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Using Site Occupation Disorder and Surface Energies to Determine Pt3Pd2 and Pt2Pd3 Electrocatalyst Models

A potential means of energy production is the hybrid sulphur (HyS) cycle [1]. The HyS cycle is a two-step water-splitting process, which is used to produce hydrogen, with no harmful by-products that are typically produced from hydrocarbon fuels. The HyS cycle oxidises aqueous SO2 to produce hydrogen and H2SO4, with the H2SO4, then thermally decomposed back to SO2. The oxidation can be promoted by use of an electrocatalyst, typically a platinum catalyst.[2] Research are conducted at North-West University [3] to improve these electrocatalysts, by developing multi-metal thin film catalysts. The best preforming catalysts were Pt3Pd2 and Pt2Pd3, which had the same performance as the standard platinum catalyst, but had increased stability, chemical resistance. Thus in order to understand the surface properties of these catalysts, computational models of these catalysts were constructed using Sutton-Chen interatomic potentials. The program, Site-Occupation Disorder (SOD),[4] was used to prepare all possible bulk combinations of Pt3Pd2 and Pt2Pd3 catalysts, and the configurations were optimised using General Utility Lattice Program (GULP).[5] Each catalyst had 101 possible combinations, however at a thermal annealing temperature of 800℃ [6] all combinations were possible and had to be taken into account. Thus to find optimum configurations, surface energies were calculated, using the two-region method, for 101 configurations of both catalysts. The bulks were cut to expose the (111) surfaces using the program METADISE [7] (minimum energy techniques applied to dislocation interface and surface energies). Figure 1 shows the calculated surface energies for both Pt3Pd2 and Pt2Pd3 catalysts. The surface energies of Pt3Pd2 catalyst indicated that one configuration, namely configuration 3, had a notably lower surface energy (0.817 J.m-2) than the rest. The Pt2Pd3 system had three configurations with lower than average energy, namely configurations 17, 81 and 8 with energies calculated at 0.887 J.m-2, 0.888 J.m- 2 and 0.889 J.m-2 respectively. These configurations will be used in future studies as the representative surfaces of these catalysts. This study was made possible thanks to the computational resources obtained from the Centre for High Performance Computing (CHPC), which provided both hardware and software used in this study.

a)
b)
Figure 1: Surface energies for the 101 configurations of the (a) Pt3Pd2 and (b) Pt2Pd3 catalysts

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