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Reactivity of H2O and SOx (x = 0 – 3) on a Pt (111) surface – A DFT Study

The current global energy demand is met by burning fossil fuels, which releases large amounts of carbon dioxide, contributing to climate change. Hydrogen is considered a sustainable energy carrier, when it is produced from renewable sources. An attractive technology currently receiving attention is the Hybrid Sulphur (HyS) cycle, which is a thermo-electrochemical process where sulphuric acid (H2SO4) is thermally decomposed (> 800℃) to produce sulphur dioxide (SO2), water (H2O) and oxygen (O2). In a second step SO2 reacts with H2O to form H2SO4 and H2. The net reaction of this cycle is the splitting of H2O into H2 and O2. The current catalyst of choice is platinum (Pt), a very expensive and rare noble metal. While various other metals have been investigated, Pt is still the best performing catalyst in terms of activity and stability.1

The SO2 oxidation mechanism on the Pt surfaces is shown in Figure 1, where the adsorption modes of SO2 and H2O was determined. The most stable adsorption mode for SO2 on the Pt (111) surface had a S,O-bonded geometry, where S-O were in the plane of the surface on the fcc binding site. For H2O, the most stable adsorption occurred when all the atoms were parallel to the surface2. The second part of this work will cover the effect of increasing the SO2 and H2O coverage on the surface, in terms of surface morphology and reactivity. This will be concluded with a discussion on the behaviour of the other SOx species on the Pt surface.

Figure 1. Wulff morphology of a Pt nanoparticle, showing the structures and adsorption sites on the Pt (111) surfaces along with the surface coverages of H2O and SO2.

In this paper, we have used density functional theory (DFT) calculations3 to predict the behaviour of SO2 and H2O on the Pt (111) surface. To that regard, we aim to develop a comprehensive understanding of the SOx chemistry, including the adsorption, desorption, oxidation, as well as side reactions to determine the species that may occur on the major electro-catalytic surfaces of Pt. This work was made possible by the CHPC (Centre for High Performance Computing)4 and Supercomputing Wales5 were 16.6 years of CPU time was used.

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