal undercooled region surrounding the cylinder, as shown in Figure 6. Additionally, the fraction of \( \alpha \)-Al crystals formed during quenching decreased with the increase of temperature, as shown in Figure 6. It is possible that the fractions of the untagged \( \alpha \)-Al crystals obtained are inaccurate due to the insufficient cooling of such large samples. This is supported by the fact that the two populations of untagged \( \alpha \)-Al crystals could not be distinguished for the samples of superheats higher than 60 °C. It is important to note that the fractions of the different crystals populations were determined at about 15mm from the bottom of the longitudinal cross section. Therefore, the layer of crystals formed around the cylinder (so-called freeze-on layer) for the sample of 60 °C superheat was shown in Figure 6. The equiaxed crystal region at the bottom of the 10 °C superheat sample was more homogeneous compared to the top region where the cylinder remained almost intact. Therefore, the fractions of the different crystals populations were determined at about 15mm from the bottom for all samples.

E. Liquid Penetration

Weck’s reagent can also reveal HAGB in the interior of the \( \alpha \)-Al phase.\textsuperscript{[20]} Figure 7 shows the microstructures of the samples of 40 °C superheat and 60 °C superheat. Figure 7(b) shows a black line, crossing through the \( \alpha \)-Al phase, revealed by Weck’s reagent, suggested to be HAGB by Gao et al.\textsuperscript{[20]} Additionally, liquid penetration seems to occur in both tagged \( \alpha \)-Al crystals from the original cylinder, and untagged crystals nucleated in the thermal undercooled liquid adjacent to the cylinder wall after immersion, as shown in Figure 7(a) and Figure 7(b), respectively. The disintegration of \( \alpha \)-Al crystals can be occurred by liquid wetting at the HAGB\textsuperscript{[7]} formed in \( \alpha \)-Al crystals bent at high temperatures.\textsuperscript{[37,38]} Liquid wetting followed by disintegration can occur in the tagged \( \alpha \)-Al crystals from the cylinder and those nucleated in the thermal undercooled region, as shown in Figure 7.

The tagged \( \alpha \)-Al crystals traced in the sample of 60 °C superheat [Figure 5 (a)] appear more dispersed compared to that observed at 40 °C superheat [Figure 4(c)]. The dissolution rate of the \( \alpha \)-Al crystals at higher superheats and their disintegration by liquid wetting may promote a quicker dispersion of the tagged \( \alpha \)-Al crystals from the cylinder in the bulk liquid.

F. Tag-and-Trace Method to Study Solidification

In this study, the potential of a new tag-and-trace method of \( \alpha \)-Al crystals was shown in a study of the dissolution of a solid alloy immersed into a superheated alloy. Titanium was added to an aluminum alloy to tag all the \( \alpha \)-Al crystals with titanium microsegregation which remained and could be detected. The peritectic forming solute titanium has a partition coefficient, \( k > 1 \), and a low diffusion rate in the \( \alpha \)-Al phase.\textsuperscript{[22]} During the solidification of a titanium-containing aluminum alloy, the first solid to form has a high titanium concentration that decreases in the adjacent growing regions of the same crystal.\textsuperscript{[22]} A distinctly dendritic structure (higher titanium concentration) was observed in the interior of the \( \alpha \)-Al phase in aluminum alloys containing solute titanium after anodization\textsuperscript{[22]} or etching with Weck’s reagent.\textsuperscript{[21]} Therefore, when solute titanium is added to an aluminum alloy, a “tag” (titanium microsegregation) is printed in all the primary \( \alpha \)-Al crystals formed during solidification, which can be revealed by color etching or anodizing. Like titanium, other peritectic forming solutes in aluminum alloys such as chromium,\textsuperscript{[39]} vanadium, zirconium, and niobium\textsuperscript{[40]} may be used to tag the primary \( \alpha \)-Al crystals and later trace the tagged crystals in the solidified samples.

Similarly, the addition of the peritectic forming solute zirconium into pure magnesium resulted in zirconium rich core structures observed after solidification in the magnesium grains in the backscattered electron mode.\textsuperscript{[41,42]} Therefore, magnesium crystals can be tagged by the addition of solute zirconium. The addition of the peritectic forming solute copper into pure zinc tags the \( \eta \)-Zn grains with a distinctly dendritic structure that is observed using the optical microscope under polarized light after color etching.\textsuperscript{[43]} These dendritic structures in the interior of the \( \eta \)-Zn grains reveal copper microsegregation. Therefore, this tagging method of primary crystals formed in an alloy by the microsegregation of a peritectic forming solute is not limited to aluminum alloys. Different characterization techniques can be used to reveal the tagged primary crystals in different alloys.
after solidification, such as color etching, anodizing, composition measurements, or backscattered electron images depending on the alloys and solute elements used.

In this study, the tag-and-trace method of z-Al crystals with titanium microsegregation was used to study the dissolution of an aluminum alloy cylinder immersed while rotating into a superheated aluminum alloy. This procedure is comparable to the Rheometal™ process used to prepare slurries that later are injected into a die-cavity to produce high integrity semisolid process used to prepare slurries that later are injected into a die-cavity to produce high integrity semisolid castings. Niu et al. [32] used a transparent model alloy to study the grain refinement that result from the addition of a solid alloy into a superheated liquid alloy. Eskin and Wang [44] studied the grain refinement mechanisms active during the immersion of a solid alloy into a superheated alloy combined with ultrasonic vibration by post-solidification metallographic characterizations. Both Niu et al. [32] and Eskin and Wang [44] studied alternative methods of grain refinement to improve the quality of products obtained by continuous casting processes, and the tag-and-trace method is perfectly suited to be applied in these type of studies.

The size, fraction, and morphology of tagged crystals in a slurry can be controlled by a careful selection of the alloy, weight fraction, cooling conditions, and the temperature before immersion of the alloy cylinder immersed into a superheated alloy. After the cylinder alloy’s dissolution, a certain fraction of tagged crystals with a certain size and morphology are dispersed in the slurry. After preparation, the slurry with both tagged and untagged crystals surrounded by liquid can be gravity poured (for low solid fractions) into an open-end die to allow a continuous flow through the cavity until the progress of solidification interrupts the flow. The tagged crystals can later be traced in solidified castings by color etching or other techniques to study the flow and solidification phenomena during die-casting of aluminum, magnesium, or zinc alloys.

The semisolid welding joining process is a new process used to produce welds with low gas entrapment and shrinkage porosity. In this process, a partially solidified filler material is deposited continuously on the joint section to join two different parts. The change of the welding parameters, joint shape and size, fraction, and morphology of the partially solidified filler material’s primary crystals can result in different flow and solidification conditions during the semisolid welding process. A filler material with a certain fraction of tagged crystals can be used to study the flow and solidification in the joint region during semisolid welding by tracing the tagged crystals in the solidified joint.

IV. CONCLUSIONS

A new tag-and-trace method of z-Al crystals was used to study the dissolution of a rotating aluminum alloy cylinder immersed into a superheated aluminum alloy. Solute titanium was added to the aluminum alloy used to cast the cylinder to tag all the z-Al crystals with titanium microsegregation. This titanium microsegregation tag in the interior of the z-Al crystals was revealed by color etching. Similar to titanium, other peritectic forming solutes in aluminum alloys can be used to tag the z-Al crystals. After the cylinder’s disintegration, the tagged z-Al crystals from the original cylinder dispersed in the sample were traced among the non-tagged z-Al crystals formed in the thermally undercooled liquid adjacent to the cylinder walls and during quenching. Liquid wetting at high-angle grain boundaries and coalescence of z-Al crystals can also be detected by color etching. This new tag-and-trace method has the potential to be extended to different alloy systems and can be used to study different solidification phenomena in die-casting or semisolid welding joining processes of aluminum alloys and other alloy systems.

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CONFLICT OF INTEREST

The authors declare that they have no known conflicts interest or personal relationships that could have appeared to influence the work reported in this paper.

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SUPPLEMENTARY INFORMATION

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