

# Strengthening of High Entropy Alloys by Dilute Solute Additions: CoCrFeNiAl<sub>x</sub> and CoCrFeNiMnAl<sub>x</sub> Alloys

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## Abstract

A theory to predict the initial flow stress of an arbitrary  $N$ -component fcc random alloy is extended to predict the additional strengthening when a dilute concentration of a substitutional element is introduced. Assuming properties for the  $N$ -component alloy to be established, the theory requires only information on the elastic and lattice constants of the new  $N + 1$ -alloy, and makes a parameter-free prediction for the strength-increment due to the added  $N + 1^{\text{st}}$  element. The theory is applied to the CoCrFeNiAl<sub>x</sub> and CoCrFeNiMnAl<sub>x</sub> systems, achieving good agreement with experiments. The theory thus serves as a valuable tool for guiding design of new fcc random alloys.

*Keywords:* High Entropy Alloys, Mechanical properties, Solution Strengthening, Modeling

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High Entropy Alloys (HEAs) are nominally multicomponent, equi-composition, **single phase, random solid solution alloys** [1, 2, 3]. A number of **HEA materials having no reported short-range-order** have shown an excellent combination of mechanical properties, e.g. high strength and high fracture toughness at low temperatures. Many of such new material systems have emerged recently, ranging from the steel-like Cantor alloy (Co-Cr-Fe-Ni-Mn)[4], lightweight metals (Al-Mg-Li-Ti-Sc) [5], and noble metals (Pd-Pt-Rh-Ir-Au-Ag-Cu-Ni) [6]. The equicomposition alloys do not necessarily have the best mechanical properties, which has rejuvenated the investigation of random alloys in search of stable compositions with attractive properties. An emerging strategy is to “dope” a HEA with a dilute addition of another element to achieve higher strength or otherwise tweak the material properties [7, 8, 9, 10, 11]. Given the vast range of possible random alloys, theoretical guidance is of high value.

The present authors have recently developed a theory for the initial yield strength of random fcc alloys having arbitrary composition. The theory is a

generalization of a Labusch-type model [12, 13], and has been applied with good quantitative success to real HEAs and simulated alloys studies [14, 15, 16]. Here, we extend the model to study the “doping” of an existing HEA by dilute addition of another element, **assuming such an addition preserves the random fcc structure**. We show that the expected associated strengthening can be predicted from a minimal set of experimental information: mechanical properties of the original alloy and elastic and lattice constants estimates for the new alloys. We apply the model to predict the strengthening of the CoCrFeNiMn and CoCrFeNi fcc HEA alloys by the addition of dilute Al over the range  $c_{\text{Al}} < 0.08$  where the system remains in a single-phase fcc structure. Very good agreement with experiments is achieved, which further supports the use of this simplified model for alloy design.

The full theory for the finite- $T$ , finite-strain-rate initial yield strength of arbitrary random fcc alloys has been presented in Refs. [14, 17]. The theory treats the random alloy as a high-concentration solute-strengthened alloy in which each elemental component is a “solute” within an “effective-medium matrix” describing the average alloy properties [14, 18]. Strengthening is due to the interactions of dislocations in the effective matrix

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with the spatial concentration fluctuations of the multi-component random alloy. **The full theory includes all solute/dislocation interaction effects through the general interaction energy  $U^n(x_i, y_j)$  for a solute at position  $(x_i, y_j)$  relative to the center of the dislocation line [17]. The full theory can be reduced to an analytical model when the solute/dislocation interactions are described by elasticity, as  $U^n(x_i, y_j) = -p(x_i, y_j)\Delta V_n$  where  $\Delta V_n$  is the misfit volume of the solute  $n$  at concentration  $c_n$  in the  $N$ -component alloy and  $p(x_i, y_j)$  is the elastic pressure field generated by the dislocation into the “effective medium matrix” at position  $(x_i, y_j)$ . This elasticity model thus includes the generalized lattice misfit effect of traditional solute strengthening models, and part of the modulus effect through the use of the alloy elastic moduli at the actual alloy composition [17].** The zero-temperature yield stress  $\tau_{y0}$  and energy barrier  $\Delta E_b$  for thermally activated flow are then

$$\tau_{y0} = 0.051\alpha^{-\frac{1}{3}}K_\tau f_\tau \left[ \sum_{n=1}^N c_n \Delta V_n^2 \right]^{\frac{2}{3}}, \quad (1)$$

$$\Delta E_b = 0.274\alpha^{\frac{1}{3}}K_{\Delta E} f_{\Delta E} \left[ \sum_{n=1}^N c_n \Delta V_n^2 \right]^{\frac{1}{3}}, \quad (2)$$

with

$$K_\tau = \mu \left( \frac{1+\nu}{1-\nu} \right)^{\frac{4}{3}} b^{-4}; \quad K_{\Delta E} = \mu \left( \frac{1+\nu}{1-\nu} \right)^{\frac{2}{3}} b. \quad (3)$$

The elastic constants  $(\mu, \nu)$  and Burgers vector  $b$  are those for the  $N$ -component random alloy at the specified composition  $\{c_n\}$ .  $f_\tau$  and  $f_{\Delta E}$  are two numerical coefficients associated with the detailed dislocation core structure, as described in Ref. [14], but are nearly constant at  $f_\tau = 0.35$  and  $f_{\Delta E} = 5.7$  over a wide range of structures corresponding to stacking fault separations above  $d \approx 10b$ . The theory involves the dislocation line tension, expressed as  $\Gamma = \alpha\mu b^2$  with  $\alpha = 0.123$  obtained from various atomistic simulations [14].

At finite temperature  $T$  and finite strain-rate  $\dot{\epsilon}$ , standard thermal activation theory then leads to the predicted yield stress and activation volume as

$$\tau_y(T, \dot{\epsilon}) = \tau_{y0} \left[ 1 - \left( \frac{kT}{\Delta E_b} \ln \frac{\dot{\epsilon}_0}{\dot{\epsilon}} \right)^{\frac{2}{3}} \right], \quad (4)$$

$$V_{\text{act}}(T, \dot{\epsilon}) = \frac{3}{2} \frac{\Delta E_b}{\tau_{y0}} \left( \frac{kT}{\Delta E_b} \ln \frac{\dot{\epsilon}_0}{\dot{\epsilon}} \right)^{\frac{1}{3}}. \quad (5)$$

At temperature  $T$ , the quantities  $\tau_{y0}$  and  $\Delta E_b$  are computed using the finite-temperature elastic constants  $\mu(T)$  and  $\nu(T)$ , but this  $T$ -dependence is left implicit here. Application of this fully-analytic, parameter-free model requires information about the misfit volumes  $\Delta V_n$  of all  $N$  elements in the alloy at composition  $\{c_n\}$  and the elastic constants at the relevant temperature.

Now we envision adding an  $N+1$ <sup>th</sup> element as a dilute solute,  $c_{N+1} \ll 1$  into the  $N$ -component alloy. The new alloy has Burgers vector  $b'$  and elastic constants  $(\mu', \nu')$ . The elemental concentrations in the new alloy are then  $c'_n = c_n(1 - c_{N+1})$  for  $n \in [1, N]$ , with  $\sum_{n=1}^{N+1} c'_n = 1$ . This assumes a partition of the dilute solute according to the concentrations in the initial alloy, which - for equicomposition HEAs with  $c_n = 1/N$  - treats the new alloy as a pseudo-binary system. The new misfit volumes are denoted as  $\Delta V'_n$ . Application of the theory requires computation of  $\sum_{n=1}^{N+1} c'_n \Delta V_n'^2$ . Using Vegard's law [14] for the average atomic volume  $\bar{V}'$  of the new alloy in terms of the elemental volumes  $V_n$  leads to

$$\bar{V}' = \sum_{n=1}^{N+1} c'_n V_n; \quad \Delta V'_n = V_n - \bar{V}'. \quad (6)$$

Denoting  $\bar{V} = \sum_{n=1}^N c_n V_n$  as the volume of the original alloy and using the  $\sum_{n=1}^N c_n \Delta V_n = 0$ , we obtain

$$\begin{aligned} \sum_{n=1}^{N+1} c'_n \Delta V_n'^2 &= (1 - c_{N+1}) \sum_{n=1}^N c_n \Delta V_n^2 \\ &+ c_{N+1} \Delta V_{N+1}^2 - (\bar{V}' - \bar{V})^2. \end{aligned} \quad (7)$$

The zero-temperature flow stress and energy barrier for the new alloy can then be expressed in terms of the corresponding quantities in the original  $N$  component alloy as

$$\begin{aligned} \tau'_{y0} &= 0.051\alpha^{-\frac{1}{3}}K'_\tau f_\tau \left[ \left( \frac{\tau_{y0}}{0.051\alpha^{-\frac{1}{3}}K_\tau f_\tau} \right)^{\frac{3}{2}} \right. \\ &\quad \left. \times (1 - c_{N+1}) + c_{N+1} \Delta V_{N+1}^2 - (\bar{V}' - \bar{V})^2 \right]^{\frac{2}{3}}, \\ \Delta E'_b &= 0.274\alpha^{\frac{1}{3}}K'_{\Delta E} f_{\Delta E} \left[ \left( \frac{\Delta E_b}{0.274\alpha^{\frac{1}{3}}K_{\Delta E} f_{\Delta E}} \right)^3 \right. \\ &\quad \left. \times (1 - c_{N+1}) + c_{N+1} \Delta V_{N+1}^2 - (\bar{V}' - \bar{V})^2 \right]^{\frac{1}{3}}. \end{aligned} \quad (8)$$

The above prediction requires only measurable macroscopic quantities on both the original and new alloy. Specifically, the elastic constants and lattice parameters of the old and new alloy allow for the computation of the  $K_\tau, K_{\Delta E}, \Delta V_{N+1}$ , and  $\bar{V}'$ , and the quantities  $\tau_{y0}$  and  $\Delta E_b$  for the original alloy are accessible from either (i) the measured temperature dependence  $\tau_y(T)$  of the original alloy, or (ii) from the activation volume  $V_{act}$  plus the strength  $\tau_y$  at a given temperature.

We now apply the model above to predict the strength of  $\text{CoCrFeNiAl}_x$  and  $\text{CoCrFeNiMnAl}_x$  alloys<sup>1</sup>. The  $(\tau_{y0}, \Delta E_b)$  for the initial HEAs are obtained by fitting the temperature-dependent strengths using Eq. (4) on large-grain-size materials (to avoid Hall-Petch contributions to strength) available in [19, 20] and shown in Table 1. The theory then requires (i) the apparent reference atomic volume  $V_{Al}$  for Al that accurately predicts changes in alloy lattice constant upon addition of Al, and (ii) **the changes in elastic moduli upon addition of Al to each base alloy.**

To obtain  $V_{Al}$ , we use a Vegard's law for the atomic volume of the new alloys, *i.e.*  $V(c_{Al}) = (1 - c_{Al})\bar{V} + c_{Al}V_{Al}$ , with  $\bar{V}$  the average atomic volume in the HEA. The effective Al volume  $V_{Al}$  is then obtained as a best-fit to the experimental data on the  $\text{CoCrFeNiAl}_x$  and  $\text{CoCrFeNiMnAl}_x$  alloys. Assuming a common  $V_{Al}$  for both sets of alloys, based on the assumption that the chemical environments are similar in the two cases, gives  $V_{Al} = 14\text{\AA}^3$ . Assuming different values in each alloy yields  $V_{Al} = 13.8\text{\AA}^3$  and  $16.2\text{\AA}^3$ , for  $\text{CoCrFeNiAl}_x$  and  $\text{CoCrFeNiMnAl}_x$ , respectively. Predicted versus experimental atomic volumes for the different alloys are shown in Fig. 1a, together with previous values for equiatomic alloys in the Co-Cr-Fe-Ni-Mn family, given for comparison. A good agreement is achieved in both cases, with a dispersion which is well within what can be expected from the assumption of fixed individual elemental volumes, like for the Co-Cr-Fe-Ni-Mn alloys. The  $V_{Al}$  in  $\text{CoCrFeNiMnAl}_x$  has a rather large uncertainty with respect to the fitting procedure, partly due to both the experimental spreading and because the available alloys show larger deviations in actual composition, which is not considered here; below we make predictions using both values.

<sup>1</sup>The correspondence between  $x$  and  $c_{Al}$  is  $c_{Al} = x/(x+4)$  for  $\text{CoCrFeNiAl}_x$ , and  $c_{Al} = x/(x+5)$  for  $\text{CoCrFeNiMnAl}_x$ , respectively.

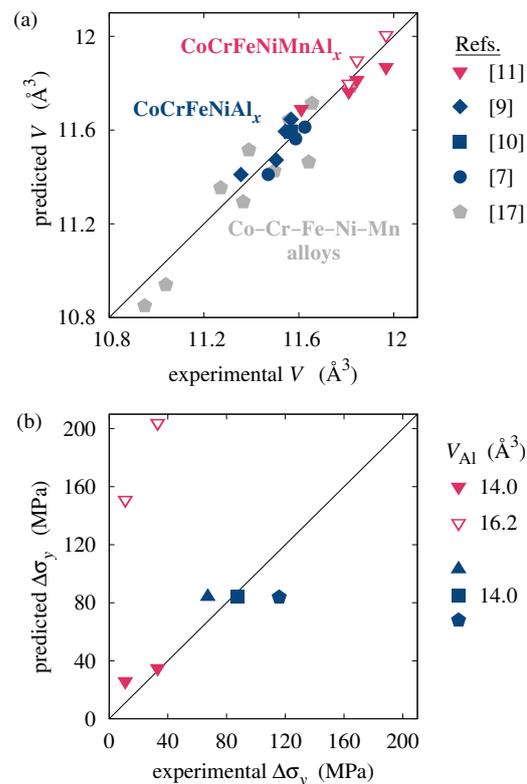


Figure 1: (a) Vegard's law predictions vs. experimental values of atomic volumes  $V(c_{Al})$  for the  $\text{CoCrFeNiAl}_x$  (blue) and  $\text{CoCrFeNiMnAl}_x$  (red) alloys. Values for alloys in the Co-Cr-Fe-Ni-Mn family (grey) are from Ref. [17]. (b) Predicted vs. experimental strength increments at  $T = 293\text{K}$  due to the Al addition in the same alloys. The predicted strength increments are converted to uniaxial tensile strengths by multiplying by the Taylor factor 3.06 for untextured polycrystals. The filled and empty symbols correspond to values obtained assuming either a unique or distinct values for  $V_{Al}$ , as indicated. Experimental strengths are from Refs [11] (red triangles), [10] (blue square: polycrystal; blue triangle: single crystal) and [24] (blue octogon).

The elastic constants for  $\text{CoCrFeNiMnAl}_x$  and  $\text{CoCrFeNiAl}_x$  have not been reported. So, we use extensive data on single-crystal elastic constants for Ni-Al solid solutions up to  $c_{Al} = 12.69\text{at.}\%$  at  $T = 300\text{K}$  [22] and *ab initio* computations [23] of the single crystal elastic constants in  $\text{CoCrFeNiAl}_x$  alloys at  $T = 0\text{K}$ . The Voigt- and Reuss-averaged shear moduli scale as  $\mu'_{\text{NiAl}} = (85\text{GPa})(1 - 1.41c_{Al})$  and  $\mu'_{\text{CoCrFeNiAl}_x} = (110\text{GPa})(1 - 1.61c_{Al})$ , and so we use  $\mu' = \mu'(T)(1 - 1.51c_{Al})$  for the HEAs with Al. For the Poisson ratios,  $\nu'_{\text{NiAl}} = 0.375 + 0.2c_{Al}$  and  $\nu'_{\text{CoCrFeNiAl}_x} = 0.275 + 0.2c_{Al}$  and so we use  $\nu' = \nu'(T) + 0.2c_{Al}$ . The base HEA values  $\mu$  and  $\nu$

are obtained from experiments [19]. The estimated elastic constants for specific Al contents are given in Table 1.

The predicted strengthening by the addition of Al “solutes” to the HEAs, using the above inputs and  $V_{\text{Al}} = 14 \text{ \AA}^3$ , is compared to experimental values [10, 11, 19, 20, 24] at  $T = 293 \text{ K}$  in Fig. 1b. Fig. 1b also shows our prediction, using the average Al volume  $V_{\text{Al}} = 14 \text{ \AA}^3$ , for the strength increment of an independently-measured single-crystal yield strength in  $\text{CoFeCrNiAl}_{0.3}$ , where we have converted to an effective uniaxial tensile strength by multiplying by the Taylor factor 3.06. The agreement is very good, with no adjustable parameters in the theory. Slight deviation for one sample [24] is associated with the exact composition that is unknown, and with traces of bcc phase ( $\sim 0.1\%$ ). The theory demonstrates the larger strengthening of Al solutes in  $\text{CoFeCrNiAl}_x$  alloys as compared to  $\text{CoFeCrNiMnAl}_x$  alloys, consistent with experiments. The strength increment in both cases results from a competition between the relatively large Al misfit volumes in the HEAs and the elastic softening accompanying Al additions. The elastic softening appears small, but multiplies the entire strength of the base alloy, because the base solute elements are all now embedded in a softer elastic matrix. Inclusion of both misfit and elastic constant effects is therefore essential in making quantitative contact with experiments. Strength predictions obtained using the alloy-specific  $V_{\text{Al}}$  are also shown in Fig. 1b. Similar results are obtained for  $\text{CoFeCrNiAl}_x$  (thus not shown), but large differences are found for  $\text{CoFeCrNiMnAl}_x$  alloys. The latter difference is in part due to deviations from the ideal pseudo-binary composition, which can be rectified by using the full theory with all misfit volumes and concentrations of all elements in the alloy, and to the spreading in lattice parameter measurements. This reinforces the need for a careful experimental characterization of alloy samples, with respect to lattice constants and chemical composition. Refined predictions can also easily be made using experimental elastic constants and more detailed measurements of lattice constants vs.  $c_{\text{Al}}$  once available.

We have demonstrated that it is possible to predict the additional “solute strengthening” of HEAs due to dilute additions of another solute, **assuming the new materials retain the random fcc structure**. Note that “strengthening” is not assured - the dilute solute addition could (i) decrease the

overall misfit volumes of all the solutes in the alloy and/or (ii) decrease the alloy elastic constants. Addition of large-volume solutes likely involves an interplay of these effects, since elements with large atomic volumes tend to be elastically softer. In contrast, additions of small-volume solutes might provide strengthening due to both factors. For the present HEAs, however, there are no obvious candidate solutes since Ni and Co are already quite “small” solutes with fairly high elastic moduli.

The present theory enables design, *i.e.* estimation of the effect of small additions of *any* substitutional solute to pre-existing fcc HEAs, with some simplifications. First, elastic constants could be estimated using a rule-of-mixtures based on the elemental (isotropic polycrystalline) elastic constants  $E_n$ ,  $G_n$ , and  $\nu_n$ , e.g.  $E_{\text{alloy}} = \sum_n c_n E_n$ ,  $G_{\text{alloy}} = \sum_n c_n G_n$ , and  $\nu_n = E_n/2G_n - 1$ . Second, as done here, Vegard’s law can be used to estimate apparent alloy atomic volumes using experimental lattice parameters of the new alloys. Uncertainty associated with both of these inputs can then also be assessed to determine whether candidate alloys and properties are robust with respect to the estimates. **The present model assumes single phase fcc random alloys, thus neglecting possible short range ordering effects and/or transformations to multiphase materials, both driven by alloy-specific thermodynamic and requiring separate assessment. Nonetheless, the model remains applicable to a wide range of multi-component substitutional fcc alloys, including transition metals, noble metals and lightweight metals where such systems have already been fabricated [5, 6, 25].** Thus, the analytic theory here provides a fairly simple but powerful *framework*, within a set of reasonable and partially-validated assumptions, for analytic estimates of the initial yield strength increment due to small solute additions to fcc HEAs. This makes it a valuable asset for experimental metallurgists seeking guidelines for design of stronger alloys and for interpreting experimental results.

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Table 1: Elastic constants, Burgers vectors, zero-temperature flow stress and energy barriers, vs.  $c_{Al}$  for  $CoCrFeNiMnAl_x$  and  $CoCrFeNiAl_x$  alloys. Values in italic font are from or deduced from experiments [19, 20]. Elastic constants for finite Al content are estimated from available literature data (see main text). Model-prediction of  $\tau_{y0}$  and  $\Delta E_b$  for finite  $c_{Al}$  are obtained with the common  $V_{Al} = 14\text{\AA}^3$ , and with the distinct  $V_{Al} = 13.8$  and  $16.3\text{\AA}^3$  (values in parenthesis).

Alloy	$c_{Al}$ (at%)	$\mu$ (GPa)	$\nu$	$b$ (Å)	$\tau_{y0}$ (MPa)	$\Delta E_b$ (eV)
$CoCrFeNiAl_x$	0	<i>84</i>	<i>0.280</i>	<i>2.527</i>	<i>101.9</i>	<i>0.9</i>
	7.1	75	0.294	<i>2.540</i>	134.0 (131.8)	1.16 (1.17)
$CoCrFeNiMnAl_x$	0	<i>80</i>	<i>0.260</i>	<i>2.544</i>	<i>117.7</i>	<i>1.05</i>
	4.6	74.4	0.269	<i>2.559</i>	128.4 (181.4)	1.14 (1.46)
	7.2	71.3	0.274	<i>2.568</i>	132.2 (204.7)	1.16 (1.56)

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