Effects of Chemical Short-Range Order on the Percolation Model of Passivation for Binary Alloys

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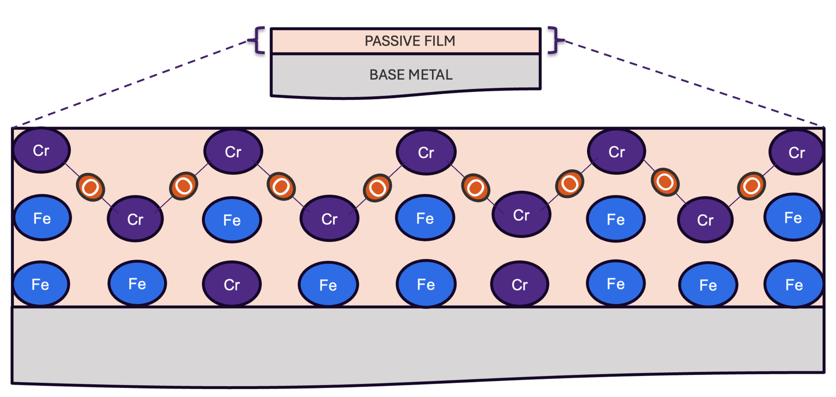
Our model reveals the mechanism of protective passive film formation in metallic alloys, paving the way for the design of advanced corrosion-resistant materials.

Introduction

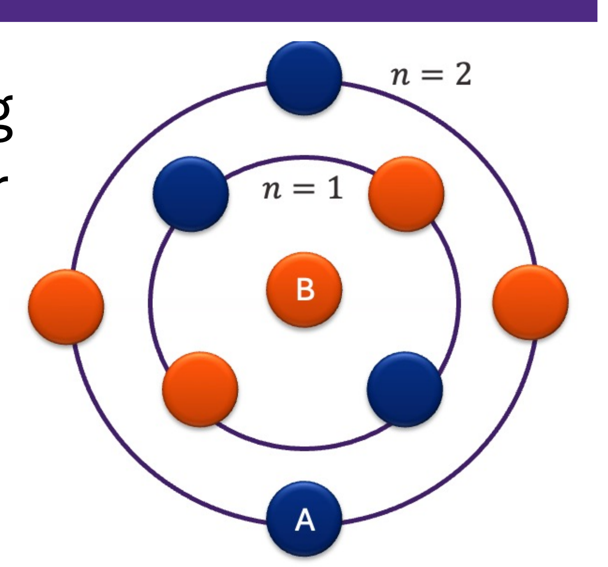
- Corrosion-resistant metallic alloys, such as stainless steel, survive aggressive environments owing to their ability to form nanometer-scale, self-healing, protective passive oxide films.
- We quantify the chemical short-range order (SRO) effects on percolation in a practical binary Cu-Rh alloy and study the related passivation behavior.
- This knowledge can inform the design of the next generation of corrosion-resistant metallic alloys.



Methods

(1) Computer-generated lattice using Warren-Cowley short-range order parameter:

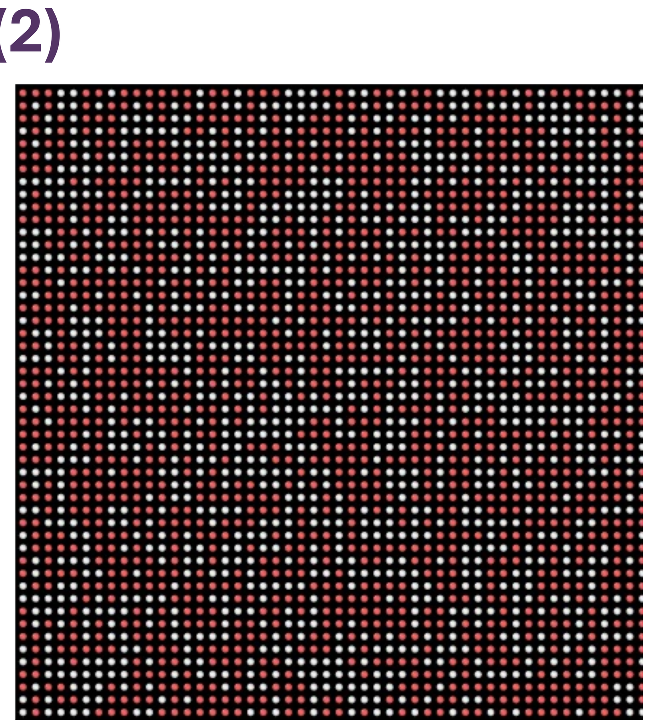
$$\alpha^{(n)}(\chi, T) = 1 - \frac{p_{AB}^{(n)}}{\chi_A}$$



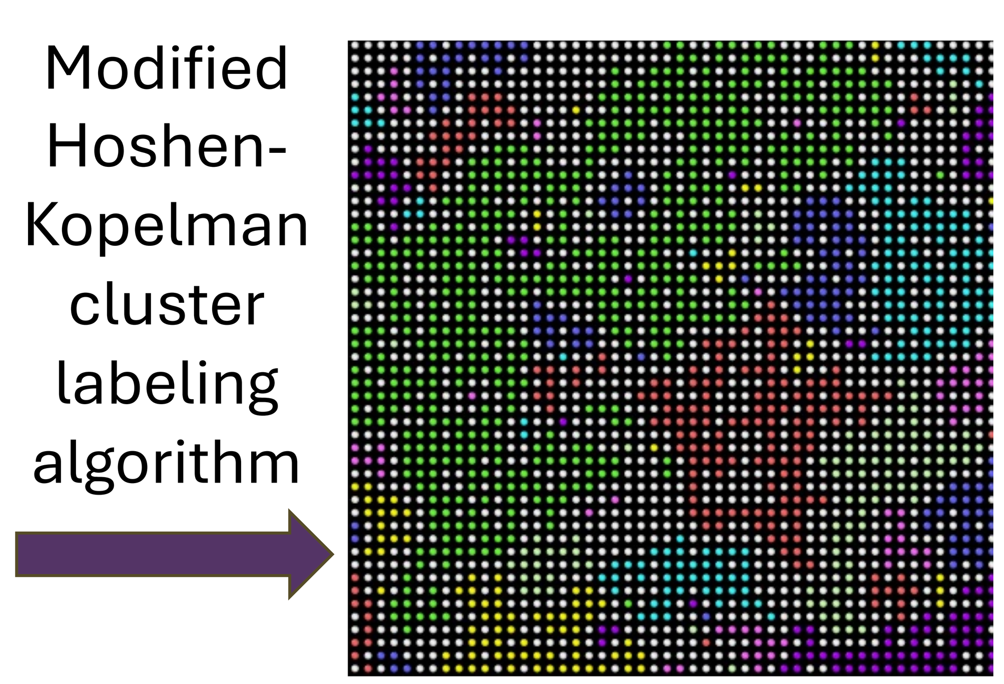
Short-range ordering ($\alpha < 0$): Predominance of A-B bonds

Short-range clustering ($\alpha > 0$): Predominance of A-A and B-B bonds

(2)

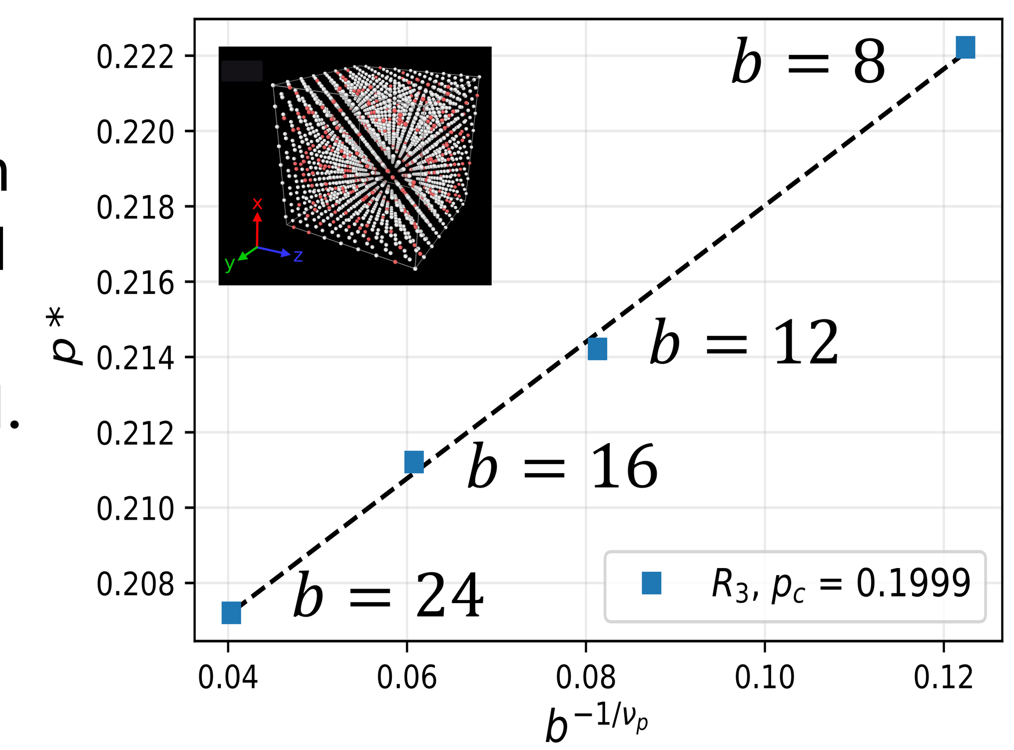


Modified Hoshen-Kopelman cluster labeling algorithm



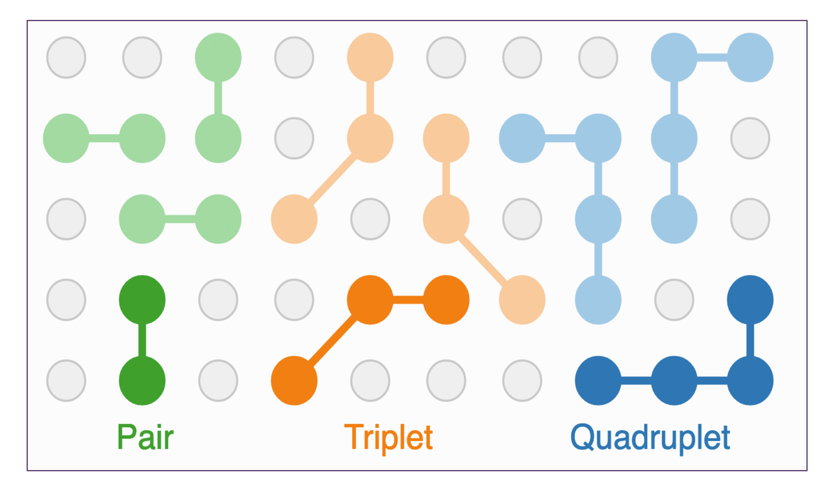
(3) The large cell Monte Carlo renormalization group method is used to calculate the percolation threshold.

$$p^* \propto b^{-1/\nu_p}$$



(4) Cluster expansion model for Cu-Rh alloy trained on DFT mixing energy data.

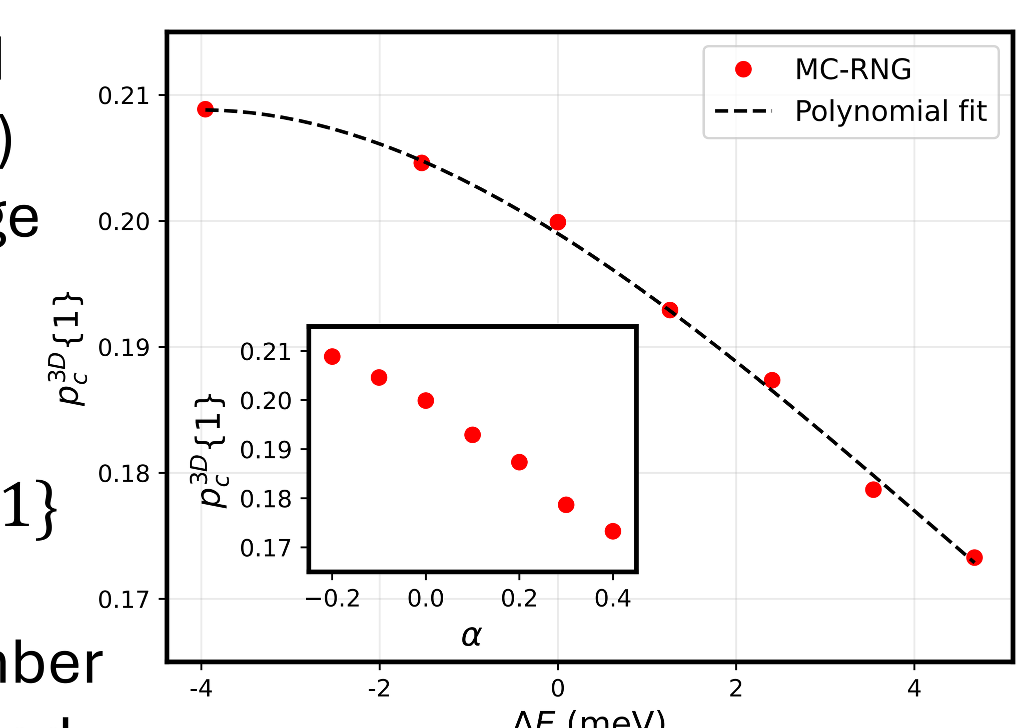
$$E(\sigma) = \sum_{\alpha} m_\alpha J_\alpha \langle \Gamma_{\alpha'}(\sigma) \rangle_\alpha$$



Results

(1) Percolation threshold (1st nearest neighbor) varies with short-range order in the alloy.

$$p_c^{3D}(\alpha) = \gamma_1 \alpha + \gamma_2 \alpha^2 + \gamma_3 \alpha^3 + p_c^{3D}\{1\}$$



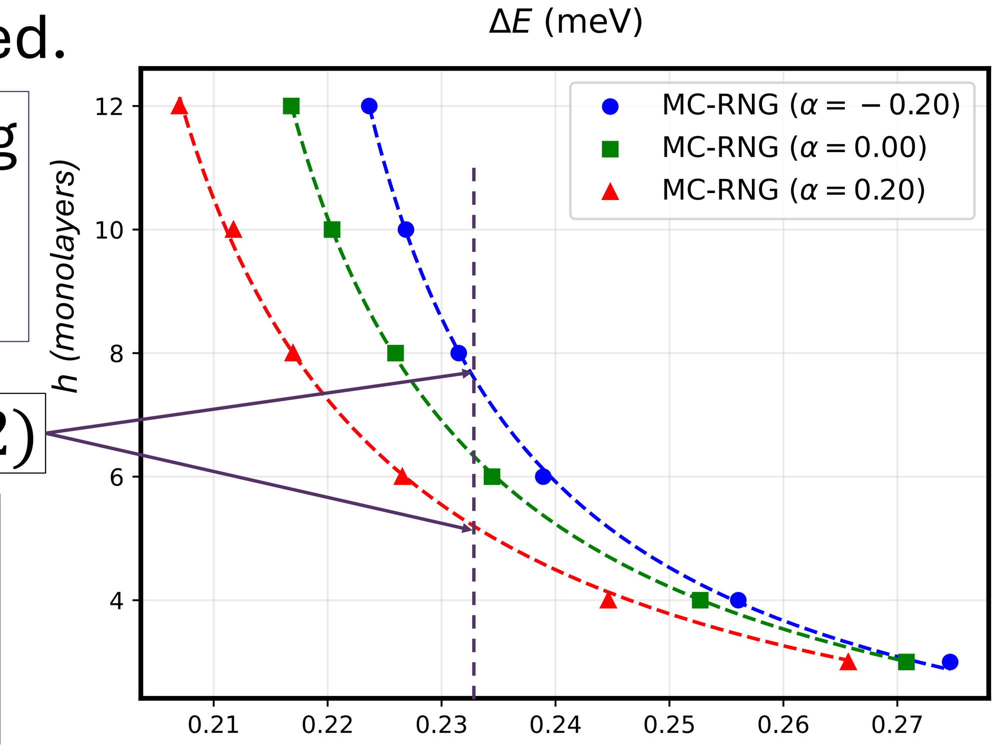
(2) h: The equivalent number of monolayers dissolved.

Short-range clustering promotes corrosion resistance.

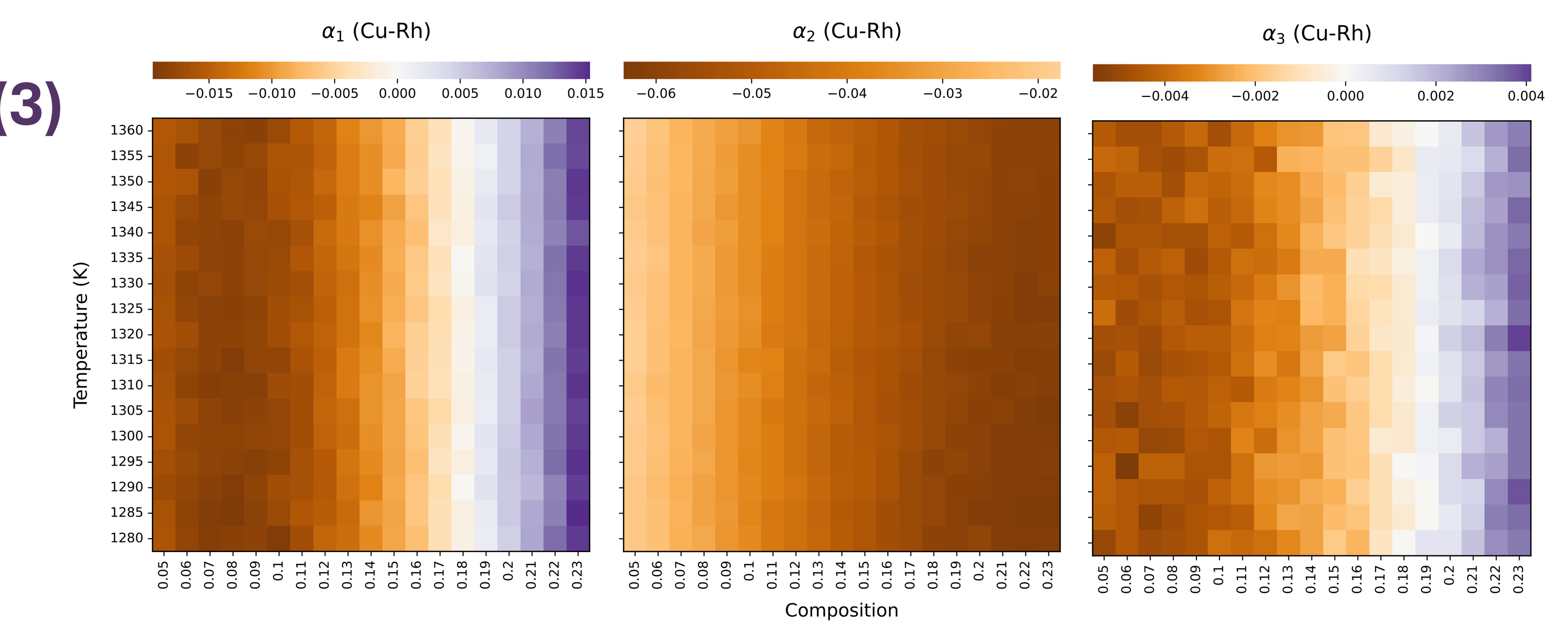
$$h(\alpha = 0.2) < h(\alpha = -0.2)$$

The percolation crossover model captures SRO.

$$h(\alpha) = \delta[p_c(h, \alpha) - p_c^{3D}(\alpha)]^{-\nu_{3D}}$$

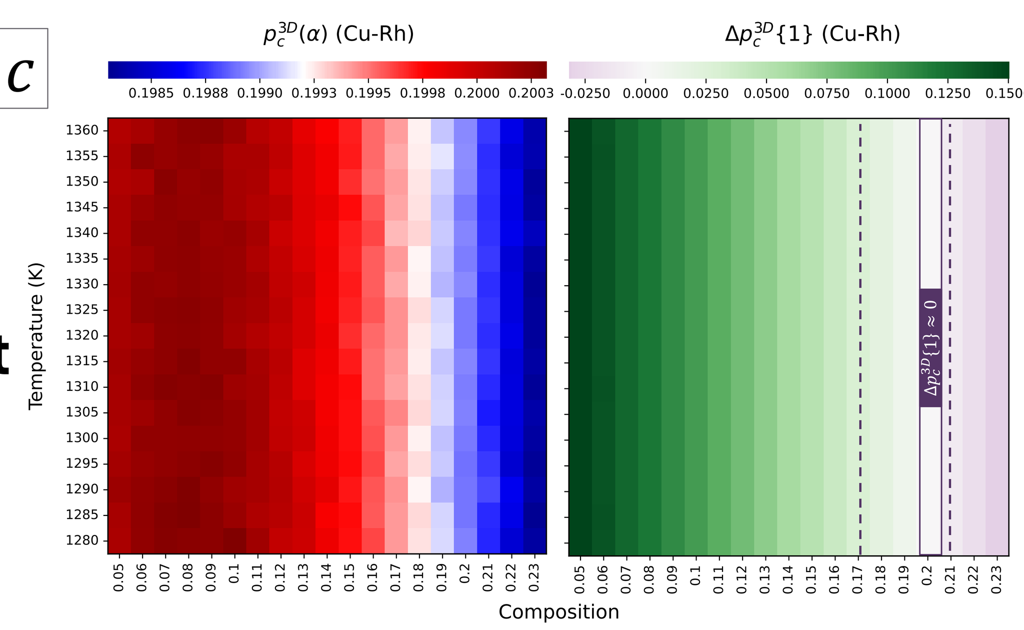


(3)



$$\Delta p_c^{3D}\{1\} = p_c^{3D}(\alpha) - c$$

The "critical region" $\Delta p_c^{3D}\{1\} \approx 0$ **should be shifted to the left** to design corrosion-resistant alloys.



Conclusions & Next Steps

- The percolation crossover model captures ***the effects of SRO on primary passivation.***
- Short-range clustering ***promotes corrosion resistance.***
- Percolation threshold difference maps can be used to determine the ***"critical region" that should be shifted to the left*** to design corrosion-resistant alloys.
- We must consider the ***effects of coherency strain energy*** and ***magnetism on SRO*** in lattice-mismatched magnetic alloys.

References & Acknowledgement

1. Roy et al. *Physical Review B* 110.8 (2024): 085420.
2. Xie et al. Nature materials 20.6 (2021): 789-793.
3. Sotta et al. The European Physical Journal E 11.4 (2003): 375-388.

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