Supplementary Material

If one inserts Eqs. (5-6) into Eq. (7), the Gibbs energy G_{self} becomes a function of the parameters Ωc_T and y. Its equilibrium value $G_{\text{self,eq}}(y)$ is then given by using the value $\Omega c_{T,\text{eq}}$ from Eq. (12) for Ωc_T as

$$G_{\text{self,eq}}(y) = (1-y)G_A + yG_B + \left(Zy - \Omega c_{T,\text{eq}}\right)E + R_gT\left\{\begin{array}{l}(1-Zy)\left[\dfrac{y-\Omega c_{T,\text{eq}}}{1-Zy}\ln\left(\dfrac{y-\Omega c_{T,\text{eq}}}{1-Zy}\right)+\left(1-\dfrac{y-\Omega c_{T,\text{eq}}}{1-Zy}\right)\ln\left(1-\dfrac{y-\Omega c_{T,\text{eq}}}{1-Zy}\right)\right]\\ +Zy\left[\dfrac{\Omega c_{T,\text{eq}}}{Zy}\ln\left(\dfrac{\Omega c_{T,\text{eq}}}{Zy}\right)+\left(1-\dfrac{\Omega c_{T,\text{eq}}}{Zy}\right)\ln\left(1-\dfrac{\Omega c_{T,\text{eq}}}{Zy}\right)\right]\end{array}\right\}. \quad \text{(S1)}$$

As $\Omega c_{T,\text{eq}}$ is a function of y, see Eq. (12), its derivative with respect to *y* reads as

$$\frac{\mathrm{d}\Omega c_{T,\text{eq}}}{\mathrm{d}y} = \Omega c'_{T,\text{eq}} = \frac{(1+Z)}{2} \pm \frac{2(1+Z)^2\, y\left(\tilde{E}-1\right)^2 + 2\left[(1+Z) - 4Zy\tilde{E}\right]\left(\tilde{E}-1\right)}{4\left(\tilde{E}-1\right)\sqrt{(1+Z)^2\, y^2\left(\tilde{E}-1\right)^2 + 2\left[(1+Z)\,y - 2Zy^2\tilde{E}\right]\left(\tilde{E}-1\right)+1}}. \quad \text{(S2)}$$

The derivative $\mathrm{d}G_{\text{self,eq}}/\mathrm{d}y$ follow from Eqs. (S1) and (S2) after some analysis as

$$\frac{\mathrm{d}G_{\text{self,eq}}}{\mathrm{d}y} = -G_A + G_B + \left(Z - \Omega c'_{T,\text{eq}}\right)E + R_gT\left\{\begin{array}{l}Z\left(\begin{array}{l}-\dfrac{y-\Omega c_{T,\text{eq}}}{1-Zy}\ln\left(\dfrac{y-\Omega c_{T,\text{eq}}}{1-Zy}\right)-\left(1-\dfrac{y-\Omega c_{T,\text{eq}}}{1-Zy}\right)\ln\left(1-\dfrac{y-\Omega c_{T,\text{eq}}}{1-Zy}\right)\\ +\dfrac{\Omega c_{T,\text{eq}}}{Zy}\ln\left(\dfrac{\Omega c_{T,\text{eq}}}{Zy}\right)+\left(1-\dfrac{\Omega c_{T,\text{eq}}}{Zy}\right)\ln\left(1-\dfrac{\Omega c_{T,\text{eq}}}{Zy}\right)\end{array}\right)+\\ \dfrac{\Omega c'_{T,\text{eq}}(1-Zy)+Z\Omega c_{T,\text{eq}}-1}{1-Zy}\ln\left(\dfrac{1-Zy}{y-\Omega c_{T,\text{eq}}}-1\right)+\dfrac{\Omega c'_{T,\text{eq}}\,y-\Omega c_{T,\text{eq}}}{y}\ln\left(\dfrac{\Omega c_{T,\text{eq}}}{Zy-\Omega c_{T,\text{eq}}}\right)\end{array}\right\}. \quad \text{(S3)}$$

The second derivative $\mathrm{d}^2G_{\text{self,eq}}/\mathrm{d}y^2$ represents a very lengthy expression, and it is recommended to calculate it from Eqs. (S2) and (S3) by using a mathematical software.

The chemical potential $\mu_{B,\text{CAL}}(y)$ can be calculated by means of the CALPHAD approach, see, e.g., [15] for a bcc Fe-Al system dilute in Al ($\text{A} \to \text{Fe}$ and $\text{B} \to \text{Al}$). The chemical potentials $\mu_{Al,\text{reg}}(y)$ (see Eq. $(15)_2$) and $\mu_{Al,\text{self}}(y)$ (see Eq. $(13)_2$) are fitted to the actual $\mu_{Al,\text{CAL}}(y)$ by adapting the value of E, see the values of E in Fig. S1 and Table S1. From Fig. S2 and Table S1 (see Supplementary Material [14] for Figs. S1, S2 and Table S1), it is evident that the agreement of $\mu_{Al,\text{self}}(y)$ with $\mu_{Al,\text{CAL}}(y)$ is much better than that of $\mu_{Al,\text{reg}}(y)$ with $\mu_{Al,\text{CAL}}(y)$, which means that the self-consistent solution model renders reality much better than the regular model.

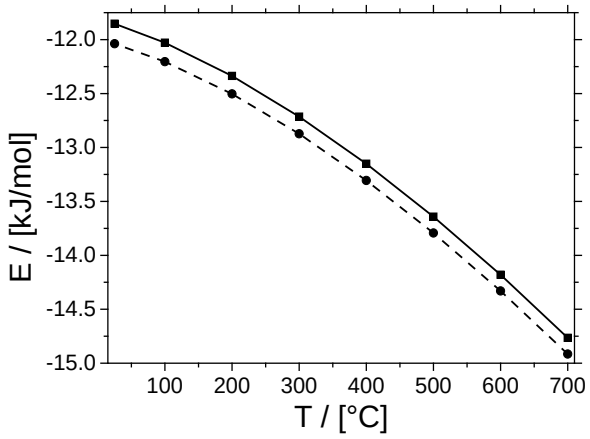

Fig. S1. Fitted values of *E* for the regular model, dashed line, and the self-consistent model, solid line, for different temperatures listed in Table 1 for a bcc Fe-Al system.

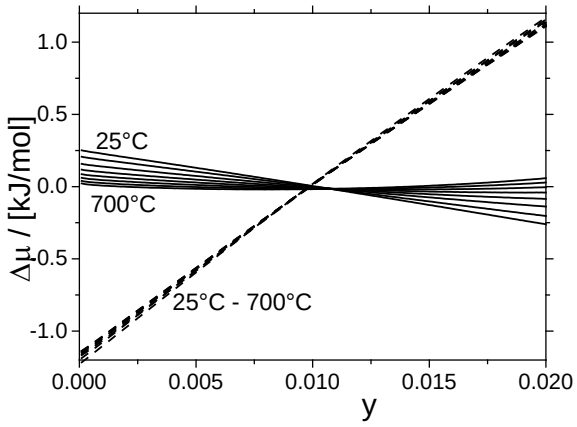

Fig. S2. Difference between $\mu_{Al,\mathrm{CAL}}(y)$ and $\mu_{Al,\mathrm{reg}}(y)$, dashed lines, and between $\mu_{Al,\mathrm{CAL}}(y)$ and $\mu_{Al,\mathrm{self}}(y)$, solid lines, both denoted as $\Delta\mu$, for different temperatures listed in Table 1 for a bcc Fe-Al system.

Table S1: Fitted values of *E* in bcc-Fe with Al (0 to 0.02 at%) at different temperatures, $Z = 8$

T / [°C]	E_{self} / [kJ/mol]	E_{reg} / [kJ/mol]	Least sum of squares	
			self	reg
700	-14.76	-14.91	0.08	92.04
600	-14.18	-14.33	0.04	88.78
500	-13.64	-13.79	0.09	86.73
400	-13.15	-13.30	0.27	85.91
300	-12.71	-12.87	0.68	86.42
200	-12.33	-12.50	1.46	88.42
100	-12.02	-12.20	2.83	92.24
25	-11.85	-12.03	4.42	96.59