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Calculation of the liquidus temperature for hypo and hypereutectic aluminum silicon alloys

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Abstract

An accurate knowledge of liquidus temperatures permits the prediction of a variety of metallurgical characteristics for a given alloy, including the melt treatment and casting temperatures (superheat). Three equations to determine liquidus for hypoeutectic aluminium alloys are reported in the literature. There is, however, no algorithm for calculation of the liquidus temperature for hypereutectic Al–Si alloys. In this paper algorithms are developed to predict the liquidus temperature of both hypo and hypereutectic 3XX series of aluminium alloys, based on their chemical compositions (minor and major elements). These algorithms are based on the new "*element-equivalency concept*". The accuracy of the calculated liquidus temperatures for 3XX hypo and hypereutectic alloys were statistically examined with the values measured by the aluminum thermal analysis platform (AlTAP).

Keywords: Aluminum alloys; Hypoeutectic; Hypereutectic; Liquidus temperature; Silicon equivalency; Quality control

1. Introduction

The modeling and control of casting processes has remained a topic of active interest for several decades, and the availability of numerous software packages (Magma, Thermo-Calc, Fact-Sage, Pro-Cast, Calphad, etc.) on the market, are a good indication of the importance that casting industries and researchers put in this field. Most of the data used in the above listed software packages are based on binary or multi-component phase diagrams but unfortunately; except for binary diagrams, many ternary or higher order phase diagrams are still not accurate enough for this purpose. Keeping in mind that most aluminum binary systems are very well established, the transferring of a multi-component system into a high reliable Al–Xi "*quasi-binary*" system has great industrial and research potential. This type of system could be used to calculate several thermo-physical and solidification process parameters of multi component aluminum alloys in as cast or melt treated conditions. These calculations are very accurate and can predict properties for relatively slow cooling rates (e.g. $\partial T/\partial t \leq 0.1\,^{\circ}\mathrm{C/s}$) [3].

In order to predict the various physical and metallurgical parameters of a solidifying aluminum casting alloy (e.g. fraction solid, the position of the moving boundary of the mushy zone, the microstructure and chemical composition of the alloy at the solid/liquid interface, the solute super saturation point, the degree of super cooling, etc.) the liquidus temperature of the alloy must be known with a high degree of accuracy. A few equations that are reported in the literature relate limited range of compositions of some hypoeutectic aluminum alloys to their liquidus temperatures [3,6,9]. However, there is no equation available to calculate the liquidus temperature of hypereutectic Al–Si alloys.

The literature on iron based alloys offers the comprehensive data for calculation of the liquidus temperature for a known chemical alloy composition, but there is no analogic methodology for aluminum alloys. A new relationship developed for aluminum multi-component alloys was titled the "silicon equivalent" (Si_{EQ}). The Si_{EQ} algorithm summarizes

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the effect of minor, major elements, and hydrogen present in the 3XX aluminum alloys, and expresses their influence on the liquidus temperatures.

Using the Si_{EQ} methodology some other thermo-physical properties such as fraction solid [4] latent heat [5] and characteristic solidification temperatures [3] can also be calculated. One of the equations found in the literature for calculating the liquidus temperature of hypoeutectic aluminum alloys (T_{LIQ}), based on a known chemical composition, was proposed by Drossel [6] using the multiple regression analysis of the experimental data:

$$T_{LIQ} = 661 - 4.97Si - 0.15(Si)^2 - 6.13Cu - 17.4Mg + 2.72Zn + 5.08CuMg\,(^\circ C) \quad (1)$$

Unfortunately, Eq. (1) is valid only for the following ranges of chemical compositions (expressed in wt.%), therefore its applicability is limited:

$Si \leq 9.30$; $Cu \leq 2.50$; $Mg \leq 0.60$;

$Fe \leq 1.15$; $Mn \leq 0.40$; $Zn \leq 0.63$;

$Ni \leq 0.43$; $Ti \leq 0.05$

The next equation for predicting T_{LIQ} of aluminum alloys, for Al–Si–Cu systems, recently developed by Vijayaraghavan et al. [9], based on the aluminium–silicon–copper ternary phase diagram through the use of multiple regression analysis is given below:

$$T_{LIQ} = 664 - 6.9Si - 2.5Cu\,(^\circ C) \quad (2)$$

Eq. (2), based on Al, Si and Cu concentrations only, fails to account for the potentially important influence of other elements (for example, Mg, Mn, Fe, Zn and Ti) therefore substantial error may be involved. This is evident from the fact that the equation's constant (664 °C) is considerably higher than the known melting point of pure aluminum (660.452 °C).

The purpose of this paper is to develop the general method for the calculation of the liquidus temperature of multi-component hypo and hypereutectic aluminum alloys, based on their known chemical compositions. The accuracy of the developed algorithms will be determined by comparing calculated values of the liquidus temperatures with the measured values by using the high resolution technique known as the aluminum thermal analysis platform (AlTAP). This comprehensive novel technology was developed by the NSERC/FORD-NEMAK/University of Windsor Industrial Research Chair (IRC) in light metals casting technology for advanced analysis of melt characteristics [3–5,7,8].

2. Calculation of the liquidus temperature of 3XX Al alloys

For a binary aluminum alloy the liquidus temperature of a given chemical composition, can easily be derived, with a high level of accuracy, from its respective phase diagram. However, there are only a limited number of sources that reliably report the relationship between liquidus temperature and chemical composition for a multi-component hypoeutectic aluminum alloy, that can be used for industrial applications [2,6], and moreover these equations do not address melt treatment or its cleanliness.

Recently, some co-authors of this paper developed an algorithm based on the Si_{EQ} methodology, which took into consideration the effect of minor, major elements, hydrogen dissolved in the alloy and melt treatment on the liquidus temperature for the hypoeutectic Al–Si alloy [3]. The Si_{EQ} was calculated using binary equilibrium phase diagrams for Al–Xi systems. In most cases the liquidus temperature drops uniformly from the melting point of pure aluminum to the temperature of the alloy eutectic state. For example, by increasing the amount of silicon in Al–Si alloys, the liquidus temperature decreases gradually from 660.452 °C (pure aluminum), reaching the minimum of 577 °C at the eutectic composition that was chosen as the most probable at 12.3 wt.% silicon [10].

Mathematically, the liquidus line of most binary Al–Xi phase diagrams can be approximated by the second-order polynomial as follows:

$$T_{LIQ}^{Al-Xi} = A - BXi - CXi^2\,(^\circ C) \quad (3)$$

where Xi is the content of the major, minor elements or hydrogen in wt.%, A is the melting point of pure aluminum, 660.452 °C, B and C are the polynomial coefficients.

The analysis of two liquidus lines of the binary systems, Al–Si and Al–Xi, shows that the "equivalent effect" on the liquidus temperature of the aluminum alloy can be obtained by using "equivalent" concentrations of Si and Xi alloying or impurity elements. This means that the influence of any element in the Al–Si melt on its liquidus temperature can be expressed as the effect of an "equivalent amount" of silicon (wt.%). The isothermal concentration difference between the Si and Xi elements can be mathematically expressed as follows:

$$Si_{EQ,\,@T=\text{constant}}^{Xi} = Si\,(\text{wt.\%}) - Xi\,(\text{wt.\%}) \quad (4)$$

where T is the melt temperature (°C).

Silicon as a major element for the 3XX series of alloys, was chosen as the reference element. Silicon is also known to have the most significant influence on the casting properties of 3XX family of alloys (e.g. fluidity, latent heat and shrinkage).

Taking into consideration the whole temperature range between the melting temperature of pure aluminum and the eutectic temperature of binary alloy the following relationship can be established between Si_{EQ}^{Xi} and the concentration of the element Xi:

$$Si_{EQ}^{Xi} = a_0^{Xi} + b_0^{Xi}Xi + c_0^{Xi}Xi^2 \quad (5)$$

where $a_0^{Xi} + b_0^{Xi} + c_0^{Xi}$ = polynomial coefficients, Xi is the concentration of elements in wt.%.

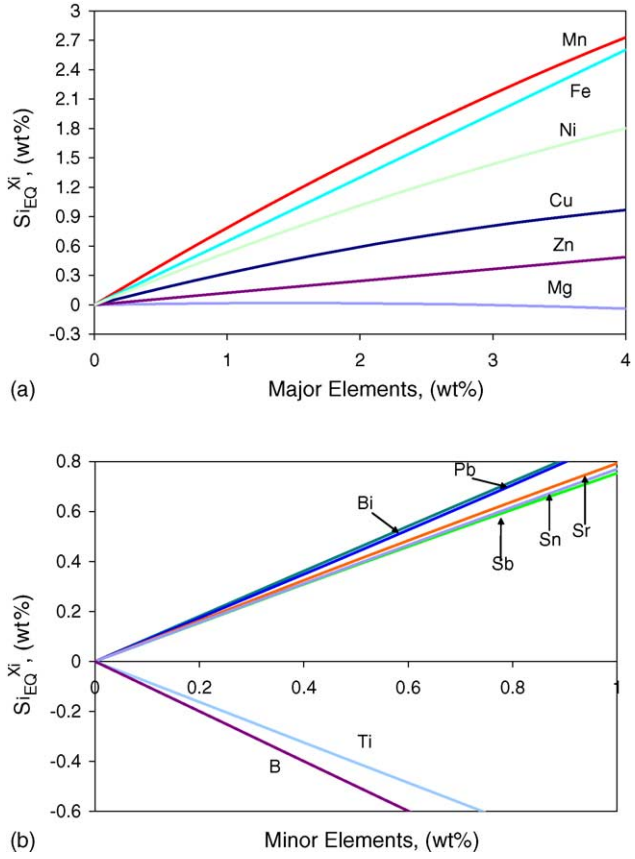

Fig. 1. The effect of Xi elements on the Si_{EQ}^{Xi} for hypo and hypereutectic Al–Si alloys.

Fig. 1a, b and Table 1 summarize the effect of some major, minor and other elements, including grain refiners, silicon modifiers, etc. on the Si_{EQ}^{Xi}. Their effect has been expressed in the polynomial form, see Eq. (5).

Manganese (Mn) has the strongest effect on the Si_{EQ}^{Xi} (Fig. 1a). The effect of the other major elements decreases in the following order Mn, Fe, Ni, Cu, Zn and Mg, respectively. It is important to mention that until now Fe has not been considered as an alloying element, however, current research conducted by the IRC demonstrates the opportunities for the control of cast component integrity by optimizing Fe content. The above-mentioned order of the major elements corresponds to the slope value ($\Delta T/\Delta Xi$) of the liquidus line for a particular binary Al–Xi phase diagram. The effect of some minor elements on the value of the Si_{EQ}^{Xi} has been presented on Fig. 1b.

Table 1
Polynomial coefficients of the Si_{EQ}^{Xi} for various binary Al–Xi alloys representing the most common major and minor elements of the 3XX hypo and hypereutectic aluminum alloys

Al–Xi alloy	b_o	c_o
Al–Cu	0.350	−0.027
Al–Mg	0.0258	−0.0088
Al–Mn	0.8221	−0.0349
Al–Fe	0.6495	0.0003
Al–Zn	0.1227	−0.0002
Al–Sn	0.7849	−0.0313
Al–Bi	0.9076	−0.0092
Al–Pb	0.859	0.02976
Al–Ca	0.0594	0.00685
Al–Sb	0.8255	−0.0327
Al–Ni	0.5644	−0.0285
Al–Sr	0.7854	−0.0157
Al–Ti	−0.8159	0.009927
Al–B	−0.9977	0.00007506

Note: $a_o = 0$ for the elements presented in this table.

The Si_{EQ} for minor, major elements as well as some elements including hydrogen can be determined as the sum of individual contributors ($\sum Si_{EQ}^{Xi}$) plus the effect of silicon itself, and can be expressed as follows:

$$Si_{EQ} = Si + \sum Si_{EQ}^{Xi} (wt.\%) \quad (6)$$

It is interesting to note that elements such as grain refiners (Ti and B), silicon modifiers (Sr and Sb) or low melting point (Bi and Pb) have similar effect on the Si_{EQ}^{Xi} value, thus in liquidus temperature (Fig. 2). The slope for some major and minor elements is expressed as a value of the change in liquidus temperature (ΔT in °C) caused by the addition of 1 wt.% of the given element Xi (Xi: Mn, Fe, Cu, Ni, Zn, etc.) into the Al–Si melt (Fig. 2). It is clear that the biggest effect on the $Si_{EQ}^{Xi}$was the low melting points elements, the grain refiners and silicon modifiers, but because of the addition of these elements in an aluminum melt is on the level of parts per million their effect on the liquidus temperature is negligible. However, their combined effect on other characteristic solidification temperatures could be significant. Therefore, the contributions of these elements to the value of the Si_{EQ}^{Xi} needs to be taken into consideration. Due to the relatively high content of major elements including Cu, Mn, Mg and Zn in the aluminum melt they are the main contributors to the depression of the liquidus temperature.

Based on initial studies a second degree polynomial Eq. (7) correlates the chemical composition and the liquidus

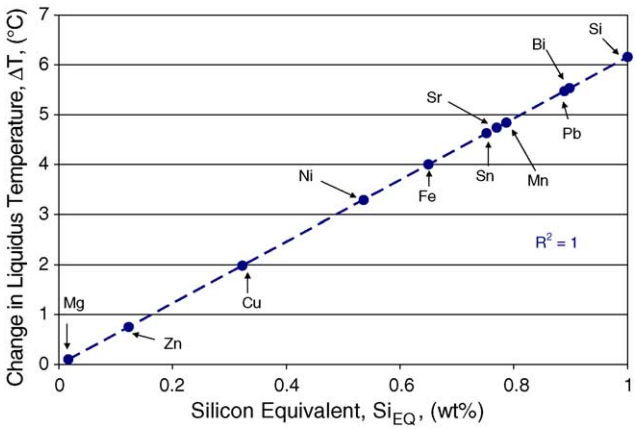

Fig. 2. The Si_{EQ}^{Xi} and ΔT values for 1 wt.% addition of major and minor elements in the 3XX series of hypo and hypereutectic aluminum alloys.

temperature curve of the Al–Si binary hypoeutectic systems [11].

$$T_{\mathrm{LIQ}}^{\mathrm{Al-Si}\sum \mathrm{Xi}} = 660.452 - 6.110 \sum \mathrm{Si}_{\mathrm{EQ}}^{\mathrm{Xi}} - 0.057 \sum \mathrm{Si}_{\mathrm{EQ}}^{\mathrm{Xi}} 2\,(^{\circ}\mathrm{C}) \quad (7)$$

A review of the available literature revealed that there is no equation that can be used to determine the liquidus temperature of hypereutectic Al–Si aluminum alloys. Therefore, the authors developed a new analytical expression to calculate the liquidus temperature of the aluminum 3XX series of alloys that has more than 12.3 wt.% silicon. Applying the regression analysis method, the equation of the liquidus line for the hypereutectic Al–Si binary phase diagram (for a Si wt.% between 12.3 and 35) can be mathematically expressed in the second-order polynomial form as follows:

$$T_{\mathrm{LIQ}}^{\mathrm{Al-Si}} = 389.79 + 15.855\mathrm{Si} - 0.0561\,\mathrm{Si}^2\,(^{\circ}\mathrm{C}) \quad (8)$$

The liquidus temperatures for multi-component hypereutectic 3XX aluminum alloys can be obtained, using Eq. (9):

$$T_{\mathrm{LIQ}}^{\mathrm{Al-Si-}\sum \mathrm{Xi}} = 389.79 + 15.855\,\mathrm{Si} - 0.0561\,\mathrm{Si}^2 + [3.14 \sum \mathrm{Si}_{\mathrm{EQ}}^{\mathrm{Xi}} + 0.057 \sum \mathrm{Si}_{\mathrm{EQ}}^{\mathrm{Xi}} 2]\,(^{\circ}\mathrm{C}) \quad (9)$$

3. Experimental procedure

3.1. Materials

Fourteen hypo and hypereutectic 3XX Al–Si-based alloys were prepared and analyzed at the University of Windsor's Metallurgical laboratory. Eight of them were hypoeutectic Al–Si alloys, with a nominal chemistry of Al-5, 7, 9 and 11 wt.% Si with added 1 and 4 wt.% Cu. Additionally, six alloys were prepared using hypereutectic Al–Si alloys with nominal chemistry of Al-14, 17, 25 and 29 wt.% Si with added 1, 2, 4 and 6 wt.% Cu, respectively. The liquidus temperature of alloys 15 and 16 shown in Table 2 were found in literature [1]. Tables 2 and 3 show the chemical compositions of the alloys analyzed using optical emission spectroscopy (OES) as well as the liquidus temperatures.

Table 2
Chemical compositions (wt.%), of the hypoeutectic aluminum alloys and their T_{LIQ} (°C) temperatures measured with AlTAP (only major elements are given)

Alloy	Si	Cu	Fe	Mg	Mn	T_{LIQ} measured (°C)
1	4.85	1.03	0.09	0.14	0.01	626.6
2	4.89	3.85	0.09	0.16	0.01	618.3
3	7.00	0.96	0.29	0.21	0.01	610.8
4	7.18	4.68	0.17	0.26	0.01	602.6
5	9.12	1.18	0.18	0.28	0.01	597.9
6	9.85	4.38	0.14	0.27	0.01	585.2
7	10.84	0.94	0.11	0.19	0.01	579.4
8	10.55	4.36	0.13	0.17	0.01	567.0

Note: for all alloys $0.005 \leq \mathrm{Mn} \leq 0.01$ wt.%.

3.2. Melting procedure

The alloys were melted in an electric resistance furnace. During melting, the melt was covered with a protective nitrogen gas atmosphere to prevent hydrogen and oxygen contamination. No grain refining or silicon modification agents were added to the melt. All alloys were melted at their respective superheat temperature that is 150 °C above the liquidus temperature. The molten alloys were degassed with the aim to reduce the hydrogen level below 0.100 ± 0.005 mL H_2/100 g of aluminum. This eliminates the hydrogen as a potential independent variable affecting the liquidus temperature.

3.3. Thermal analysis (TA) procedure

The aluminum thermal analysis platform (AlTAP [12]) consists of a high speed-high resolution National Instruments Data Acquisition system that was calibrated (traceable) according to the standard of the National Institute of Standards and Technology (NIST) [7]. The data acquisition system (DAQ) is linked to a personal computer where the AlTAP software is used to record the cooling curves, calculate the first derivative and makes an automated determination of the liquidus temperature [12]. Samples with masses of approximately $500\,\mathrm{g} \pm 10\,\mathrm{g}$ were poured into stainless steel cups. A K-type thermocouple was inserted into the liquid alloy and automatically the cooling curve started to be recorded by AlTAP and until the alloy reached a temperature of 400 °C.

Table 3
Chemical compositions (wt.%) of the hypereutectic aluminum alloys and their calculated and measured liquidus temperatures

Alloy	Si	Cu	Fe	Mg	Mn	T_{LIQ} calculated (°C)	T_{LIQ} measured (°C)
9	13.14	4.11	0.51	0.99	0.21	600.2	601.5
10	25.00	1.18	0.39	0.05	0.12	752.5	751.8
11	25.00	5.64	0.64	0.09	0.15	756.9	760.0
12	24.16	2.46	0.93	0.17	0.36	738.4	735.3
13	28.64	2.43	0.90	0.15	0.36	794.9	798.0
14	13.80	4.00	0.61	1.30	0.23	599.7	599.9
15	17.2	4.85	0.20	0.57	0.02	654.8	654.0
16	17.5	4.80	0.70	0.56	0.27	658.4	656.2

Alloys 9–14 were made at the University of Windsor [8], the data for alloys 15 and 16 were are taken from the literature [1].

Tables 2 and 3 show the measured liquidus temperatures of the experimental aluminum alloys.

4. Results and discussion

The currently available Si_{EQ} methodology for rapid calculation of the liquidus temperature of 3XX Al–Si hypoeutectic alloys was further extended in the present research to developed a novel algorithm for Al–Si hypereutectic alloys. At the present time, algorithms with this level of applicability to both hypo and hypereutectic silicon alloys has not been reported yet (see Eqs. (7) and (9)).

Fig. 3 depicts the calculated liquidus temperatures versus the measured liquidus temperatures for the hypoeutectic compositions used in this paper. In addition, the liquidus temperatures calculated by Drossel [6] and Vijayaraghavan [9] have been compared with the experimentally determined liquidus temperatures using AlTAP. The main purpose for this was to consider the eight alloys shown in Table 2 and to statistically compare the previously developed silicon equivalency concept, and to show the high level of reliability ($R^2 = 0.99$) of Eq. (7) over wide ranges of chemical composition for the hypoeutectic 3XX aluminum alloys. Fig. 3 shows the results of comparing Eqs. (1), (2) and (7) with the measured liquidus temperature using AlTAP for the Al–Si hypoeutectic alloys. A statistical analysis conducted for the above-mentioned evaluation shows the following standard deviations for Eqs. (1), (2) and (5): 5.7, 4.7 and 2.3 °C, respectively, and the maximum differences for the 95% confidence limit were 13.3, 13.2 and 1.2 °C. This demonstrate that the liquidus temperatures determined using Eqs. (1) and (2) are more scattered than if calculated using Eq. (7) when compared with measured values (see Fig. 4). Using the previously discussed statistical data can be concluded the considerable higher level of accuracy for Eq. (7).

Fig. 5 depicts the liquidus lines of the Al–Si phase diagram (solid lines) [11] and the calculated liquidus lines (dashed lines) using Eqs. (7) and (9) for hypo and hypereutectic alloys, respectively. The liquidus temperatures calculated using the Si_{EQ} methodology for the chemistries presented in Tables 2 and 3 were used in Fig. 5 to construct an Al–Si quasi-binary phase diagram. The liquidus temperatures for the alloys, used in this paper were measured by the AlTAP and were also plotted in Fig. 5 as dots [8]. The accuracy of the calculated liquidus temperatures depends strongly on the

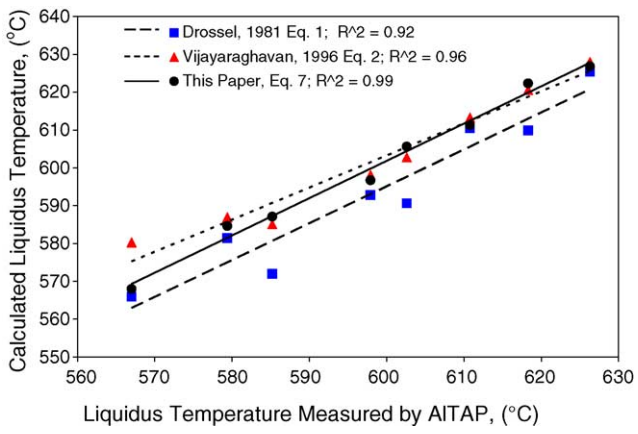

Fig. 3. Calculated vs. measured liquidus temperatures for 3XX hypoeutectic Al–Si alloys.

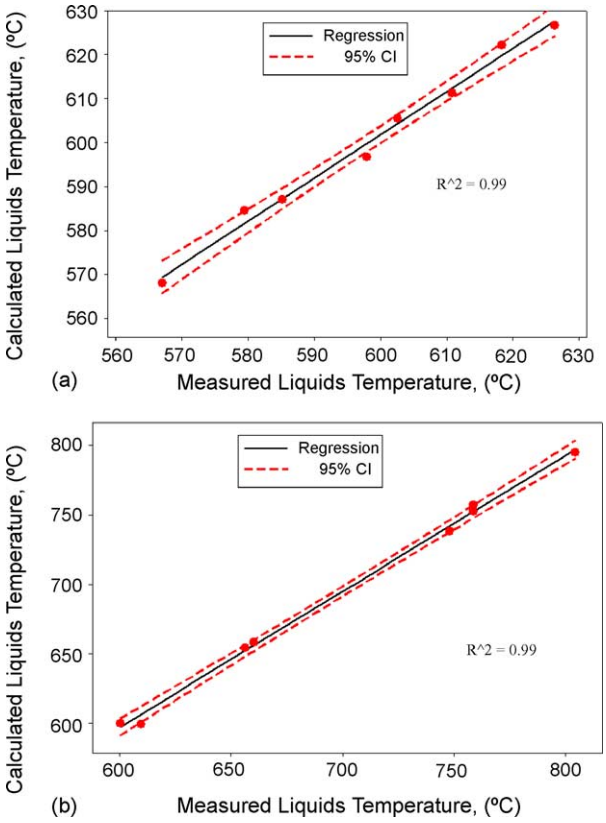

Fig. 4. Coefficient interval (CI) curves showing the 95% confidence limit for the calculated liquidus temperature for Al–Si: (a) hypoeutectic and (b) hypereutectic 3XX alloys using Eqs. (7) and (9), respectively.

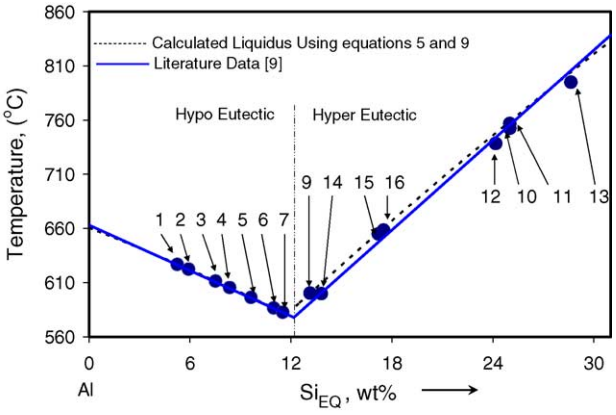

Fig. 5. Phase diagram containing the literature (solid line) [10], calculated (dashed line) and measured (dots) liquidus temperatures superimposed on a "quasi binary Al–Si phase diagram" obtained using the Si_{EQ} methodology for the different 3XX compositions.

accuracy of determination of the coefficients (Fig. 1a, b and Table 1), which allows for the conversion of the alloy composition into "an equivalent silicon" for each component, in wt.%. These coefficients are derived from the respective binary systems, and their accuracy is closely related to the accuracy with which the data for liquidus lines was experimentally determined and numerically fitted. In addition, the accuracy and precision of the OES chemical composition data and the calibration of OES equipment itself can compromise the exactitude of the Si_{EQ} methodology. In order to minimize the error, a re-examination of liquidus lines, on both aluminum and silicon rich side of the Al–Si binary system, or probably for all ternary Al–Si–Xi systems should be conducted.

The negligible differences between the measured and calculated values of the liquidus temperatures for hypo and hypereutectic Al–Si alloys can be observed in Fig. 4. However, these variances can be reduced further by taking into consideration the interactions between elements. The Si_{EQ} mathematical approach of this paper is based on binary alloy systems only and no interactions between three or more elements were taken into consideration for the Si_{EQ} methodology. However, considering an interaction among at least three elements in an alloy (for example, using ternary phase diagrams) might bring the calculated values even closer to the measured ones.

Analyzing Figs. 3, 4 and Table 2 can be observed highly accurate results with an $R^2 = 0.99$ using the Si_{EQ} methodology Eq. (7) in comparison with the available in the literature algorithms to determine the liquidus temperature for multicomponent Al–Si alloys. In addition, the statistical analysis shows that similar precision can be obtained for the determination of liquidus temperature for Al–Si hypereutectic alloys using Eq. (9) (Fig. 4). The high correlation coefficient ($R^2 = 0.99$) between the measured and the calculated liquidus temperature for the hypo and hypereutectic Al–Si alloys confirms that the Si_{EQ} methodology can be used to accurately predict the liquidus temperature for the 3XX series of alloys that solidifies at a cooling rate close to equilibrium conditions.

Additionally, the Si_{EQ} methodology can be used as an off line quality control tool by establishing upper and lower limits for specific chemistries. Especially when the aluminum foundry industry is using recycled materials, the main concern is how to keep the compositions of major and minor elements and impurities within the required boundaries. Today casting plants are using secondary ingots for which the chemical compositions need to be closely monitored, because the presence of some elements in excessive amounts (sometimes even at the parts per million level) can result in serious deficiencies in as- cast components characteristics. Therefore, there is a need to control the quality of the incoming ingots and by using Si_{EQ} this task can be made easier. The accuracy of the results of these algorithms under laboratory conditions indicates that this methodology can be industrially implemented (i.e. as an algorithm built into the OES analysis software) as a tool for daily control of incoming materials in the casting plant.

5. Conclusions

A novel Si_{EQ} methodology has been used to calculate the liquidus temperatures of hypo and hypereutectic multicomponent Al–Si alloys. The calculation procedures presented in this paper are more accurate than those found in the literature. The results of the statistical analysis showed that both algorithms, for hypo and hypereutectic compositions, indicate an approximately 1 °C variance for a 95% confidence limit when the calculated values are compared with the measured ones (Figs. 4 and 5). However, the accuracy to predict the liquidus temperature using the Si_{EQ} methodology depends on the precision in determination of the coefficients presented in Fig. 1 and Table 1.

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