Modelling of materials properties and behaviour critical to casting simulation

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Abstract

Thermo-physical and physical properties of liquid and solid phases during solidification are critical data for casting simulations. However, the number of alloys for which such information is available is limited, primarily due to the difficulty in experimentally determining these properties during the casting/solidification process. Because small variations in composition can have marked effects, it is unwise to extrapolate properties derived from other alloys. The purpose of this paper is to demonstrate the value of integrating the modelling of solidification and the associated thermo-physical and physical properties for multi-component alloys, using Java-based materials properties software (JMatPro). This includes how changes in the composition of an alloy within its specification range can substantially affect its properties during solidification and how properties of the liquid can vary significantly in the mushy zone.
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1. Introduction

Throughout the manufacturing industry, casting process simulation is now widely accepted as an important tool in product design and process development to improve yield and casting quality. Casting simulation requires high-quality information concerning thermo-physical and physical properties during solidification. Some properties have been measured for specific alloys, but the number of alloys for which information is available is limited. Furthermore, the information may be incomplete in the sense that not all properties have been measured and sometimes, disparate information from a variety of sources is used to build up the database for one specific alloy. The latter situation can lead to inconsistent results, as the composition of the alloys used for database creation may not be the same and consequently critical temperatures, such as for the solidus and invariant reactions, may differ between the alloys. To overcome the lack of data and achieve a better understanding of how changes in composition within a specification range of an alloy may affect solidification properties, it is highly desirable to develop computer models for calculation of the thermo-physical and physical properties of multi-component alloys during solidification.

Java-based materials properties software (JMatPro) provides extensive information on how the properties of an alloy may change within its specification range, as well as providing detailed information on the properties of each individual phase [1,2], such as the liquid phase in the mushy zone, which is usually beyond the capability of measurement. Thermodynamic modelling based on the CALPHAD methodology is a well-established technique for modelling of phase equilibria in multi-component alloys [3]. By extending it to non-equilibrium solidification using the so-called Scheil–Gulliver (SG) model, excellent results for the phases forming during solidification as well as their composition and temperature range of formation can be obtained [4–8]. Successful predictions for fraction solid versus temperature, phase formation and latent heat evolution have been incorporated into casting simulation packages, where they have achieved high-quality results.

However, JMatPro has gone one step further by developing an extensive capability for the calculation of physical and thermo-physical properties [1,9], which can be integrated with the thermodynamic calculations to provide wide-ranging thermo-physical and physical properties for use in solidification modelling.

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The properties of individual phases in multi-component systems, such as molar volume and thermal conductivity, are first expressed by functions similar to those used to model thermodynamic excess functions in multi-component alloys [3], taking account as far as possible of the relevant underlying theory. Once the properties of the individual phases have been defined, the property of the final alloy is calculated using well-established mixture models that can account for the effect of microstructure on the final property [10,11]. Such models, which were initially developed for two-phase systems, have been extended to allow calculations to be made for multi-phase structures [12]. Properties that are critical to casting simulation, such as volume, density, thermal expansion coefficient, thermal conductivity and liquid viscosity, can be modelled. Extensive databases of relevant parameters now exist for most of the major phases in Al-, Fe- and Ni-based alloys, which have been validated against experimental measurement for both the liquid and solid state [13–15]. Readers interested in Mg-, Ti- and Zr-based alloys or properties, such as Young's, bulk and shear moduli and Possion's ratio are referred to Refs. [2,16].

2. Technical background

Accurately modelling the continuous change in physical properties on cooling depends on first reproducing the fractions of all participating phases present at each temperature. Historically, this started with the assumption of equilibrium solidification, linearized phase boundaries, the use of the lever rule and assuming that complex alloys would exhibit pseudo binary behaviour. As is well known, this is rarely realized in practice, a major improvement was made by Scheil, who assumed that solute diffusion in the solid phase is small enough to be considered negligible. However, the traditional derivation of the Scheil equation has severe restrictions when applied to multi-component alloys [17]. It is not possible to derive this equation using the same mathematical method if the partition coefficient is dependent on temperature and/or composition. Furthermore, the Scheil equation is applicable only to single-phase solidification. It, therefore, cannot be used for eutectic alloys that are common in Al- and Mg-alloys or cast irons and it cannot be used to predict the formation of intermetallics during solidification.

Using more extensive thermodynamic modelling, all of the above disadvantages can be overcome. Instead of using Fig. 1 as a representation of the actual phase changes, this becomes merely a schematic diagram, with the actual amounts and compositions being calculated rigorously for the multi-component system. The appearance of additional phases can be easily taken into account in this approach, keeping the assumption that no back diffusion occurs in them. Therefore, all transformations can be accounted for, including the final eutectic solidification. Following the scheme outlined in Fig. 1, it is necessary to specify the magnitude of each isothermal step, but as this can be set as small as one wishes, it provides results that are almost completely equivalent to those which would be obtained from continuous cooling. A further advantage of using a thermodynamic approach is that the heat evolution during solidification is a straightforward product of the calculation.
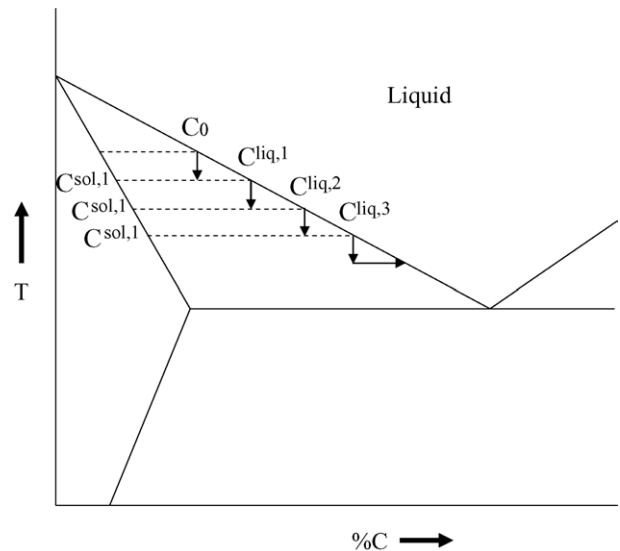

Fig. 1. Schematic representation of solidification occurring under Scheil conditions.

3. Sample calculations

3.1. Solidification of cast irons

A model for fast C and N diffusion has been proposed in a previous paper [13] and applied to cast irons. However, it is noted that because the activity of C remains fairly constant and close to unity over the solidification range, little difference is found in predicted solidification behaviour between a fast C diffusion model or a "standard" Scheil–Gulliver model. Of particular interest is the case of so-called eutectic graphite volume expansion [18–21], which offers a demonstration of how physical property calculations have been linked with the solidification calculation.

Figs. 2 and 3 show examples of the accuracy that can be obtained for calculated properties of the austenite and liquid phases in binary systems Fe–C and Fe–Si [22,23], while the graphite molar volume is taken from Gustafson [24]. Linking
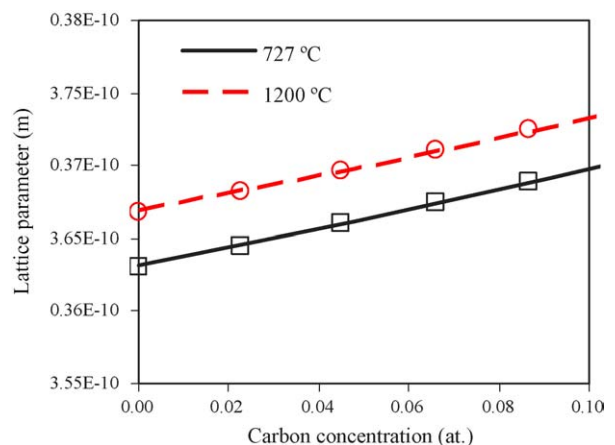

Fig. 2. Lattice parameter of austenitic Fe–C alloys at various temperatures. Markers are experimental values [22].
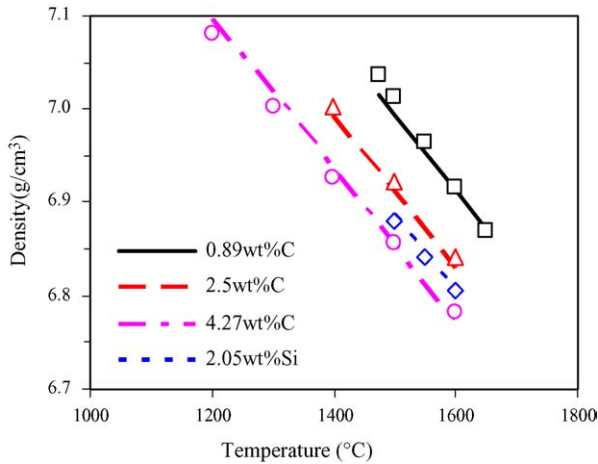
Fig. 3. Density of liquid Fe–C and Fe–Si alloys vs. temperature. Markers are experimental values [23].

the solidification model to the physical property calculations in a commercial multi-component grey iron provides the volume change on solidification as shown in Fig. 4. It can be seen the initial solidification of austenite involves shrinkage. However, at the graphite eutectic temperature, expansion occurs and continues throughout the eutectic reaction. Clearly, the total volume change will be critically dependent on alloy concentration.

3.2. Solidification of Ni-based superalloys

A result of the current approach is that fine detail can be obtained, for example, for the density of the liquid in the mushy zone (Fig. 5). This can be utilized for the calculation of casting defects and potentially, macrosegregation in ingot casting and re-melting processes. In this case, we have shown the varying behaviour of liquid phase in the mushy zone for three superalloys, showing the quite different behaviour of alloy 706, which shows signs of a density inversion, in comparison to 718 and 625. Such behaviour is consistent with observations of defects in these alloys and various calculations of liquid density of Auburtin et al. [25].
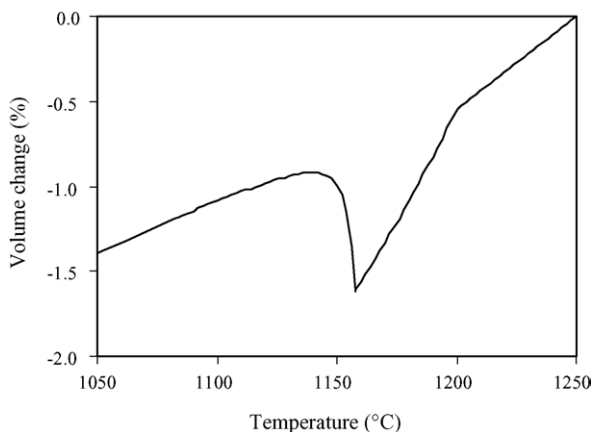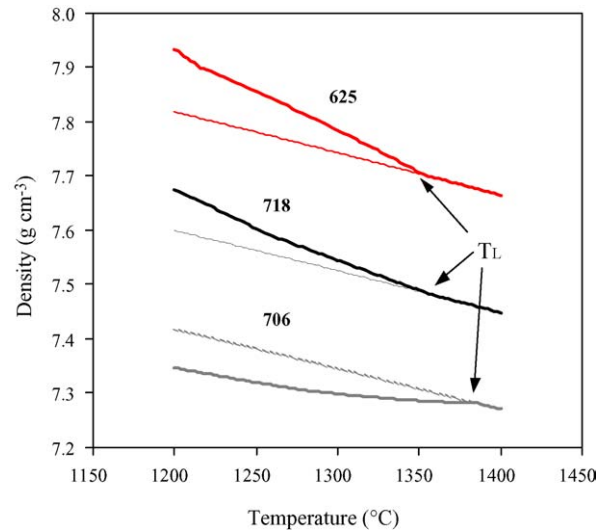

Fig. 5. Calculated densities of the liquid phase during solidification of 625, 718 and 706 alloys (bold lines). Fine lines show the density of the liquid alloys extrapolated from high temperature (T_L is the liquidus).

In terms of calculation of the Rayleigh number and its application to defect formation, it is also important to obtain liquid viscosities in the mushy zone. Fig. 6 shows the calculated liquid viscosity in two alloys, 713 and 718, during solidification. Experimental measurements of the fully liquid alloys are shown for comparison [26]. It can be seen that a simple extrapolation of the high temperature liquid properties into the mushy zone will seriously underestimate the viscosity for both alloys.

In all cases of solidification, the properties of the liquid in the mushy zone are sensitive to partitioning of the various elements of the alloy. By combining the thermodynamic calculation with the physical property calculation, it is now possible to explicitly consider the effect of changing liquid composition on a wide variety of physical properties.

Fig. 4. Volume change of a grey cast iron of composition Fe–2Si–3.4C–0.75Mn during solidification.
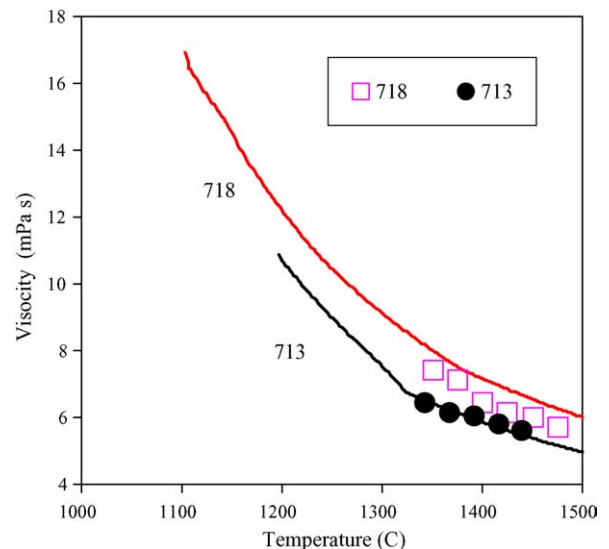

Fig. 6. Calculated viscosity of the liquid during solidification of alloys 713 and 718. Experimental data for the fully liquid state [24] shown for comparison.

Table 1
Composition of ADC12 alloy used in calculation

Composition (wt.%)	Cu	Fe	Mg	Mn	Ni	Si	Zn	Al
Specification	1.5–3.5	≤1.3	≤0.3	≤0.5	≤0.5	9.6–12.0	≤1.0	Balance
Low specification	1.5	0.25	0.1	0.1	0.1	9.6	0.1	88.25
High specification	3.5	1.3	0.3	0.5	0.5	12.0	1.0	80.90

3.3. Variations in behaviour within an alloy specification range

The ability to calculate thermo-physical and physical properties at will for many types of alloys is important for a number of reasons. Firstly, information can be readily gained for alloys, where measurements are not available. Because there is a distinct lack of reliable measurements for solidification properties of many alloys, this is in itself a highly valuable capability. It is also possible to calculate (quickly and self-consistently) how such properties change as the alloy composition varies within its permitted specification range. Furthermore, a great deal of detailed information comes with the current calculation route that is otherwise either impossible or extremely difficult, to determine experimentally. For example, properties of the liquid in the mushy zone, which is the controlling factor in many types of defect, e.g. freckles, macrosegregation and shrinkage porosity.

Because of the lack of data on thermo-physical and physical properties for multi-component alloys, properties are often given for a typical alloy in its class, without defining its composition. Certain alloys are rather sensitive to small composition variations, while others, for example, solid solution alloys, may not be so sensitive. It may also be that the composition specification for that alloy is wide, which is the case for many well-known and well-used alloys.

An Al-alloy ADC12 (Japanese designation, for composition, see Table 1) will be taken as an example here. This is an alloy containing high Si and Cu, with quite large variations on the permissible levels of other elements, such as Fe and Ni. Two compositions have been tested, one at the low level of specification for each element, the other at the high level. While such a choice might be expected to show the largest difference in behaviour, this is not necessarily the case. Depending on partition behaviour and effect on invariant reactions, it is possible for an increase in one element to cancel out the effect of another. In the event, the calculated difference in behaviour of the two ADC12 alloys is very striking.

Fig. 7 shows fractions solid versus temperature plots for the two alloys. The high specification (HS) alloy is hypereutectic, with primary Si and intermetallics forming over a significant temperature range, while the low specification (LS) alloy forms about 22% of primary Al. At the start temperature of eutectic solidification for the HS alloy (565 °C), the fraction solid for the LS alloy is ∼65%, in comparison to ∼5% for the HS alloy. The discrepancies between fraction solid at any temperature remain high for much of solidification sequence, though both finally solidify via a eutectic reaction involving Al₂Cu. The very different behaviour of the two alloys, will subsequently affect all
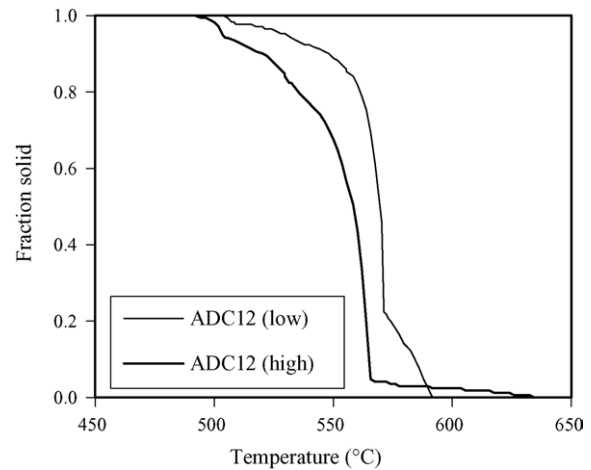

Fig. 7. Calculated fraction solid vs. temperature plots of two ADC12 Al-alloys.

of the properties as a function of temperature. An example is the volume change (in the range 450–650 °C), which again, for most of the temperature range, is quite different for the two alloys (Fig. 8).

Differences between alloys with much smaller composition variations can produce quite substantial variations in the behaviour of the physical properties of the liquid. The Al-alloy 356 is taken as an example in the present work. Fig. 9 shows the density changes of an alloy with the composition Al–0.01Cu–0.2Fe–0.3Mg–0.02Mn–7Si–0.025Zn (wt.%). For this composition, there is a slight density inversion as Mg segregates into the liquid below the silicon eutectic. However, when Cu, Mn and Zn levels increase to higher levels (0.25Cu,
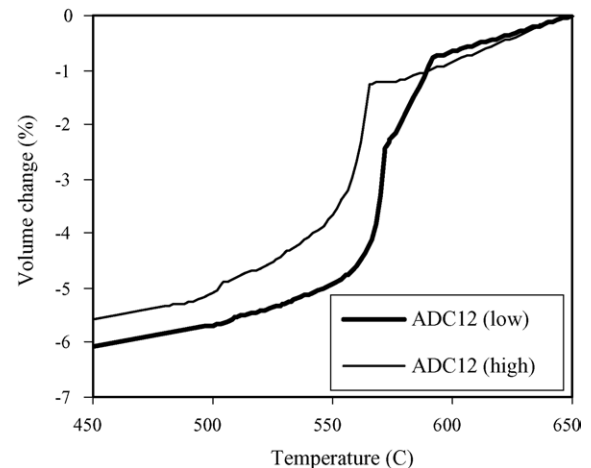

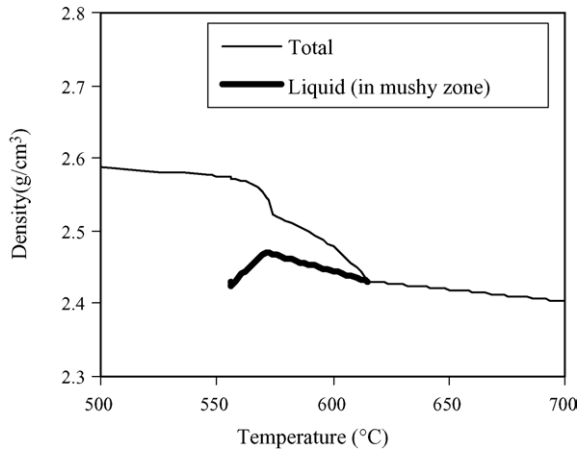Fig. 8. Calculated volume change vs. temperature plots of two ADC12 Al-alloys.

Fig. 9. Calculated density of a 356 Al-alloy (with low Cu, Mn and Zn) during solidification. Bold line shows density of the liquid in the mushy zone.
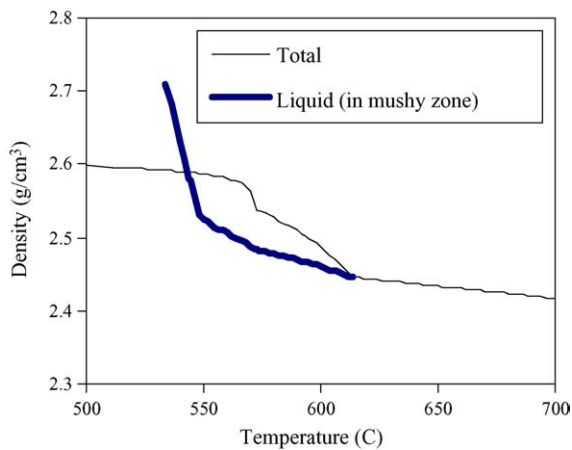

Fig. 10. Calculated density of a 356 Al-alloy (with high Cu, Mn and Zn) during solidification. Bold line shows density of the liquid in the mushy zone.

0.3Mn, 0.35Zn) the behaviour of the liquid in the mushy zone changes dramatically (Fig. 10). During the initial Al solidification, the behaviour is similar; however, the behaviour during the eutectic part of solidification is quite different, with the liquid phase now being much denser.

4. Summary and conclusions

Models have been developed for the calculation of the change in various thermo-physical and physical properties in several different multi-component alloys during solidification. This augments earlier predictions for fraction solid versus temperature, phase formation and latent heat evolution already incorporated into casting simulation packages. The new models have been successfully combined with the calculation of non-equilibrium solidification behaviour using the modified Scheil–Gulliver approach. Properties can now be calculated as a function of temperature for many alloys where there is no current experimental information and incomplete data sets can be augmented in a self-consistent manner. In principle, such data sets can also be used as input for inverse modelling to obtain other properties, which are difficult to measure, such as the heat-transfer coefficients at the ingot/mould interface.

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