Thermodynamic Simulation on Thixoformability of Aluminium Alloys for Semi-solid Metal Processing

M.S. Salleh, M.Z.Omar, J. Syarif, M.N. Mohammed, K.S. Alhawari

Abstract—Semi-solid metal (SSM) processing is a relatively successful method for forming alloys in the semi-solid state to near net shaped products. One type of SSM processing is thixoforming which is used widely in the automotive industry to produce near net-shaped parts. All the alloys that have been used to date for thixoforming were developed originally for either casting or forging purposes. Therefore, it is necessary to modify existing commercial alloys in order to improve the potential of these alloys for use in thixoforming. In the present work, thermodynamic simulations were utilised to determine the working window temperature, solidification temperature range and fraction liquid sensitivity of modified A319 commercial alloy. All the calculations in the present work are performed using JMatPro software. The conditions of the non-equilibrium solidification are determined using the Scheil-Gulliver equation. A key advantage of the JMatPro software is that it allows the calculation of the formation conditions for the various stoichiometric compounds (exp: α-Al(Fe,Mn)Si, β-AlFesi, $Al_5Cu_2Mg_8Si_6$, Al_7Cu_2M and Al_2Cu), which has a great influence on the mechanical properties of aluminium alloys. The results indicate that these modified alloys are potential materials for thixoforming.

Keywords—Alloys design criteria, thermodynamic simulation, thixoforming, aluminium alloys, phase transformation

I. INTRODUCTION

Semi-solid metal (SSM) processing is a relatively new technology that offers several advantages over liquid and solid processing, such as weight savings in components with less porosity than conventional die casting and low forming forces during the shaping process [1]. This process relies on the thixotropic behavior of alloys which have a spheroidal

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rather than a dendritic microstructure in the semi-solid state [2]. One type of SSM processing is thixoforming which involves the forming of alloys in the semi-solid state to near net shaped products. Over the past few years, there has been extensive interest in the development of new alloys specially tailored for SSM processing [3-5]. Some of these alloys show improvements in their properties such as less temperature sensitivity of the liquid fraction, a suitable solidification range and the potential for age hardening.

However, to date, only a few materials were selected for SSM processing such as A319, A356 and A357 to produce commercial products, mainly in the automotive industry. Hence, there is a need to extend the range of aluminium alloys that can be used in this process. A new method has been developed based on thermodynamic simulation that is suitable for predicting the behavior of aluminium alloys during solidification [6]. This method can be used as a first approximation, for example to calculate the solidification temperature, working window temperature and fraction liquid sensitivity, hence avoiding any unnecessary experimental work, which is both time consuming and costly [7].

Recently, a few researchers have carried out some thermodynamic simulations on various aluminium alloy grades such as AA7000, AA2024, A380, A356, A357, AM60 and A206. For instance, Maciel et al. [8] studied the suitability of commercial AA7000 (Al-Zn-Mg-Cu) series alloys for SSM processing; Han and Viswanathan [9] investigated the ideal composition of hypoeutectic aluminium-silicon alloys (A356/A357) to make them more suitable for SSM processing; Liu et al. [3] investigated the thixoformability of alloys based on Al-Si-Cu and Al-Si-Cu-Mg systems; Solek et al.[10] studied the thixoformability of A356 and AA7000 aluminium alloys; and Yuan Dong et al.[6] investigated a commercial AM60 alloy for SSM processing.

Thixoformability is the term usually used to indicate the suitability of alloys for thixoforming. Thixoformability depends on parameters such as solidification range, fraction liquid sensitivity and window processing temperature. Thermodynamic simulation software is very useful for calculating the thixoformability parameters for multicomponent alloys, commercial foundry alloys and potential SSM alloys.

The focus of this paper is to investigate the effect of the addition of magnesium or zinc to A319 alloys and the

suitability of the consequent potential candidate materials for thixoforming. This study is an extension of previous work which only focused on the addition of zinc to A319 alloys [11]. A commercial software package, "Java based Material Properties" (JMatPro) was used for the simulation based on the alloy design criteria for SSM processing. Our results found that, the A319 alloy can be modified thereby, allowing a larger processing window temperature and reduction in fraction liquid sensitivity as well as improved the mechanical properties.

II. ALLOY DESIGN CRITERIA AND THERMODYNAMIC CALCULATION

Existing alloys were originally designed for casting or forging purposes, therefore new alloys that are more suited to thixoforming are desired. In this respect, some modifications can be made in order to fully utilize the potential of aluminium alloys in the SSM processing. Previous research has identified some general criteria that affect the thixoformability of aluminium alloys which are outlined below.

A. Working Window Temperature ($\Delta T^{0.3/0.5}$ °C)

The working window temperature can be defined as the interval of temperature for a fraction liquid from 0.3wt% to 0.5wt% in the fraction liquid versus temperature curve. A large working window temperature represents the stability of the alloy for semi-solid processing.

B. Solidification Temperature Range (ΔT_{S-L})

The solidification temperature range is defined as the temperature range between the solidus and the liquidus of an alloy. It is usually dependent on the alloy composition and processing conditions. The solidification temperature range should not be too wide, e.g. from 100K to 150K because it may leads to poor resistance to hot tearing and poor fluidity of the liquid alloy. On the other hand, solidification range that is too narrow will makes it difficult to control the temperature of the semi-solid billets during thixoforming.

C. Temperature Sensitivity of Liquid Fraction (df_L/dT)

The temperature sensitivity is usually defined by the slope of the fraction liquid versus temperature curve. Alloys for thixoforming should have a small temperature sensitivity of liquid fraction. It also believed that, for good processability (df_L/dT) should be less than $0.020K^{-1}$ at the SSM processing temperature, which means less than 2% change in the fraction liquid is allowed around the processing temperature. In addition, according to Liu et al. [3], the binary eutectic reaction on the fraction liquid versus temperature curve should occur at a liquid fraction of 30-50%, as shown in Fig.1. This is the area where the α-solid solution starts melting. If this occurs at a liquid fraction above 50%, the liquid formation is uncontrollable and will affect the thixoforming process.
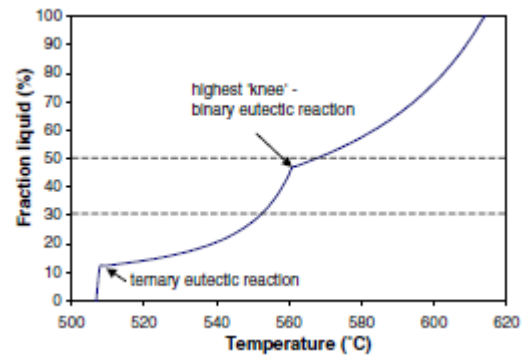

Fig. 1 Schematic of a typical fraction liquid versus temperature curve [3].

A thermodynamic calculation allows the solidification parameters to be investigated without the need to prepare any actual material. This is very useful when designing new alloys, especially potential candidates for SSM processing. In this work, the analysis of the solidification process of A319 alloys was carried out using JMatPro software. This software has the capability to calculate the working window temperature, solidification temperature and temperature sensitivity of liquid fraction, as discussed above. The calculation is based on Scheil-Gulliver's equation for multicomponent systems. In addition, this software also allows the prediction of the formation conditions for various compounds which influence the mechanical properties of alloys during solidification.

In equilibrium solidification, the composition of the solid c_s as a function of the fraction solid f_s is given as [10]:

$$c_s = \frac{kc_0}{f_s(k-1)+1} \tag{1}$$

where k is the distribution co-efficient and c_0 is the composition of overall liquid alloy.

$$k = \frac{c_s}{c_l} \tag{2}$$

By substituting equation (2) to (1), the equation can be written as

$$f_s = \left(\frac{1}{1-k}\right)\left(\frac{T_l - T}{T_s - T}\right) \tag{3}$$

where T_l and T_s are the equilibrium liquidus and solidus temperatures, respectively. Assuming that the solute diffusion in the solid phase is very low, the equation (1) can be written as

$$c_s = kc_0(1-f_s)^{k-1} \tag{4}$$

and equation (3) as

$$f_s = 1 - (\frac{T_s - T}{T_s - T_l})^{\left(\frac{1}{1-k}\right)} \tag{5}$$

To determine the liquid fraction f_L in SSM alloys, the Scheil equation in (5) can be written as

$$f_L = \left(\frac{T_m - T}{T_m - T_l} \right)^{\left(\frac{1}{1-k} \right)} \qquad (6)$$

where T_m is the melting temperature of the SSM alloys.

It should be noted that the Lever rule and the Scheil's model describe two different conditions namely equilibrium and non-equilibrium solidification processes, respectively. The liquid fraction can be estimated using the Lever rule, which assumes that complete equilibrium, is maintained between the solid and liquid phases, and Scheil's model, which assumes that there is a complete mixing in the liquid and no diffusion in the solid phase. An extremely slow rate of cooling is required if this condition is to be achieved. In practice, most of aluminium alloys experience non-equilibrium solidification. Therefore, we need to use Scheil's model equation to analyze the solidification of A319 and modified A319 alloys tailored for SSM processing.

III. THERMODYNAMIC SIMULATIONS OF A319 CAST ALUMINIUM AND A319 MODIFIED ALLOYS

A. Working Window Temperature ($\Delta T^{0.3/0.5}$ °C)

The working window temperature is also referred to as the operational temperature window. Since the temperature fluctuates during the forming of the specified alloys, a relatively large working window temperature is needed. The prediction of the fraction liquid and a suitable processing temperature is useful in the selection of alloying elements that need to be are tailored for thixoforming. For this analysis, all the modified A319 alloys are assumed to have the same nominal chemical composition as the A319 alloy given in Table 1, with the exception of magnesium and zinc, which are varied over specified ranges, as given in Tables 2 and 3.

The thixoformability of alloy A319 can be improved by increasing the amount of magnesium or zinc content. The working window temperature at between 0.3wt% and 0.5wt% fraction liquid is enlarged from 18°C to 26°C by magnesium addition and from 18°C to 24°C by zinc addition, as shown in Figs. 2 and 3.

Fig. 3 shows that the addition of 2.0wt% and 2.5wt% of zinc does not appear to increase the working window temperature. The addition of either magnesium or zinc also reduces the processing temperature of modified A319. Figs. 4 and 5 show the effect of the addition of either magnesium or zinc addition at 0.4wt% fraction liquid. The processing temperature is reduced from 562°C to 547°C and 562°C to 551°C when magnesium and zinc is added, respectively, to A319.

Table.1 Chemical composition of A319

Starting alloy	Si	Cu	Mg	Zn	Fe	Mn	Al
A319	5.5	3.0	0.10	1.0	1.0	0.5	Bal

Table. 2 Chemical composition of A319 with magnesium addition

Alloys	Si	Cu	Mg	Zn	Fe	Mn	Al
A	5.5	3.0	1.0	1.0	1.0	0.5	Bal
B	5.5	3.0	1.5	1.0	1.0	0.5	Bal
C	5.5	3.0	2.0	1.0	1.0	0.5	Bal
D	5.5	3.0	2.5	1.0	1.0	0.5	Bal

Table 3. Chemical composition of A319 with zinc addition

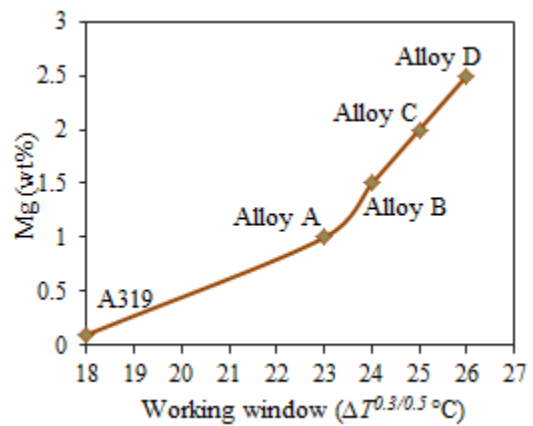| Alloys	Si	Cu	Mg	Zn	Fe	Mn	Al
E	5.5	3.0	1.0	1.5	1.0	0.5	Bal
F	5.5	3.0	1.0	2.0	1.0	0.5	Bal
G	5.5	3.0	1.0	2.5	1.0	0.5	Bal

Fig. 2 Working window temperature as a function of magnesium concentration in A319 and A to D alloys.
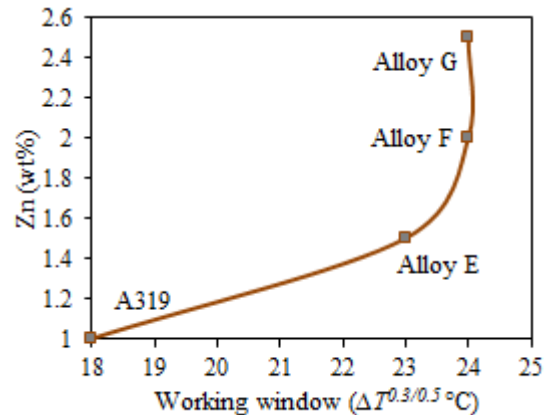

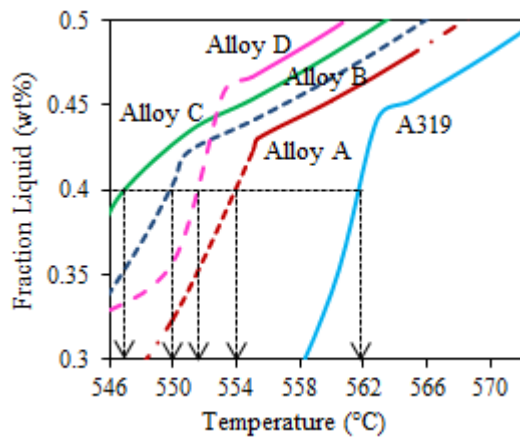Fig. 3 Working window temperature as a function of zinc concentration in A319 and E to G alloys.

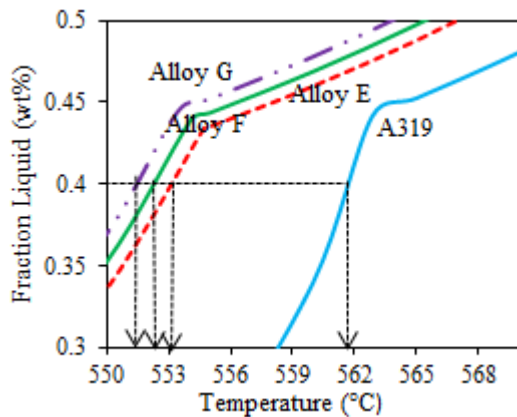Fig.4 Relationship between fraction liquid of A319 alloy and A to D alloys (magnesium addition).

Fig.5 Relationship between fraction liquid of A319 alloy and E to F alloys (zinc addition).

B. Solidification Temperature Range (ΔT_{S-L})

The solidification temperature ranges of A319 and modified A319 are shown in Fig. 6. For A319, the solidification temperature is 149.9K whereas for alloys A, B, C and D, the corresponding temperature is 129.8K, 131.3K, 132.6K and 133.8K, respectively. These temperatures show good agreement with the alloys design criterion which specifies that the solidification temperatures should be less than 150K. All alloys with a magnesium addition shows improvement, i.e the solidification temperature range is reduced, thereby avoiding the risk of hot tearing [12]. For alloys E, F, and G, the solidification temperature range is 149.8K, 154.6K and 165.3K, respectively. It seems that, alloys F and G tend to expose to hot tearing.
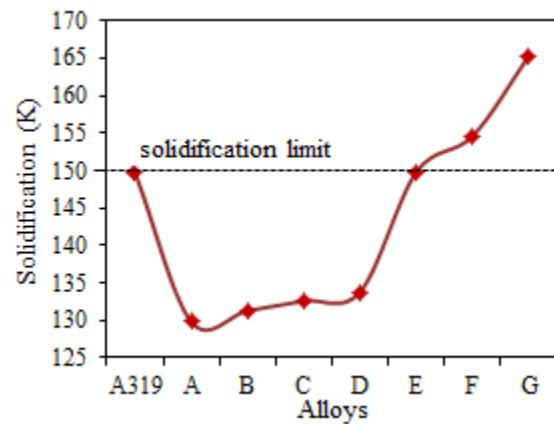

Fig. 6 Solidification temperature ranges of studied alloys.

C. Temperature Sensitivity of Liquid Fraction ($\mathrm{d}f_L/\mathrm{d}T$)

It is difficult to fully control the processing temperature during SSM processing. The temperature sensitivity of liquid fraction is the most important parameter in achieving a well-controlled temperature during semi-solid processing. Fraction liquid sensitivity should be as small as possible. Large values will result in substantial variation of the fraction liquid with a small change in temperature, and will consequently affect the mechanical properties of the final product. Hence, the smaller the temperature sensitivity of the liquid fraction, the easier it is to control the processing temperature. The variation in the liquid sensitivity with processing temperature and fraction liquid at 0.4wt% is shown in Fig. 7. The temperature sensitivity of fraction liquid of alloys A to F decreases at 0.4wt% fraction liquid. Alloys A to D shows a reduction in temperature sensitivity from 0.018 K to 0.008 K while alloys E to G shows a reduction in sensitivity from 0.018 K to 0.003 K.

The processing temperature of alloys A319 falls dramatically (562°C to 548°C) when magnesium is added from 0.1wt% to 2.0wt%, while for alloy D, with 2.5wt% magnesium, it increases from 548°C to 552°C. For alloys E to G with zinc addition, there is a reduction in the processing temperature from 562°C to 551°C. The binary eutectic reaction for all alloys group occurred between 0.3wt% to 0.5wt% fraction liquid, as shown in Figs.8 and 9.
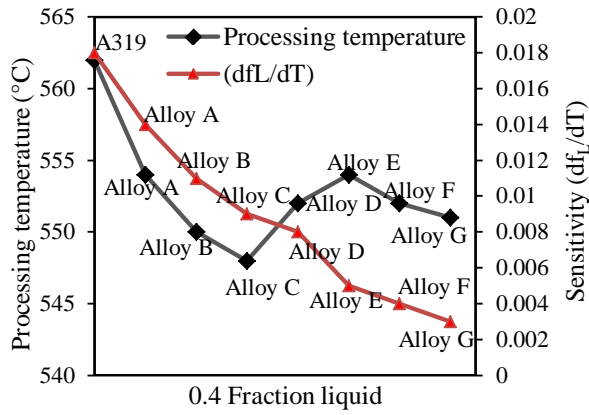
Fig. 7 Relationship between fraction liquid 0.4wt%; sensitivity and processing temperature of all alloys.
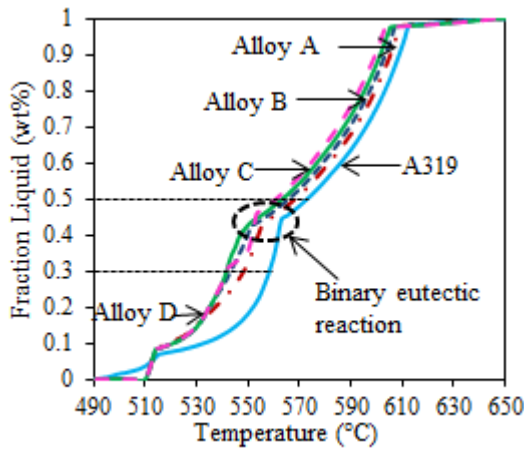

Fig. 8 Relationship between fraction liquid and temperature of A319 modified with magnesium addition.
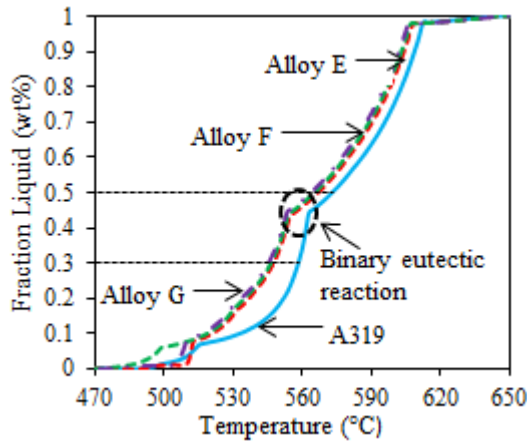

Fig. 9 Relationship between fraction liquid and temperature of A319 modified with zinc addition.

D. Eutectic temperature

Eutectic temperature also plays an important role in determining the processing temperature of aluminium alloys. The processing temperature of the alloy should be higher than the eutectics temperature in order to avoid large fraction liquid variation with processing temperature. Figs. 10 and 11 show the relationship between the addition of magnesium and zinc to A319 aluminium alloys, respectively. As shown in Fig. 10, the eutectic temperature for modified alloys A to C at 0.4wt% fraction liquid decrease from 556°C to 544°C when the addition of magnesium ranges from 0.1wt% to 2wt%. The eutectic temperature increases to 550°C when the addition of magnesium is 2.5wt%. As shown in Fig. 11, for alloys with zinc addition, the eutectic temperature decrease from 556°C to 548°C when the addition of zinc is 2.5wt %. These results suggest that both magnesium and zinc can reduce eutectic temperature during SSM processing.

It should be noted that, the fraction liquid is set at 0.4wt %, so that the processing temperature, according to the fraction liquid, is usually above the eutectic temperature for all modified alloys. It is very important to ensure that the processing temperature is above the eutectic temperature in order to obtain a homogenous microstructure [13]. The zinc concentration does not have a big effect on the eutectic temperature. On the other hand, varying the magnesium concentration provides an opportunity to vary the eutectics temperature and is useful for SSM processing.
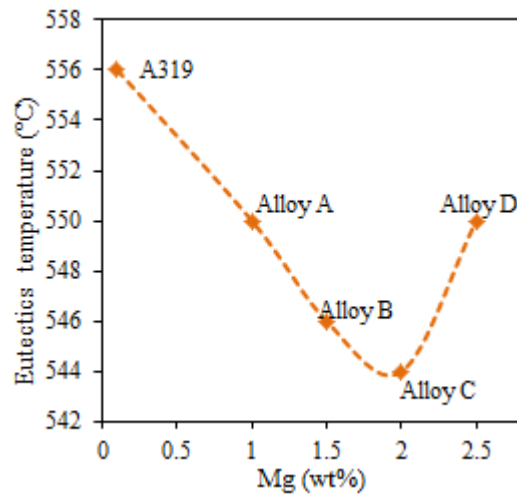

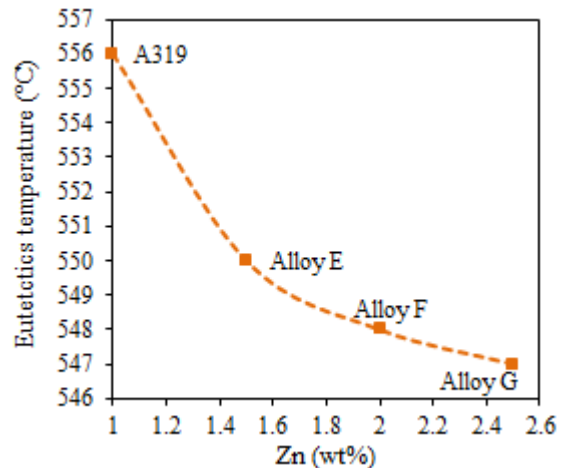Fig. 10 Relationship between magnesium addition and eutectic temperature of modified alloys.

Fig. 11 Relationship between zinc addition and the eutectic temperature of modified alloys.

IV. PHASE TRANSFORMATION

The presence of various compounds in modified A319 alloys plays an important role in influencing the alloy's mechanical properties. These compounds are formed by incorporating transition elements, especially iron, zinc and magnesium, into an Al-Si alloy in order to increase its strength and improve wear resistance [14]. Main elements such as Al, Si, Cu, Zn and Mg are taken into account in this analysis, whereas other elements are not considered since their effect to the mechanical properties is very minimal. The equilibrium diagrams in Figs. 12 to 19 demonstrate the various compounds that affect the mechanical properties of A319 and modified A319. Primary stoichiometric compounds such as α-Al(Fe,Mn)Si, β-Alfesi, $Al_5Cu_2Mg_8Si_6$, Al_7Cu_2M and Al_2Cu are formed in A319 alloy. When the magnesium content in A319 is increased from 0.1wt% to 1.5wt%, a new phase of $Al_8FeMg_3Si_6$ is formed and when magnesium content is increased from 1.5wt% to 2.5wt%, a new phase of Mg_2Si is formed. For alloys E to G, when the zinc content is increased from 1.0wt% to 1.5wt%, a new phase of $Al_8FeMg_3Si_6$ is formed and when zinc content is increased to 2.0wt %, a new Mg_2Si and Zn_HCP are formed.

Table 3 shows the compounds of A319 and modified A319 in alloys A to D. The volume fraction of the $Al_8FeMg_3Si_6$ phase becomes greater with a magnesium concentration from 1wt% to 2.5wt%. The formation of certain intermetallic compounds such as Mg_2Si in alloys C, D and F will increase the strength and heat treatability [15]; hence improving the mechanical properties of the modified alloys.

Fig. 14 Equilibrium diagram of alloy B obtained from JMatPro simulation

Fig. 15 Equilibrium diagram of alloy C obtained from JMatPro simulation

Fig. 16 Equilibrium diagram of alloy D obtained from JMatPro simulation
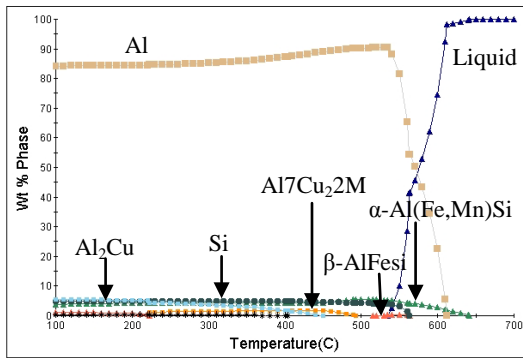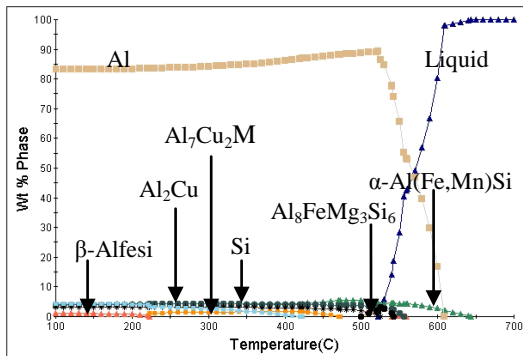

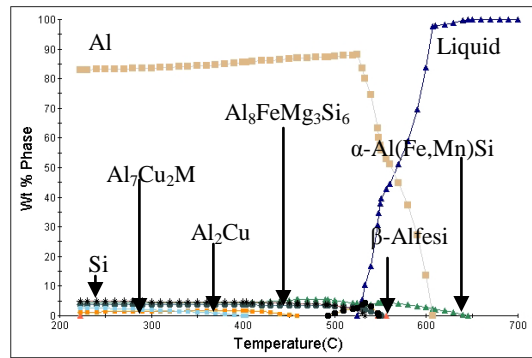Fig. 12 Equilibrium diagram of A319 obtained from JMatPro simulation

Fig. 13 Equilibrium diagram of alloy A obtained from JMatPro simulation

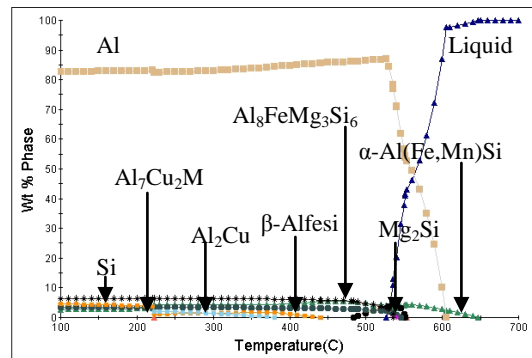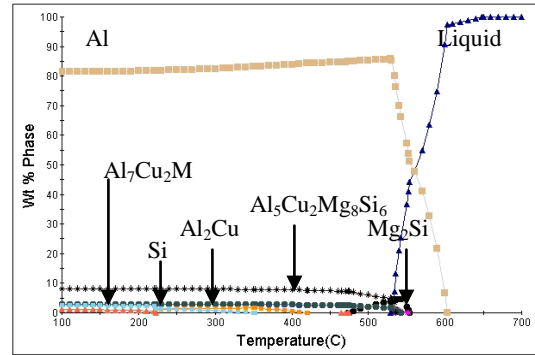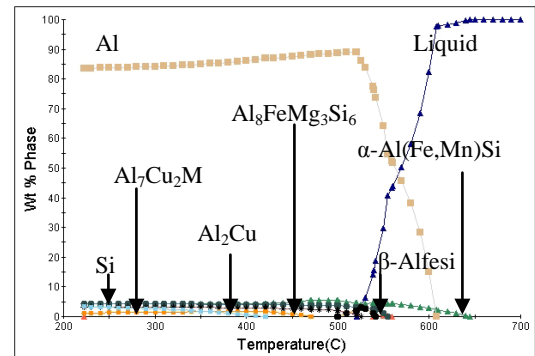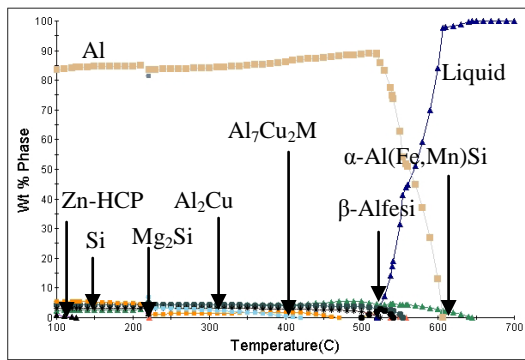Fig. 17 Equilibrium diagram of alloy E obtained from JMatPro simulation

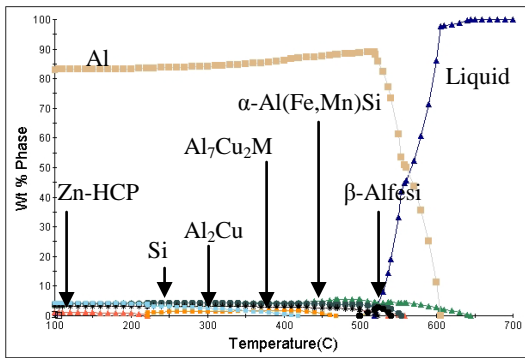Fig. 18 Equilibrium diagram of alloy F obtained from JMatPro simulation

Fig. 19 Equilibrium diagram of alloy G obtained from JMatPro simulation

Table. 3 Various compounds of A319 and modified A319 (A to G)

Alloys	Compounds	Temperature (°C)
A319	Al_2Cu	100-449
	Al_7Cu_2M	222-493
	β-Alfesi	100-322 / 515-563
	$Al_5Cu_2Mg_8Si_6$	100-400
	α-Al(Fe,Mn)Si	100-641
A	Al_2Cu	100-423
	Al_7Cu_2M	222-472
	β-Alfesi	540-560
	α-Al(Fe,Mn)Si	100-644
	$Al_5Cu_2Mg_8Si_6$	100-526
	$Al_8FeMg_3Si_6$	500-542
B	Al_2Cu	223-403
	Al_7Cu_2M	222-458
	β-Alfesi	547-556
	α-Al(Fe,Mn)Si	222-645
	$Al_5Cu_2Mg_8Si_6$	100-533
	$Al_8FeMg_3Si_6$	492-548
C	Al_2Cu	222-380
	Al_7Cu_2M	483-551
	β-Alfesi	551-553
	α-Al(Fe,Mn)Si	100-646
	$Al_5Cu_2Mg_8Si_6$	100-535
	$Al_8FeMg_3Si_6$	483-551
	Mg_2Si	535-550
D	Al_2Cu	100-351
	Al_7Cu_2M	222-420
	β-Alfesi	464-474
	α-Al(Fe,Mn)Si	100-647
	$Al_5Cu_2Mg_8Si_6$	100-550
	$Al_8FeMg_3Si_6$	474-553
	Mg_2Si	530-558
E	Al_2Cu	222-420
	Al_7Cu_2M	222-471
	β-Alfesi	538-558
	α-Al(Fe,Mn)Si	222-644
	$Al_5Cu_2Mg_8Si_6$	222-525
	$Al_8FeMg_3Si_6$	499-540
F	Al_2Cu	210-419
	Al_7Cu_2M	100-469
	β-Alfesi	537-558
	α-Al(Fe,Mn)Si	100-644
	$Al_5Cu_2Mg_8Si_6$	100-524
	$Al_8FeMg_3Si_6$	500-540
	Mg_2Si	220
	Zn_HCP	100-126
G	Al_2Cu	100-418
	Al_7Cu_2M	221-468
	β-Alfesi	536-558
	α-Al(Fe,Mn)Si	100- 645
	$Al_5Cu_2Mg_8Si_6$	100-518
	$Al_8FeMg_3Si_6$	500-540
	Zn_HCP	100-105

V. SELECTION OF ALLOY COMPOSITION

The results from the previous section can be used to select the appropriate alloy composition for thixoforming. Each of the approaches used to design the alloys discussed in this section ensures that only those alloys that fulfill specific criteria can be chosen for use in SSM processing. The design criteria are the working window temperature, solidification temperature range and fraction liquid sensitivity. Note that all the calculations are based on 0.4wt% fraction liquid.

We now discuss with the reference to the specific examples in Section III. The first example is that of A319 aluminium alloy, while the subsequent examples relate to A319 modified by the addition of either magnesium (alloys A to D) or zinc (alloys E to F)

Figs. 2 and 3 in section III show the working window temperature as a function of the magnesium and zinc concentration in A319 aluminium alloys, respectively. Magnesium and zinc have the ability to enlarge the working window temperature of A319 alloys, thereby making them more stable for SSM processing. Figs.4 and 5 show the processing temperature as a function of the fraction liquid. It was observed that in alloy D, the processing temperature increased from 547°C to 551°C when 2.5wt% of magnesium was added.

Fig.6 shows the solidification temperature range for A319 and modified A319. All the alloys have a solidification range

below 150K except for alloys F and G. Therefore, alloys F and G will be affected by hot tearing. These results show that the addition of magnesium has a significant effect on the solidification temperature range of A319 aluminium alloys when compared to the addition of zinc.

Fig. 7 shows the fraction liquid as a function of processing temperature and fraction liquid sensitivity. The addition of magnesium and zinc from 0.1wt % to 2.5wt% and 1.5wt% to 2.5wt%, respectively, causes a reduction of fraction liquid sensitivity from 0.018K to 0.08K and 0.018K to 0.003K. It would therefore appear that, the liquid formation for all the studied alloys tends to be controllable and suitable for semisolid processing.

Table 3 shows the various compounds for alloy A319 and modified A319. All alloys have the formation of intermetallic compounds of Al_2Cu. Alloys C, D and F have higher magnesium content and these alloys are observed to have primary Mg_2Si, which increase strength and heat treatability. For alloy F, Mg_2Si is formed in a small amount at 220°C and such an amount does not contribute to the improvement of the mechanical properties of the alloy. Therefore, in this work, the most promising compositions that fulfill the alloys design criteria were found to be alloys A to E.

VI. CONCLUSION

Thermodynamic simulation is a very important tool in the modification or design of new alloys for thixoforming. There are three design criteria that should be considered in the selection of the suitable alloys; working window temperature, solidification temperature range and fraction liquid sensitivity. The working window temperature is enlarged from 18°C to 26°C by magnesium addition up to 2.5wt % and from 18°C to 24°C by zinc addition up to 2.5wt%. The processing temperature for all modified alloys is decreased at 0.4wt % fraction liquid, except for alloy D where the processing temperature is increased to 551°C. All the alloys have a eutectic reaction temperature below the processing temperature. The effect of magnesium is different from that of zinc, in that the zinc content does not appear to have a significant effect on the working window temperature.

The solidification range of all the modified alloys is below 150K, except for alloys F and G, with a 2.0wt % and 2.5wt% zinc addition. The sensitivity of the alloys group is decreased when either magnesium or zinc is added to A319 alloys. The binary eutectic reaction of all studied alloys occurs between 0.3wt% and 0.5wt% fraction liquid.

The primary formation of α-Al(Fe,Mn)Si, β-AlFesi, $Al_5Cu_2Mg_8Si_6$, Al_7Cu_2M and Al_2Cu occurs during the solidification process of the A319 alloy. When the magnesium content in A319 is increased from 0.1wt% to 1.5wt%, a new phase of $Al_8FeMg_3Si_6$ takes place and when the magnesium content is increased from 1.5wt% to 2.5wt%, a new phase of Mg_2Si is formed. For alloys E to G, when the zinc content is increased from 1.0wt% to 1.5wt%, a new phase of $Al_8FeMg_3Si_6$ is formed and when the zinc content is increased to 2.0wt %, a new Mg_2Si and Zn_HCP are formed.

Based on the simulation results of modified A319, it is recommended that alloys A to E can be used as potential candidates for thixoforming. These alloys also have a potential for age hardening, hence improve their mechanical properties.

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