Supplementary Materials for:

"The origins of segregation behaviors of solute atoms and their effects on strength of α-Al//θ' -Al_2Cu interfaces in Al-Cu alloys"

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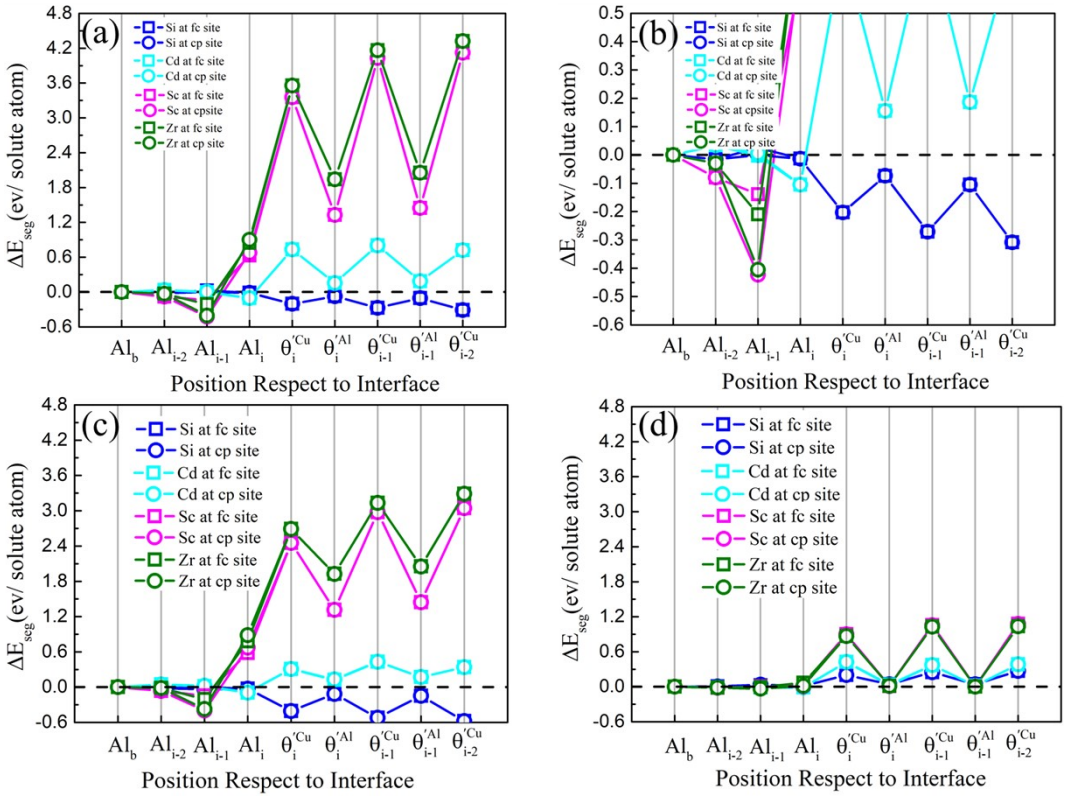

Fig. S1 The segregation energies of Zr, Sc, Cd and Si at the different substitution segregation positions of different atomic layers at the $(001)_{\alpha\text{-Al}}//(001)_{\theta'}$ interface under the condition of fixing the volumes of the interface supercells. (a) The total segregation energy. (b) The enlarged view of figure (a). (c) The chemical contribution of segregation

energy. (d) The mechanical contribution of segregation energy.

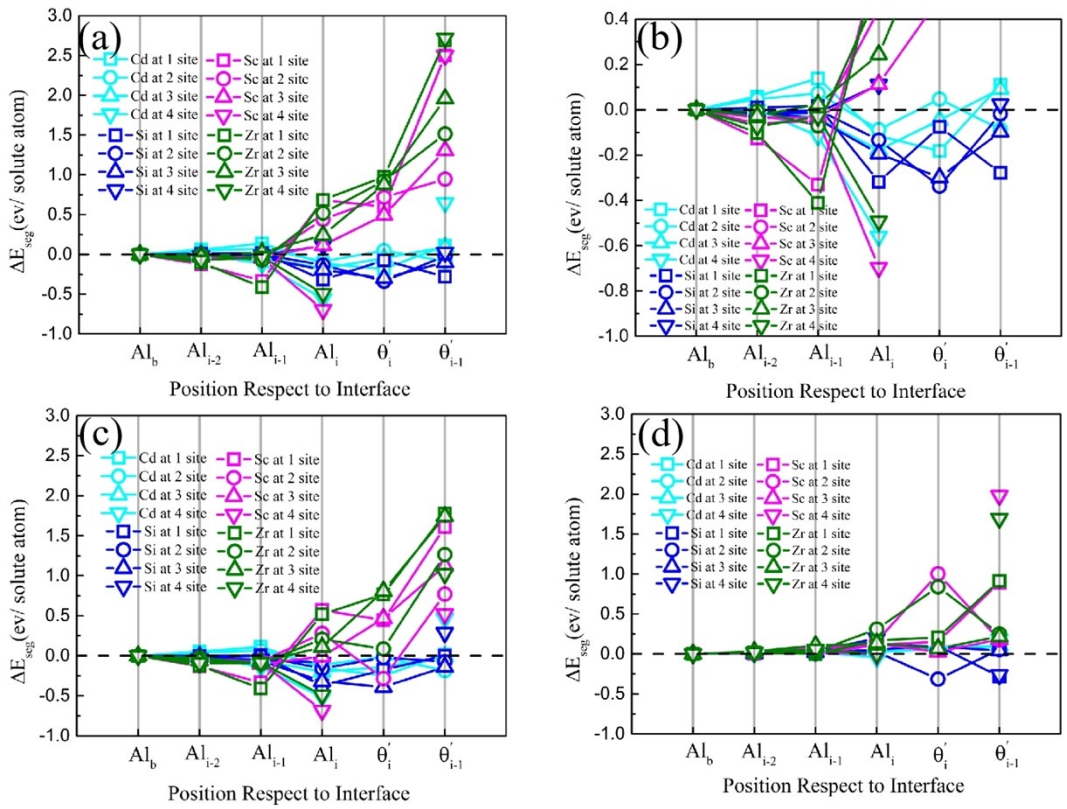

Fig. S2 The segregation energies of Zr, Sc, Cd and Si at the different substitution segregation positions of different atomic layers at the $(010)_{\alpha\text{-Al}}//(010)_{\theta'}$ interface under the condition of fixing the volumes of the interface supercells. (a) The total segregation energy. (b) The enlarged view of figure (a). (c) The chemical contribution of segregation energy. (d) The mechanical contribution of segregation energy.

1. Charge density difference for the $(001)_{\alpha\text{-Al}}//(001)_{\theta'}$

Fig. S3 shows three-dimensional charge density difference ((a), (c), (e), (g) and (i)) and two-dimensional charge density difference ((b), (d), (f), (h) and (j)) along [001] direction for the clean $(001)_{\alpha\text{-Al}}//(001)_{\theta'}$ and $(001)_{\alpha\text{-Al}}//(001)_{\theta'}$ interfaces with Cd, Si, Sc and Zr at sites Al_i-fc, θ'^{Cu}_{i-2}, Al_{i-1}-cp and Al_{i-1}-cp, respectively. It is observed that the charge

redistribution is significantly localized at the interface. That is to say, the interface-induced changes in the electron structure are very short-ranged. Compared with the clean interface (Figs. S3(a) and (b)), it is observed from Figs. S3(c) and (d) that the charge depletion is significant in the region between the interface Al layer on the Al matrix side and $\theta'^{\mathrm{Al}}_{\mathrm{i\text{-}1}}$ layer on the θ' side, which leads to the significant weakening effect of solute atom Cd on the interface. In contrast, the charge accumulation occurs between the solute atom Sc (or Zr) and the host atom Al, as shown in Figs. S3(g), (h), (i) and (j). The result explains the strengthening effect of solute atom Sc (or Zr) on the interface. In particular, Figs. S3(e) and (f) suggest that Si segregation at site $\theta'^{\mathrm{Cu}}_{\mathrm{i\text{-}2}}$ within θ' leads to a very small charge change around Si, explaining why the effect of Si on the interface strength is insignificant.

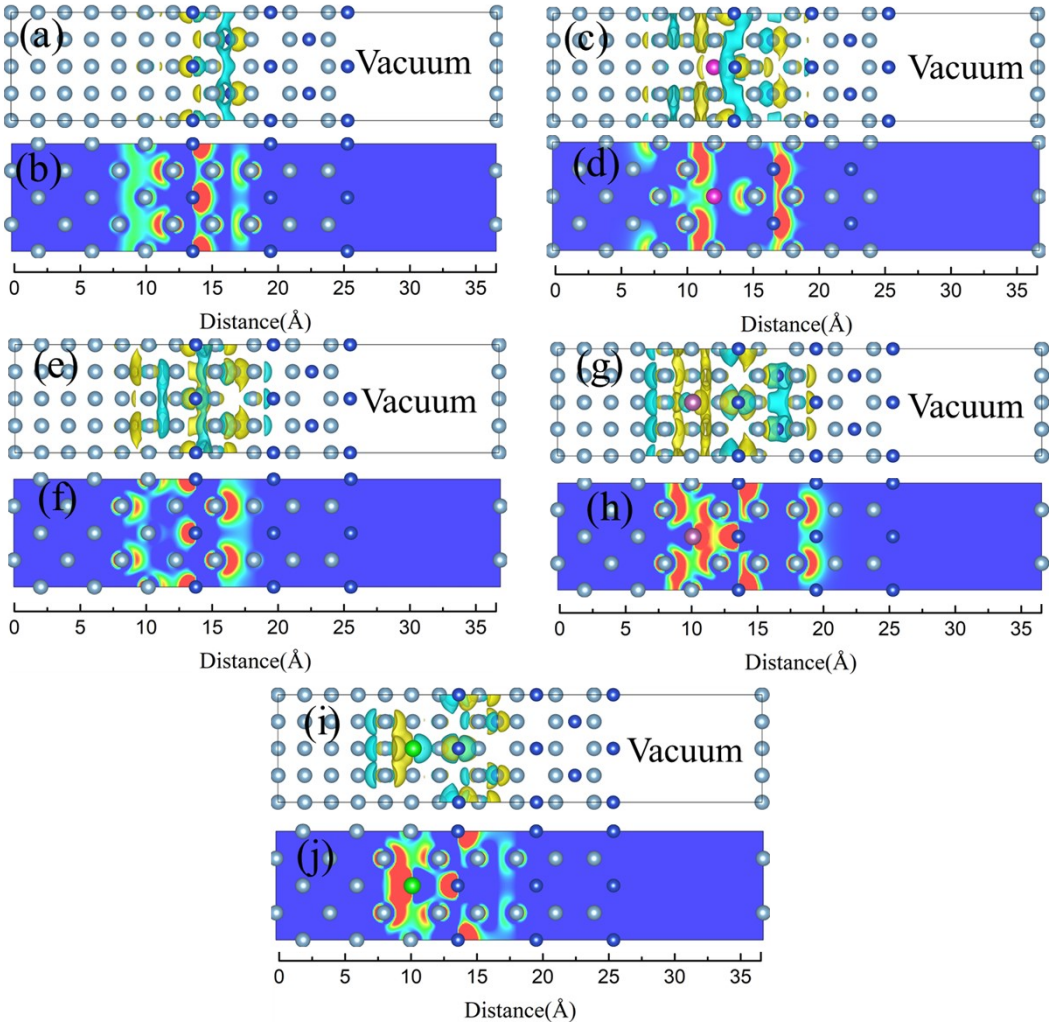

Fig. S3 Three-dimensional and two-dimensional charge density difference along (100) plan for the clean $(001)_{\alpha\text{-Al}}//(001)_{\theta'}$ and $(001)_{\alpha\text{-Al}}//(001)_{\theta'}$ interfaces with Cd, Si, Sc and Zr at sites Al_i-fc, $\theta'^{Cu}_{i\text{-}2}$, $Al_{i\text{-}1}$-cp and $Al_{i\text{-}1}$-cp. (a), (c), (e), (g) and (i) The three-dimensional charge density difference. (b), (d), (f), (h) and (j) The two-dimensional charge density difference. The light blue and blue mean the charge depletion, while yellow and red indicate the charge accumulation, respectively. The isosurface value is set to 4×10^{-6} e/Bohr^3.