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Reactivity of H₂O and SO_x (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°C) 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 surface.² 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 SO_x 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 SO_x 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 where 16.6 years of CPU time was used.

References

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