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The effect of Si upon the interfacial reaction characteristics in SiCp/Al-Si system composites during multiple-remelting

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The composite interfaces play an important role in determining the resultant composite properties, especially the development of interfacial reaction during remelting is critical to the commercialization and sustainable-development of metal matrix composites. In this paper, the interfacial reaction characteristics of SiCp/Al-Si system composites during multiple-remelting were investigated by Differential Scanning Calorimeter (DSC). It was found that the interfacial reactions were not sensitive to remelting number, remelting temperature and reinforcement volume fraction after a degree of reaction, and the results also suggested that the preventation effects of Si upon the interfacial reaction SiCp/Al were mainly attributed to the Si released from the interfacial reaction, while the original Si content in the master alloy also has the same effect only after a given Si content.

1. Introduction

Engineering interest in aluminum-based metal matrix composites (MMCs) has increased, owing to their high specific strength and high specific stiffness. A great portion of the research effort is focused on the producing the MMCs by solidification processing, which is likely to be more economical and relatively simple in comparison with the competing solid processing [1–4]. But unfortunately, all the technological and theoretical difficulties encountered in the manufacture and application of these materials are not entirely solved. For instance, chemical interaction is still a major problem to be considered, when these material are fabricated by solidification processing.

It is well established that SiC react with molten aluminum, producing Al_4C_3 and silicon, according to the following reaction

$$4Al + 3SiC \rightarrow Al_4C_3 + 3Si \tag{1}$$

silicon formed in this reaction dissolves in unreacted aluminum, giving rise to an Al-Si alloy [5–7]. Indeed the chemical stability of SiC in contact with molten aluminum has been widely suggested in both wrought and cast Al-Si based MMCs. However, the knowledge of the interfacial reaction characteristics between the reinforcement and the matrix during multiple remelting have not been reported yet, which is valuable in the selection of composite system and also a main problem in the large commercialization and self-sustainable development of this kind of advanced materials under the effect of environmental consciousness.

It is very difficult to characterize interfaces of this nature by their intrinsic mechanical properties, the interface phenomenon in MMCs is not fully understood because of its complexity and dependence on many variables. Various techniques can be applied to determine the interfacial reaction between the matrix and the reinforcement, most of them are concentrated on carrying out the measurements on the solid reaction products after the reaction at the ambient temperature [8–10], and in the previous works, the authors reported the methods to determine the interfacial reaction using Liquid Metal X-ray Diffraction [11, 12], which allowed the observation of the reaction characteristics above liquidus when the reinforcement and the molten matrix are co-existing. In the present study, Differential Scanning Calorimitry was used to describe the interfacial

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reaction characteristics during multiple remelting for SiCp/Al-Si system composites.

2. Experimental

2.1. Material preparation

SiCp/Pure Al and SiCp/Al-Si composites used in this study were fabricated using Vacuum-high Pressure Infiltration Processing. The dominant phase of SiCp used as reinforcement was α-SiC(6H) and the average size of SiCp was about 7 μm. SiCp used in this study had not undergone any surface modification such as surface oxidation, surface coating etc. A series of SiCp/Pure Al and SiCp/Al-Si composites were used to vary the interfacial reaction characteristics during multiple remelting, i.e., SiCp/Al-5Si, SiCp/Al-8Si, SiCp/Al-25Si composites with 10% reinforcement volume fraction and SiCp/Pure Al composites with 10 and 40% reinforcement volume fraction, respectively.

2.2. Differential scanning calorimeter experiment

For the DSC work, the composite sample were remelted in a high purity alumna pan in Netsch DSC404 instrument. Multiple runs were carried out from ambient to desired temperature, subsequently cooling to ambient temperature, and under dynamic high purity argon atmosphere (80 ml/min). In order to avoid the effect of settlement of discontinuous reinforcement during remelting, the 20 °C/min of heating and cooling rate were applied [13]. High purity corundum was used as a reference, and all the DSC thermogram were normalized to the actual amount of metal (in weight percent) in each composites.

3. Results and observations

According to Equation 1, SiC could react with molten aluminum, producing Al_4C_3 and silicon. Simensen has established that carbon solubility in molten aluminum, found experimentally by Oden and McCune, is less than 2.34 ppm, the system Al-C-Si can be approximated with the well known binary subsystem Al-Si below 1100 °C [14, 15].

The acquired data were analized in the form of heating curve in order to set up the connection of interfacial reaction characteristics between the fabrication and the subsequently remelting, and the thermal analysis parameters that were analyzed in the present study were indicated in Fig. 1. The figure showed the heating curve obtained for Al-Si eutectic alloy heated at the rate of 20 °C/min, indicating that the eutectic reaction will be one main peak at this heating rate. Various parameters analyzed were indicated what they represent.

Fig. 2 showed the multiple heating curve of SiCp/pure Al composites with 10% reinforcement volume fraction. As seen in Fig. 2, in the first heating curve, the onset temperature began at 648.1 °C, and the matrix alloy became completely molten at 682.2 °C, which was the temperature that the melting of matrix alloy reached its maximum, at the same time, the undercooling marked as A in the Fig. 2 was observed. It is well known that the melting point of pure aluminum is 660 °C, while the eutectic temperature is 577 °C in binary Al-Si system and maximum solubility of Si in solid Al is 1.65 at %. Both the onset temperature of the melting and the undercooling phenomena indicated that the interfacial reaction between SiC and pure Al was less serious during fabrication, and the content of Si released from the interfacial reaction does not exceed the maximum solubility of Si in solid Al. In the second and third heating curve, the melting began at 578.8 and 577.8 °C, respectively, which was the onset temperature of eutectic reaction of Al-Si, and the matrix alloy became completely molten at 615.3 and 615 °C,

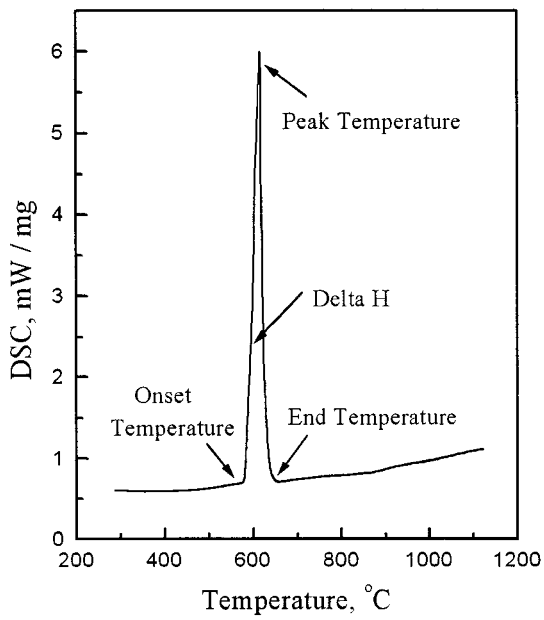

Figure 1 Representation of characteristic thermal analysis parameters used and analized in the present study; illustrated with heating curve obtained for Al-Si eutectic alloy heated at 20 °C/min.

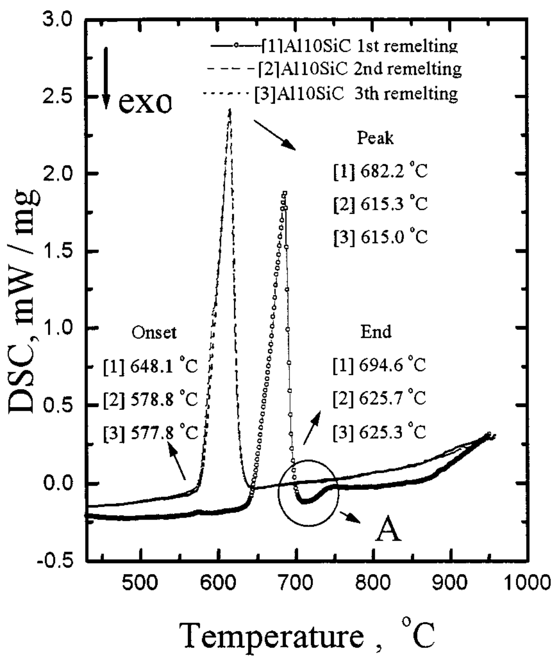

Figure 2 The DSC heating curve of 10 vol % SiCp/pure Al composites remelted at 1050 °C.

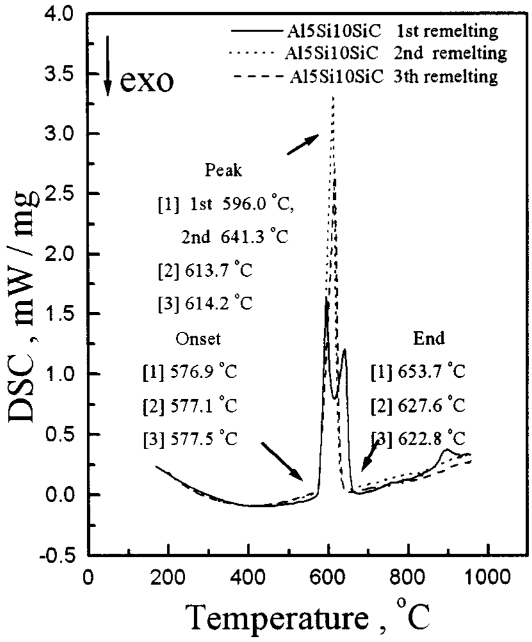

Figure 3 The DSC heating curve of 10 vol % SiCp/Al-5Si composites remelted at 1050 °C.

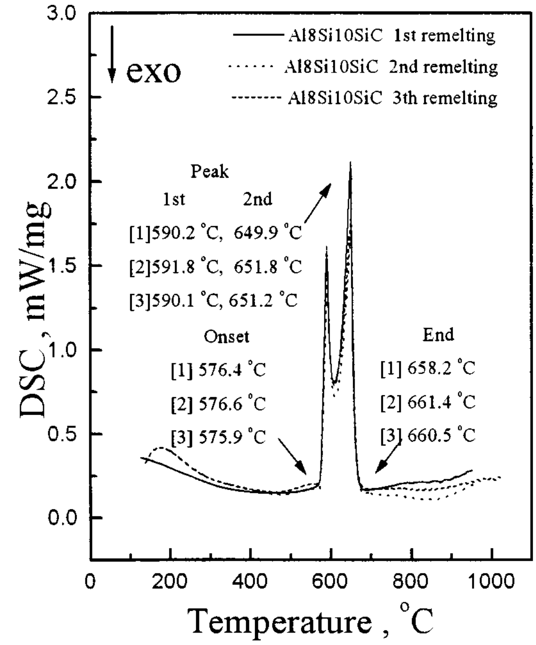

Figure 4 The DSC heating curve of 10 vol % SiCp/Al-8Si composites remelted at 1050 °C.

respectively. The onset temperature of eutectic reaction was not identical to the equilibrium eutectic temperature of 577 °C, which indicated that the superheating effects are significant for the metling of SiCp/Al composites at the heating rate of 20 °C/min.

Fig. 3 showed the multiple heating curve in SiCp/Al-5Si composite. In the first heating curve, two peaks were observed. Based on the Al-Si phase diagram where the basic microstructure was primary aluminum and Al-Si eutectic phase for Al-5Si alloy, the first peak was the melting of eutectic phase where it began at 576.9 °C and completed at 596 °C, the second one was the melting of primary aluminum. In the second and third heating curve, only one eutectic peak was observed at the heating rate of 20 °C, indicating that the interfacial reaction was serious during the second and the third remelting than that of the first remelting, where the eutectic peak was so large that the sub-peak of primary aluminum was hidden into the eutectic peak.

Fig. 4 showed the multiple heating curve of SiCp/Al-8Si composites with 10% reinforcement volume fraction. As seen in Fig. 4, every heating curve consisted of two peaks, eutectic peak and primary aluminum peak, respectively, and these three curve were nearly emerging, where the onset temperature, peak temperature of eutectic peak and the peak temperature of primary aluminum were nearly identical in every heating curve. These results suggested that the interfacial reaction in SiCp/Al-8Si composites was less during multiple remelting.

Fig. 5 showed the multiple heating curve of SiCp/Al-25Si. AS seen in Fig. 5, two peaks were observed in every heating curve, indicating the eutectic peak and primary Si peak, respectively. The heating curve were also nearly emerging, where the thermal parameters were more or less identical. These results also suggested that the interfacial reaction in SiCp/Al-25Si was nearly nought even during multiple remelting.

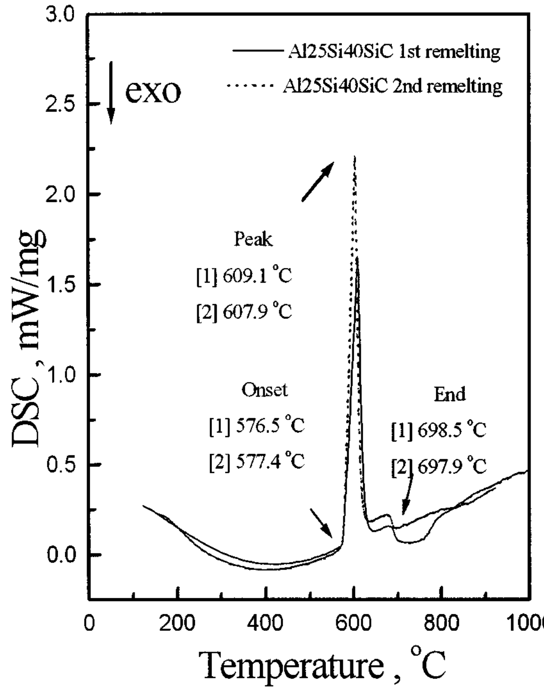

Figure 5 The DSC heating curve of 10 vol % SiCp/Al-25Si composites remelted at 1050 °C.

Figs 6 and 7 showed the multiple heating curve of SiCp/Pure Al composites with 40% reinforcement volume fraction remelted at the temperature of 1050 and 750 °C, respectively. As seen in Figs 6 and 7, in their first heating curve, only one peak and undercooling phenomena marked as B and C in the figures were also observed, where the melting of matrix alloy began at 642.8 and 641.8 °C, respectively, indicating the distribution of particle reinforcement was uniform in the matrix. But in their second and third heating curve, in Fig. 7, two thermal peaks where the first one was eutectic peak and the second one was primary aluminum, while only one main peak was observed in Fig. 6, where the

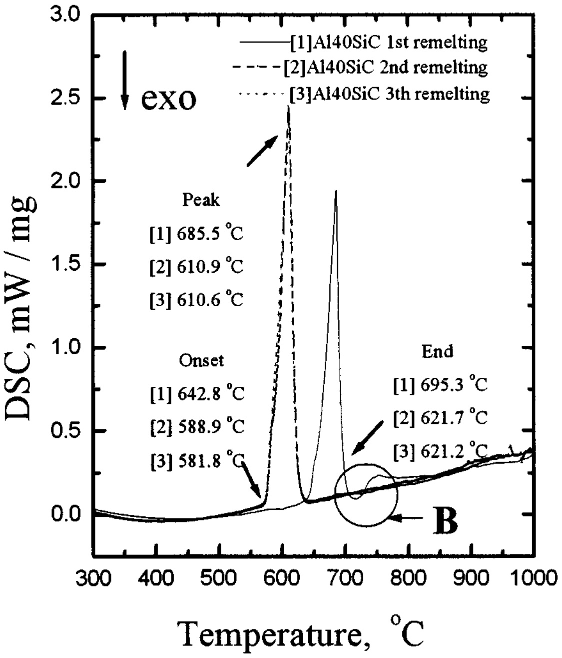

Figure 6 The DSC heating curve of 40 vol % SiCp/pure Al composites remelted at 1050 °C.

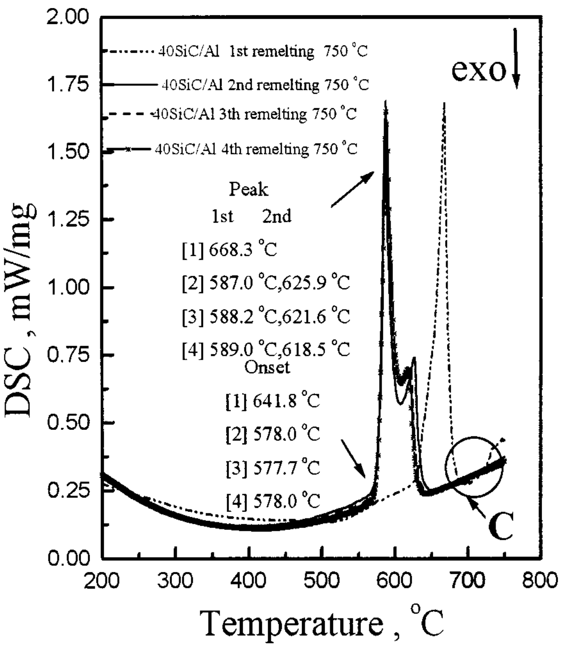

Figure 7 The DSC heating curve of 40 vol % SiCp/pure Al composites remelted at 750 °C.

sub-primary aluminum peak was hidden in the eutectic peak. These results suggested that the high remelting temperature had a greater effect upon the interfacial reaction during multiple remelting than low temperature.

4. Discussion

According to Equation 1, the addition of Si to the matrix can prevent the chemical reaction between SiC and aluminum. As indicated in Fig. 2 to Fig. 5, the second and third heating curve in SiCp/Al-5Si composites were nearly emerging after the first remelting, indicating that the interfacial reaction nearly reached the equilibrium, and all the heating curve in SiCp/Al-8Si and SiCp/

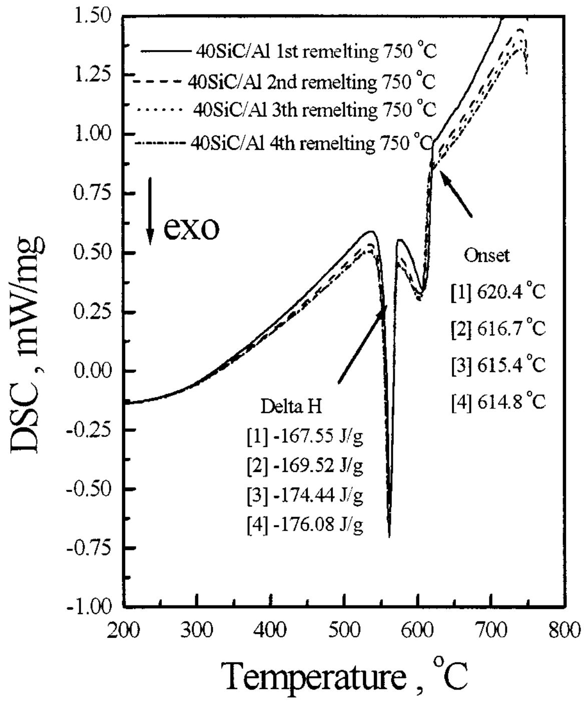

Figure 8 The DSC cooling curve of 40 vol % SiCp/pure Al composites remelted at 750 °C.

Al-25Si composites were nearly emerging, without obvious difference in their thermal parameters, respectively. Both the results suggested that during remelting, the interfacial reaction could be effectively prevented by the addition of a given content of Si in the master alloy.

As indicated in figures above, the interfacial reaction were not sensitive to remelting temperature, remelting number and reinforcement fraction after a degree of chemical reaction. Based on the Al-Si binary diagram, when Equation 1 took place, the liquidus temperature will be depressed and the eutectic reaction exothermic heat will be increased in their DSC cooling curve, So the interfacial reaction evolution in SiCp/Al can be evaluated by the changed liquidus temperature and eutectic reaction exothermic heat in the DSC cooling curve. Figs 8 and 9 showed the evolution of liquidus temperature and exothermic heat of eutectic reaction and corresponding Si content during the multiple remelting in SiCp/pure Al at 750 °C, indicating that after the first remelting, the interfacial reaction was progressively a constant. Several studies [16–21] have been made to predict the silicon contents required to prevent the interfacial reaction between SiC and molten pure aluminum, as demonstrated in Fig. 10, it was obvious that the meta-equilibrium Si content of Fig. 9 was in good agreement with the reports by Iseki *et al.* [20, 21], but much lower than other reports showed in Fig. 10. As reported by Gowri and Samul [22], the cooling rate will depress the liquidus and eutectic solidification region, so the exact Si content of the matrix are lower than that of Fig. 9. On the other hand, Fig. 11 showed the temperature dependence of equilibrium Si content in SiCp/Al-Si composites [16, 20, 21, 23]. As indicated in this figure, for 1050 °C, the equilibrium Si will at least be 13 at %, but the liquidus temperature (617.4 °C) of the third heating

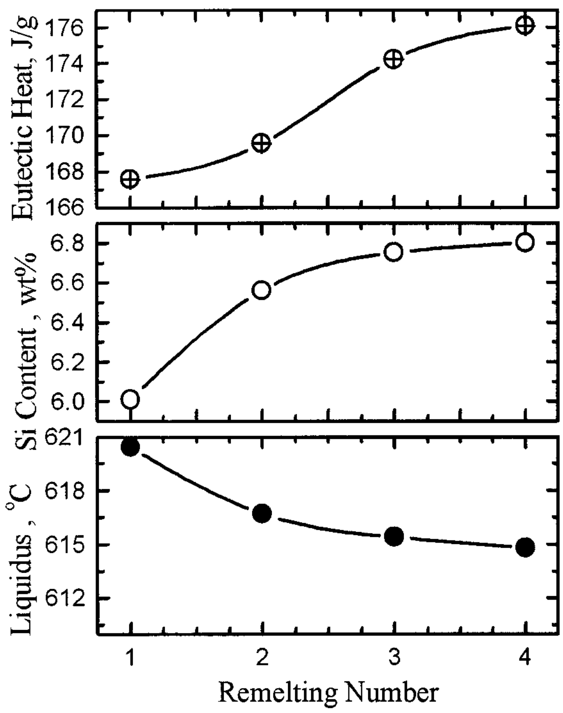

Figure 9 The evolution of interfacial reaction in SiCp/pure Al composites remelted at 750 °C.

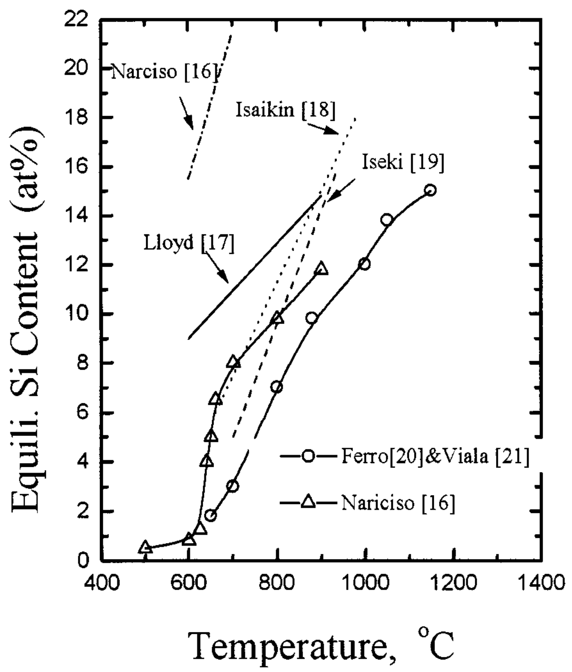

Figure 10 Variation in the equilibrium Si content in SiC/pure Al composite plotted as a function of fabrication temperature.

curve in SiCp/Al-5Si composites (see also in Fig. 9) suggested the nearly equilibrium Si content (6.45 at %) are much lower than 13 at %. These results suggested that the preventation effect of Si upon the interfacial reaction are mainly attributed to the silicon released from the chemical reaction after the chemical reaction took place.

In the previous works [11, 12], the authors investigated the melt structures above the liquidus and the interfacial reaction in SiCp/Pure Al composites using the liquid metal X-ray diffraction and discussed the reaction kinetics between SiC and molten aluminum.

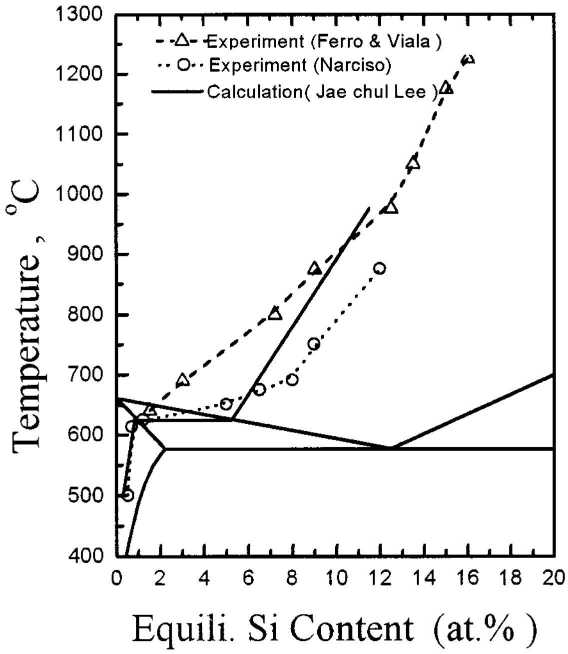

Figure 11 The temperature dependence of equilibrium Si content in SiC/Al-Si composites.

The experimental results showed that Si is not well distributed above the liquidus in SiC/Al composite melt, which main distribute around the SiC particles, and the diffusion of Si released from the interfacial reaction is possible only after a given temperature or a degree concentration fluctuation of Si. On the other hand, during the solidification of discontinuously reinforced hypoeutectic Al-Si composite, the primary phase Al nucleates in the centre of interparticle regions and grows towards the particles so as to avoid the particles. The particles are therefore associated with the last freezing liquid where a eutectic reaction take place [24]. As a result, the near-particle regions are always associated with the Si-riched regime, which will impede the lateral interfacial reaction during the subsequently multiple remelting.

5. Conclusions

In the present work, Differential Scanning Calorimetry was used to investigate the interfacial reaction characteristics during multiple remelting. The following conclusions can be drawn:

(1) The interfacial reaction in SiCp/Al-Si system composites during multiple remelting were not sensitive to the remelting number, remelting temperature and remelting fraction after a degree of chemical reaction.

(2) The preventing effect of Si upon the interfacial reaction are mainly attributed to the Si released from the chemical reaction, while the original Si content in the master alloy also have the same effect only after a given Si content.

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