

Abstract Submitted
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First Principles Study of Bulk and Surface Ordering Phenomena in Pt-X Binary Alloys in the Presence of Oxygen WEI CHEN, CHRIS WOLVERTON, Northwestern University, Department of Materials Science and Engineering, WILLIAM SCHNEIDER, University of Notre Dame, Department of Chemical and Biomolecular Engineering, Department of Chemistry and Biochemistry — Alloying in metal catalyst particles (such as platinum) may change surface structure due to segregation and ordering, which may in turn significantly impact catalytic activity. Using first principles density functional theory (DFT) calculations in conjunction with a cluster expansion (CE) technique, we have studied the ordering/phase-separation phenomena of bulk and surface Pt-X binary alloys, with a specific focus on Pt-Au. The surface DFT+CE calculations are performed both in the presence and absence of oxygen. For Pt-Au, the calculated results reveal a phase separating tendency in bulk Pt-Au and a small coherency energy between these two elements. The Pt-Au phase diagram calculated by combining DFT+CE with Monte Carlo simulation shows a slightly asymmetric miscibility gap is in good agreement with experimental results. In contrast to the bulk tendency, the surface does not show a pronounced phase-separation tendency, with several low-energy “striped” ordered structures (with a very small, negative formation energy). The presence of oxygen qualitatively changes the surface segregation tendency of Au, and our DFT+CE results show that the equilibrium structure of the Pt-Au (111) surface varies with oxygen coverage.

Wei Chen
Northwestern University, Department of Materials Science and Engineering

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