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Determination of the growth restriction factor and grain size for aluminum alloys by a quasi-binary equivalent method

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ABSTRACT

In the present research paper is suggested a new methodology to determine the growth restricting factor (Q) and grain size (GS) for various Al-alloys. The present method combines a thermodynamical component based on the liquidus behavior of each alloying element that is later incorporated into the well known growth restricting models for multi-component alloys. This approach that can be used to determine Q and/or GS based on the chemical composition and the slope of the liquidus temperature of any Al-alloy solidified in close to equilibrium conditions. This method can be modified further in order to assess the effect of cooling rate or thermomechanical processing on growth restricting factor and grain size. In the present paper is proposed a highly accurate (R^2 = 0.99) and validated model for Al–Si alloys, but it can be modified for any other Al–*X* alloying system. The present method can be used for alloys with relatively high solute content and due to the use of the thermodynamics of liquidus this system considers the poisoning effects of single and multi-component alloying elements.

1. Introduction

The microstructure refinement of aluminium castings is highly desirable, fine and equiaxed grains are usually the "preferred" microstructures. This is because finer grains reduce the possibilities for hot tearing and promote a higher homogeneity and may improve mechanical properties and service characteristics [1]. A fine grain structure in aluminium alloys cast components can be produced by promoting heterogeneous nucleation. A larger number of crystals impinge on each other hindering grain growth. Different authors have distinct methods to assess the growth restriction factor (Q) or the amount of constitutional undercooling (*P*) for non-diluted or multi-component alloys [1–9].

The mechanism of grain refinement using chemical agents can be divided in two types. First type assumes that the nucleation particles are of ultimate importance while second type recognizes the solute essential to achieve finer microstructures [1,2,4,9,10]. The solute theory, formalized by suggests that both the nucleants and the segregation have influence in the grain refinement. Q was developed to determine the degree of segregation. Q is a measure of the growth restricting effect of solute elements on the growth of solid-liquid interface of the new grains as they coarse into the liquid metal. Based on the binary Al–X_i system the Q_{X_i} is defined as $m_{X_i}(k_{X_i} - 1)C_O$, where m_{X_i} is the slope of the liquidus temperature line based on the phase diagram, C_o is the concentration of the solute in the melt, and k_{X_i} is C_S/C_L, where C_S and C_L are the solute concentrations at the interface for the solid and liquid respectively. *k* is also known as the equilibrium partitioning coefficient. Typical values of the Q parameters for common alloying elements are given in Table 1.

In theory Q and *P* can be estimated by adding the respective Q for each element present in the alloy. However, the theoretical results for this method grossly overestimate the actual value. Some authors estimate Q in multi-component alloys as the sum for each element [11]. The supercooling [11] and the growth restriction [12] parameters can be determined as follows:

$$P = -\frac{m(1-k)C_0}{k} \quad (1)$$

$$Q = m_i(k-1)C_0 = kP \quad (2)$$

where *m* is the liquidus slope, *k* is the equilibrium partition coefficient and C_0 is the alloy composition. The limitations of *P* is that this parameter is equal to the freezing range of the alloy assuming that both, the liquidus and solidus, lines are straight and the alloy solidifies as a solid solution. Therefore, *P* is the degree of undercooling induced by growth restriction.

Si is an example an element with dual function in the Al–Si systems. In the Al–Si system the GS decrease first with additions of up to 1.5 wt% Si and tapers off at approximately 3 wt% Si; further additions have a poisoning effect resulting in grain growth [13–17]. An

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Nomenclature

Q	growth restricting factor
GS	grain size
R^2	regression coefficient
P	constitutional undercooling
m_{X_i}	slope of the liquidus line
C_o	concentration of the solute in the melt
k	equilibrium partitioning coefficient
k_{X_i}	partition coefficient of individual element
k_i	partition coefficient of all elements
C_S	solute concentration in the solid
C_L	solute concentration in the liquid
d	grain size in diameter (Refs. [3,19,20])
T_{Liq}	liquidus temperature
T_{DCP}	temperature at the dendrite coherency point
($T^{E,NUC}_{AlSi}$)	temperature at the Al–Si eutectic nucleation
($T^{E,NUC}_{AlSiCu}$)	temperature at the Al–Si–Cu eutectic nucleation
T_{SOL}	solidus temperature
α-Al	primary Al grains
df_s/dt	cooling rate
Amount of Ti	wt% of Ti added to the melt
Q_d	grain growth index for casting with minimal GS
Q_b	grain growth index for original aluminum based material
C_{Eq}	carbon equivalent
Si_{Eq}	silicon equivalent
$X_{i_{Eq}}$	sum of the contribution of all elements expressed as the equivalent effect of individual element on the liquidus temperature
$a_0^{X_i}$, $b_0^{X_i}$ and $c_0^{X_i}$	polynomial coefficients
X_i	the concentration of the individual element in wt%

analysis of all the GS and thermal analysis data suggested that the GS in the above mentioned references was controlled by the rate of grain growth. The restriction effect of crystal growth by solute segregation increases with the partition coefficient (k_i) of all the elements. Therefore, the total Q should be given by Eq. (3).

$$Q_{TOT} = \sum m(k-1)C_0 = \sum m_1(k_1-1)C_1 + \sum m_2(k_2-1)C_2 + \ldots \quad (3)$$

Eq. (3) is valid for dilute alloying systems or alloys with elements that have limited or none interactions among the constitutional elements of the alloy. Therefore, this may result in an over or under-estimation of Q and prevents the accurate determination of GS.

Table 1
Data required for calculation the Q for binary Al alloys [5].

Al–X_i alloy	k_i	m_i	$m_i(k_i-1)$
Al–Si	0.11	−6.6	5.9
Al–Cu	0.17	−3.4	2.8
Al–Mg	0.51	−6.2	3.0
Al–Mn	0.94	−1.6	0.1
Al–Fe	0.02	−3.0	2.9
Al–Zn	0.88	−2.97	0.3
Al–V	4.00	10.0	30.0
Al–Mo	2.5	5.0	7.5
Al–Nb	1.5	13.3	6.6
Al–Ge	0.10	−4.66	4.2
Al–Li	0.56	−8.06	3.8
Al–Ni	0.007	−3.3	3.3
Al–Cr	2.00	3.5	3.5
Al–Ti	7.8	33.3	220.0
Al–B	0.067	1.015	3.2

Although, Eq. (3) has the advantages that it is accurate for dilute multi-component aluminum alloys and the approximation of Q and GS is conducted by a single function.

Easton et al. [3,13] and Lee et al. [14] developed empirical models (Eqs. (4) and (5), respectively) to determine the GS (d) as a function of Q and P for aluminum alloys. Eqs. (4) and (5) have been developed based on the effect of grain refiner (TiB_2) for Al-alloys.

$$d = \frac{32}{\sqrt[3]{pct\ TiB_2}} + \frac{652}{Q} \quad (4)$$

$$d = \frac{43}{\sqrt[3]{pct\ TiB_2}} + \frac{520}{Q} \quad (5)$$

1.1. Utilization of thermal analysis in grain refinement

The thermal analysis methodology allows for the assessment of several thermal characteristic such as solidification temperatures (e.g. liquidus – T_{LIQ}), dendrite coherency point (T_{DCP}), Al–Si eutectic nucleation ($T^{E,NUC}_{AlSi}$), Al–Si–Cu eutectic nucleation ($T^{E,NUC}_{AlSiCu}$) and solidus (T_{SOL}); as well as the Latent Heat of solidification and structural characteristics for multi-component 3XX Al alloys. In fact, the difference between the liquidus temperature and the undercooling for the nucleation of the α-Al grains is proportional to the number of nuclei particles. The smaller the undercooling, the larger the number of solidified nuclei particles resulting in a finer microstructure, thus smaller grains [15]. A valid correlation based on the constitutional undercooling and the number of grains can be established.

Bäckerud et al. [18] developed an algorithm showing a detailed description and understanding of how the composition of aluminum alloys affects its self refinement. Such algorithm can be used to calculate the desired level of grain refinement. The coherency point can be determined by thermal analysis based on the gradient temperature during solidification (df_s/dt) using a two thermocouples approach; one at the center of the sample and the other close to the wall [5,13,19]. The temperature gradient between both thermocouples is accumulative and increases during the initial stage of solidification. The maximum of this temperature gradient occurs at the coherency point, and then decreases after the gradient becomes lower. An alternative method using a single thermocouple approach is possible by analyzing the first derivative of the cooling history of a solidifying alloy as shown in Ref. [20].

An alternative method to determine the amount of Ti needed to reach a desired GS can be calculated with Eq. (6). Eq. (6) ignores the undercooling that takes place during the alloy solidification but considers the slope of the liquidus temperature (m_{Ti}) and the distribution coefficient of Ti (k_{Ti}) of the Al–Ti system [20].

$$\text{Amount of Ti} = \frac{Q_d - Q_b}{(k_{Ti}-1)m_{Ti}}[\text{wt\%}] \quad (6)$$

The "Amount of Ti" is the percentage by weight of Ti to be added to the melt, Q_d is the grain growth index resulting in aluminum castings having a minimal GS, Q_b is the grain growth index of the original aluminum base material.

1.2. Quasi-binary equivalent concept for grain refinement

To date, there are no comprehensive procedures for rapid prediction of structural properties of multi-component aluminum alloys in as-cast and/or heat treated conditions based on the alloy chemical composition. The equivalent concept proposed herein is similar to the well know carbon equivalent (C_{Eq}) concept used in ferrous alloys where carbon is the major alloying element [21,22]. Similar concept has been developed for the hypoeutectic and hypereutectic [5]. Similar concept has been developed for the hypoeutectic and hypereutectic [5,19] Al–Si alloy systems where