The influence of temperature on stacking fault energy in Fe-based alloys

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Abstract Temperature has great influence on the stacking fault energy (SFE). Both SFE and $d\gamma_0/dT$ for Fe-based alloys containing substitutional or interstitial atoms increase with increasing temperature. Based on the thermodynamic model of SFE, the equation $\frac{d\gamma_0}{dT} = \frac{d\gamma^{ch}}{dT} + \frac{d\gamma^{seg}}{dT} + \frac{d\gamma^{MG}}{dT}$ and those expressions for three items involved are established. The calculated $d\gamma_0/dT$ is generally consistent with the experimental. The influence of chemical free energy on the temperature dependence of SFE is almost constant, and is obviously stronger than that of magnetic and segregation contributions. The magnetic transition and the segregation of alloying elements at stacking faults cause a decrease in SFE of the alloys when temperature increases; that is, $d\gamma^{MG}/dT < 0$ and $d\gamma^{seg}/dT < 0$. Meanwhile, such an influence decreases with increasing temperature, except for the $d\gamma^{seg}/dT$ of Fe-Mn-Si alloys. With these results, the experimenal phenomena that the SFE of Fe-based alloys is not zero at the thermodynamically equilibrated temperature (T_0) of the γ and ε phases and they are positive both at $T > T_0$ and $T < T_0$ can be reasonably explained.

Keywords: **stacking fault energy(SFE), temperature, Fe-based alloys, segregation, magnetic contribution.**

The stacking fault energy(SFE) in FCC metals and alloys is an important parameter related to mechanical and physical properties. The factors influencing SFE include composition, temperature, segregation of alloying elements at stacking faults, elastic strain, magnetic contribution, and so on. Lots of work has been carried out to investigate the influence of composition on SFE[1—4], but its temperature dependence was generally neglected. Dislocation node measurement using a transmission electron microscope has shown such an influence, and it has been found that SFE increases with the increase in temperature in cubic materials and decreases in hexagonal materials and that SFE is not zero at the temperature of fcc→hcp martensitic transformation[5]. Because SFE changes with temperature, the SFE at Ms will be greatly different from that at room temperature or the equilibrium temperature (T_0) of parent and martensite phases. It was suggested that there exists the carbon pinning at dislocation upon the temperature variation in a Fe-20Mn-4Cr-0.55C alloy, and antiferromagnetic→paramagnetic transition of austenite can also be associated with SFE[6,7]. However, the calculation of magnetic contribution to the temperature derivative of SFE seems not reasonable. If SFE is considered as the difference in Gibbs free energy between a cubic thin platelet and a hexagonal one, the SFE at T_0 temperature would become zero which is inconsistent with the experiments and contradictory to $\gamma_0(T < T_0)$. $\gamma_0(T >$

$T_0) < 0$ from Hirth model[8]. In this paper, the $d\gamma_0/dT$ will be discussed in detail from thermodynamics including segregation of alloying elements at stacking faults and magnetic contribution.

1 Calculation model

According to Hirth[8], the SFE of pure metal is equal to the difference in Gibbs free energy of one-layer atoms of FCC and HCP structures. Hence, based on the change in chemical free energy due to FCC(γ)→HCP(ε) phase transformation, the SFE per unit area can be written as

$$\gamma_0 = \frac{\Delta G^{\gamma\to\varepsilon}}{V^{\frac{2}{3}}(N_0)^{\frac{1}{3}}} \approx \frac{\Delta G^{\gamma\to\varepsilon}}{8.4V^{\frac{2}{3}}}, \tag{1}$$

where $\Delta G^{\gamma\to\varepsilon}$ is the change in free energy during $\gamma\to\varepsilon$ transformation, V the molar volume of metal, N_0 the Avogadro constant. Eq. (1) may be invalid for alloys and may cause some errors. Ericsson[9] considered various influences on the SFE of an alloy and separated the γ_0 into three parts

$$\gamma_0 = \gamma^{\mathrm{TOT}} = \gamma^{\mathrm{ch}} + \gamma^{\mathrm{seg}} + \gamma^{\mathrm{MG}}, \tag{2}$$

where γ^{ch} is the free energy difference between the FCC and HCP structures per unit area of one-layer atoms, γ^{seg} is the energy change due to the segregation of alloying elements at stacking faults, γ^{MG} represents the magnetic contribution to SFE. Then, the temperature derivative of the SFE becomes

$$\frac{d\gamma_0}{dT} = \frac{d\gamma^{\mathrm{TOT}}}{dT} = \frac{d\gamma^{\mathrm{ch}}}{dT} + \frac{d\gamma^{\mathrm{seg}}}{dT} + \frac{d\gamma^{\mathrm{MG}}}{dT}. \tag{3}$$

1.1 Calculation of $d\gamma^{\mathrm{ch}}/dT$

Based on the regular solid solution model, the free energy change $\Delta G^{\gamma\to\varepsilon}$ can be written in the classical thermodynamic form

$$\Delta G^{\gamma\to\varepsilon} = \sum_i^n x_i \Delta G_i^{\gamma\to\varepsilon} + \frac{1}{2}\sum_i^n \sum_j^n \Omega_{ij} x_i x_y, \tag{4}$$

where x_i is the atomic percentage of the component i, $\Delta G_i^{\gamma\to\varepsilon}$ is the free energy change of the component i due to $\gamma\to\varepsilon$ transformation, and Ω_{ij} is a parameter related to the interaction between i and j components ($i \neq j$). Ω_{ij} is assumed to be of little dependence on temperature; that is, $d\Omega_{ij}/dT \approx 0$. Therefore, we have

$$\frac{d\gamma^{\mathrm{ch}}}{dT} = \frac{d\Delta G_i^{\gamma\to\varepsilon}}{dT} = \sum_i^n x_i \frac{d\Delta G_i^{\gamma\to\varepsilon}}{dT}. \tag{5}$$

1.2 Calculation of $d\gamma^{\mathrm{seg}}/dT$

Ishida[10] analysed the contribution from segregation, γ^{seg}, as a result of the concentration difference of alloying elements at stacking faults and in matrix(Suzuki effect). On the basis of his work, the temperature dependence of segregation can be reduced

$$\frac{d\gamma^{\mathrm{seg}}}{dT} = \frac{d\Delta G^{\mathrm{ch(s)}}}{dT} + \frac{d\Delta G^{\mathrm{sur(s)}}}{dT} + \frac{d\Delta G^{\mathrm{els(s)}}}{dT}, \tag{6}$$

where $d\Delta G^{\mathrm{ch(s)}}/dT$ is the temperature derivative of the chemical free energy change due to segregation, $d\Delta G^{\mathrm{sur(s)}}/dT$ is the temperature derivative of the surface free energy change due to the concentration difference at stacking fault and in the matrix, $d\Delta G^{\mathrm{els(s)}}/dT$ is the temperature

derivative of the elastic free energy change due to the variation of mean atom size caused by segregation. The elastic free energy changes for both the substitutional and the interstitial alloying elements in the system are considered to be negligible ($\Delta G^{\mathrm{els(s)}} \approx 0$), and we ignore the item $\frac{\mathrm{d}\Delta G^{\mathrm{els(s)}}}{\mathrm{d}T}$ in this work. According to ref. [10], we obtain

$$\frac{\mathrm{d}\Delta G^{\mathrm{ch(s)}}}{\mathrm{d}T} = \frac{\mathrm{d}\left[RT\sum_i^n x_i \ln\left(\frac{x_i^s}{x_i}\right)\right]}{\mathrm{d}T} = R\left[\sum_i x_i \ln\left(\frac{x_i^s}{x_i}\right) + T\sum_i\left(\frac{x_i}{x_i^s}\cdot\frac{\mathrm{d}x_i^s}{\mathrm{d}T}\right)\right], \tag{7}$$

where x_i^s is the concentration of ith element segregated at stacking fault. Yakubtsov et al.[2] deduced the x_i^s value using a regular solid solution model and the equilibrium condition of γ and ε phases

$$x_i^s = \frac{x_i}{x_i + \sum_j\left[x_j \exp\left(\frac{\Delta G_i^{\gamma\to\varepsilon} - \Delta G_j^{\gamma\to\varepsilon}}{RT}\right)\right]}. \tag{8}$$

For the $\mathrm{d}\Delta G^{\mathrm{sur(s)}}/\mathrm{d}T$, we have

$$\frac{\mathrm{d}\Delta G^{\mathrm{sur(s)}}}{\mathrm{d}T} = \frac{1}{2}W(x_i^s - x_i)\frac{\mathrm{d}x_i^s}{\mathrm{d}T}, \tag{9}$$

where W is the interaction energy of alloying elements with stacking fault. Yakubtsov et al.[2] have suggested that the interaction energy of substitutional atoms with stacking fault is rather small and while that of interstitial atoms with stacking fault has a substantial influence on $\Delta G^{\mathrm{sur(s)}}$. Using eq. (8), the concentration of interstitial atom at stacking fault can be expressed as

$$x_{i(\mathrm{int})}^s = \frac{x_{i(\mathrm{int})}}{x_{i(\mathrm{int})} + (1 - x_{i(\mathrm{int})})\exp\left(-\frac{w}{RT}\right)}. \tag{10}$$

1.3 Calculation of $\mathrm{d}\gamma^{\mathrm{MG}}/\mathrm{d}T$

It is assumed that the average magnetic moment would not change with temperature. From refs. [11—13] we have the temperature derivative of magnetic free energy

$$\frac{\mathrm{d}\gamma^{\mathrm{MG}}}{\mathrm{d}T} = \frac{\mathrm{d}\Delta G^{M(\gamma\to\varepsilon)}}{\mathrm{d}T}, \tag{11}$$

$$\frac{\mathrm{d}\Delta G^{M}}{\mathrm{d}T} = R\ln(\beta + 1)\left[f(\tau) + T\frac{\mathrm{d}f(\tau)}{\mathrm{d}T}\right], \tag{12}$$

where β is the average magnetic moment per atom (in Rohr magnetons) and the parameter $\tau = T/T_C$, while T is the temperature of the interested system and T_C the magnetic transition temperature. In eq. (12), $\mathrm{d}f(\tau)/\mathrm{d}T$ is defined as

$$\frac{\mathrm{d}f(\tau)}{\mathrm{d}T} = 0.4269\left[\frac{\tau^{-6}}{2} + \frac{\tau^{-16}}{21} + \frac{\tau^{-26}}{60}\right] \text{ for } \tau > 1, \tag{13}$$

$$\frac{\mathrm{d}f(\tau)}{\mathrm{d}T} = -0.4269\left[\frac{79}{140}\cdot\frac{1}{p}\cdot\frac{(-1)}{\tau^2} + \frac{474}{497}\cdot\left(\frac{1}{p} - 1\right)\cdot\left(\frac{\tau^2}{2} + \frac{\tau^8}{15} + \frac{\tau^{14}}{40}\right)\right] \text{ for } \tau < 1, \tag{14}$$

where the parameter p is equal to 0.28 for FCC and HCP structures.

2 Calculation parameters

Using the method presented in the previous sections, the $d\gamma_0/dT$ of Fe-Cr-Ni, Fe-Mn-Cr-C, Fe-Cr-Ni-C, Fe-Mn-Si and Fe-Mn-Si-C alloys are calculated, and the results are compared with the experimental. Table 1 lists the free energy changes taken from ref. [14]. The interaction energy of interstitial atoms (carbon) with stacking fault is from ref. [15]. The average magnetic moments β and the magnetic transition temperature T_C for Fe-Mn-Si alloys are calculated using the formulas[16]

$$T^{\gamma}_{C(\text{Fe-Mn-Si})} = 67x_{\text{Fe}} + 540x_{\text{Mn}}[761 + 689(x_{\text{Fe}} - x_{\text{Mn}})];$$
$$\beta^{\gamma}_{(\text{Fe-Mn-Si})} = 0.7x_{\text{Fe}} + 0.62x_{\text{Mn}};$$
$$T^{\varepsilon}_{C(\text{Fe-Mn-Si})} = 580x_{\text{Mn}};$$
$$\beta^{\varepsilon}_{(\text{Fe-Mn-Si})} = 0.62x_{Mn}.$$

Table 1 Chemical free energy change $\Delta G_i^{\gamma\to\varepsilon}$/J·mol^{-1}[14]

$\Delta G_{\text{Fe}}^{\gamma\to\varepsilon}$	$-2243.38 + 4.309T$
$\Delta G_{\text{Cr}}^{\gamma\to\varepsilon}$	$-2856 - 0.163T$
$\Delta G_{\text{Ni}}^{\gamma\to\varepsilon}$	$1045 + 1.255T$
$\Delta G_{\text{Mn}}^{\gamma\to\varepsilon}$	$-1000 + 1.123T$
$\Delta G_{\text{Si}}^{\gamma\to\varepsilon}$	$-1800 + T$
$\Delta G_c^{\gamma\to\varepsilon}$ a)	$4.88T - 0.01T\ln(T) + 135400T^{-1} + 3.3\times10^{6}T^{-2} - 9\times10^{8}T^{-3}$

a) Approximately treated as the difference in those of graphite and diamond.

3 Results and discussion

3.1 Fe-Based alloys with substitutional but not interstitial atoms

The calculated results are shown in table 2. For the Fe-(17—20)Cr-(12—15)Ni (at%) alloys, the influence of segregation ($d\gamma^{\text{seg}}/dT$) enhances gradually with decreasing temperature, which is reasonable and consistent with the theory of diffusion. Based on the experimental thermodynamic data, the temperature derivative of chemical free energy ($d\gamma^{\text{ch}}/dT$) exhibits a slight variation. However, $\dfrac{d\gamma^{\text{seg}}}{dT}\cdot\dfrac{d\gamma^{\text{ch}}}{dT} < 0$ indicates that the contributions of segregation and chemical free energy to SFE are opposite, implying that the SFE with segregation is smaller than the SFE without segregation. Though the relevant parameters of magnetic properties are not available for these alloys, the calculations without considering $d\gamma^{\text{MG}}/dT$ are quite consistent with the experiments. There exists great difference between the calculation and experiments of Remy et al's[6,7]. It seems also not reasonable that the $d\gamma_0/dT$ does not change with temperature as they suggested. There has been no work on the $d\gamma_0/dT$ of Fe-Mn-Si alloys until now. The calculation of $d\gamma_0/dT$ for a Fe-25Mn-11Si (at%) alloy (table 2) indicates that $d\gamma^{\text{MG}}/dT$ is negative and the $|d\gamma^{\text{MG}}/dT|$ decreases with increasing temperature. Though $d\gamma^{\text{seg}}/dT < 0$, its absolute value increases with an increase in temperature, contrary to that of Fe-Ci-Ni alloys. A possible reason might be that manganese belongs to a kind of volatile elements causing different segregation

behavior. On the other hand, $|d\gamma^{seg}/dT|$ (or $|d\gamma^{MG}/dT|$) $< d\gamma^{ch}/dT$ shows that segregation and magnetic contribution have rather small influence on SFE. To confirm these calculated results more experimental evidence is needed.

Table 2 Temperature dependence on SFE in ferrous alloys with substitutional atoms

Alloy(at%) $\left(\frac{d\gamma_0}{dT}\right)^{Exp.}$ mJ/m²·K	$T(K)$	$\frac{d\gamma^{ck}}{dT}$	$\frac{d\gamma^{seg}}{dT}$	$\frac{d\gamma^{MG}}{dT}$	$\frac{d\gamma^{TOT}}{dT}$	$\left(\frac{d\gamma_0}{dT}\right)^{Cal.}$ mJ/(m²·K)
		$\left(\frac{d\gamma_0}{dT}\right)^{Cal}$ (this work) mJ/(m²·K)				
Fe-17Cr-14Ni 0.08[17]	200	0.097	-0.094		0.003	0.18[7]
	300		-0.049		0.048	
	400		-0.031		0.066	
	500		-0.022		0.075	
	600		-0.016		0.081	
Fe-19Cr-13Ni 0.06[17]	200	0.096	-0.088		0.008	0.17[7]
	300		-0.045		0.051	
	400		-0.028		0.068	
	500		-0.020		0.076	
	600		-0.015		0.081	
Fe-19Cr-12Ni 0.10[18]	200	0.093	-0.082		0.016	0.17[7]
	300		-0.042		0.056	
	400		-0.027		0.071	
	500		-0.019		0.079	
	600		-0.015		0.083	
Fe-20Cr-15Ni 0.05[18]	200	0.098	-0.098		-0.005	0.16[7]
	300		-0.049		0.044	
	400		-0.030		0.063	
	500		-0.020		0.073	
	600		-0.016		0.076	
Fe-25Mn-11Si	300	0.100	-0.001	-0.007	0.092	
	400		-0.003	-0.002	0.095	
	500		-0.004	-0.001	0.095	
	600		-0.005	~0	0.095	
	700		-0.005	~0	0.095	

3.2 Fe-based alloys with interstitial atoms

Many studies have been performed to evaluate the thermodynamic behavior of interstitial elements in alloys, but few suitable thermodynamic approaches were established to deal with its magnetic behavior. It is still unclear whether interstitial atoms make contribution to the magnetic free energy because there are no relevant experiments. Remy[6] measured the SFE of an FCC Fe-20Mn-4Cr-0.5C alloy at various temperatures in the range of 100—390 K through TEM observation of dislocation nodes and shear modulus determinations and calculated the relationship between SFE and temperature. According to his experiment, γ_0 showed a nonlinear variation with temperature T and increased dramatically above 300 K. He thought that it is caused by a substantial magnetic contribution, $d\gamma^{MF}/dT = 0.29$ mJ/(m²·K) at 400 K. However, it seems too high and lacks direct experimental support. His calculations were obviously far from the experimental re-

sults——$(d\gamma_0/dT)^{Cal}[=0.20\ mJ/(m^2\cdot K)] > (d\gamma_0/dT)^{Exp.}[=0.08\ mJ/(m^2\cdot K)]$. In this paper, we deal with this alloy. The results in table 3 show that the magnetic contribution to SFE exerts slight influence on the temperature dependence of SFE, and $d\gamma^{MG}/dT$ reduces with the increase in temperature, which is inverse to Remy's relationship between $d\gamma^{MG}/dT$ and T. Compared with $\Delta G^{ch(\gamma\to\varepsilon)}$, $\Delta G^{MG(\gamma\to\varepsilon)}$ is about one fourth in Fe-Mn-Si alloys according to a thermodynamic analysis[16], and so is that in Fe-Mn alloys investigated by Huang et al.[19]. These results supported our calculations. The $d\gamma/dT$ for Fe-Cr-Ni-C alloys are also given in table 3, which are consistent with the previous experiments. For a promising shape memory material, Fe-Mn-Si alloys, in which the alloying element C atom improves its shape memory effect, a Fe-27Mn-6Si-0.4C(weight percent) alloy in a temperature range of 200—700 K is calculated as an example (table 3).

Table 3 Temperature dependence on SFE in ferrous alloys with interstitial atom

Alloy(%)	T/K	$\left(\frac{d\gamma_0}{dT}\right)^{Exp.}$ mJ/(m²·K)	$\left(\frac{d\gamma_0}{dT}\right)^{Cal.}$ (this work) mJ/(m²·K)				$\left(\frac{d\gamma_0}{dT}\right)^{Cal.[6]}$ mJ/(m²·K)	
			$\frac{d\gamma^{ch}}{dT}$	$\frac{d\gamma^{seg}}{dT}$	$\frac{d\gamma^{MG}}{dT}$	$\frac{d\gamma^{TOT}}{dT}$	$\frac{d\gamma^{MG}}{dT}$	$\frac{d\gamma_0}{dT}$
Fe-20Mn-4Cr-0.4C	200	0.01[6]	0.112	-0.072	0.0004	0.0404	0.08	0.08
	300	0.05[6]	0.113	-0.037	-0.010	0.066	0.25	0.17
	400	0.08[6]	0.113	-0.025	-0.002	0.086	0.29	0.20
	500		0.113	-0.020	-0.001	0.092		
	600		0.113	-0.016	-0.0003	0.0967		
Fe-18Cr-7Ni-0.18C	200	0.10[20]	0.103	-0.055		0.048		
	300		0.103	-0.030		0.073		
	400		0.103	-0.020		0.083		
	500		0.103	-0.012		0.091		
	600		0.103	-0.010		0.093		
Fe-27Mn-6Si-0.4C	200		0.099	-0.036	……	0.063		
	300		0.102	-0.018	-0.009	0.075		
	400		0.102	-0.012	-0.002	0.088		
	500		0.102	-0.009	-0.001	0.092		
	600		0.102	-0.007	~0	0.095		
	700		0.102	-0.006	~0	0.096		

3.3 SFE at T_0 temperature

Based on the above analyses, SFE of alloys is the function of composition and temperature, and can be generally written as (with Ms as a reference temperature)

$$\gamma_0 = \gamma_0(T, x_1, x_2, \cdots) = \gamma_0|_{T=Ms} + \int_{Ms}^{T} \frac{\partial \gamma_0}{\partial T} dT. \tag{15}$$

Experiments have already proved that SFE at Ms temperature is not zero[5]. Because $d\gamma_0/dT > 0$ as we calculated, $\gamma|_{T_0} \neq 0$ is readily deduced from (15). Meanwhile, another conclusion of $\gamma_0|_{T_1} \cdot \gamma_0|_{T_2} > 0 (T_1 > T_0 > T_2)$ can be made, which solves the inconsistency from Hirth model[8].

3.4 Magnetic contribution

Tables 2 and 3 show that the magnetic influence on SFE(γ^{MG}) is small, but the value of this part of the SFE (γ^{MS}) was not involved in the above discussion. Here we take the Fe-Mn-Si alloys as an example to calculate the γ^{MG}, including the influence of composition for Fe-22Mn-(1—7) Si (wt%) alloys. Fig. 1 shows that $\gamma^{MG} < 1.2$ mJ/m^2, and both γ^{MG} and $|d\Delta G^M/dT|$ ($d\Delta G^M/dT < 0$) decrease with an increase in Si content because Si reduces the Neel temperature (T_N). Fig. 2 shows that $\gamma^{MG} < 1.1$ mJ/m^2 when Mn% (wt%) = 32%, and γ^{MG} and $|d\Delta G^M/dT|$ ($d\Delta G^M/dT < 0$) increase with an increase in Mn content. The reason could be that Mn increases T_N. In this kind of alloys, SFE as well as $d\gamma^{MG}/dT$ increases with increasing Mn content and decreasing Si content.

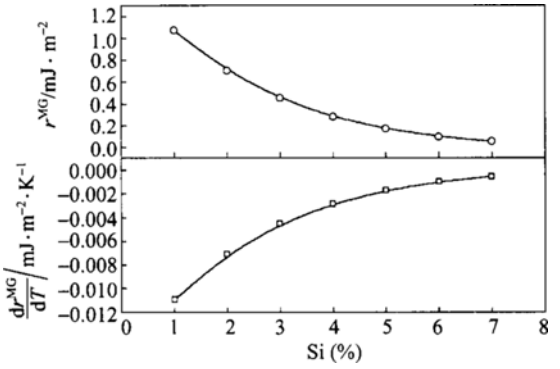

Fig. 1. Magnetic contribution to SFE versus Si content in Fe-22Mn-(1—7) Si alloys. Fe-22Mn-(1—7) Si alloys, T = 400 K.

4 Conclusions

The $d\gamma_0/dT$ of Fe-based alloys with substitutional or interstitial atoms is calculated by using thermodynamics. Based on $\frac{d\gamma_0}{dT} = \frac{d\gamma^{ch}}{dT} + \frac{d\gamma^{seg}}{dT} + \frac{d\gamma^{MG}}{dT}$, it is shown that $d\gamma_0/dT > 0$ and the values of $d\gamma_0/dT$ at various temperatures can be specified. Within a certain temperature range, $\gamma|_{T_0} \neq 0$ and $\gamma_0|_{T_0} \cdot \gamma_0|_{T_0} > 0$ ($T_1 > T_0 > T_2$) can be obtained. The chemical free energy has great effect on the SFE and $d\gamma_0/dT$, and $d\gamma^{ch}/dT > 0$. The influence of segregation ($d\gamma^{seg}/dT$) and magnetic contribution ($d\gamma^{MG}/dT$) are much smaller than $d\gamma^{ch}/dT$, and their influences are opposite to that of the chemical free energy ($d\gamma^{MG}/dT < 0$, $d\gamma^{seg}/dT < 0$). Both $|d\gamma^{MG}/dT|$ and $|d\gamma^{seg}/dT|$ decrease, and $d\gamma_0/dT$ increases with increasing temperature.

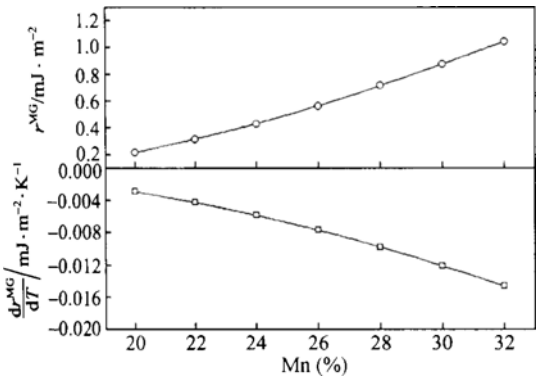

Fig. 2. Magnetic contribution to SFE versus Mn content in Fe-(20—32)Mn-6Si alloys. Fe-(22—32)Mn-6Si alloys, T = 300K.

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