

CHAPTER 5

CONCLUSION

5.1 Conclusions

The aim of this study was to establish kinetic data and determine whether a $\text{Pt}/\text{M}_x\text{O}_y\text{-C}$ could be applied for SO_2 oxidation in the SDE. Oxidation of SO_2 took place at the RDE and H_2 formed at the counter electrode.

5.1.1 Transmission electron microscopy

The images gathered from the TEM showed the particle distribution of platinum and metal oxides (ZrO_2 , Ta_2O_5 and Nb_2O_5) on carbon. Though ZrO_2 , Ta_2O_5 and Nb_2O_5 were co-supports in the prepared catalysts, the platinum particles were shown to have been more prone to attach to the carbon support in the bulk material of $\text{Pt}/\text{ZrO}_2\text{-C}$, whereas in $\text{Pt}/\text{Ta}_2\text{O}_5\text{-C}$, the Pt particles appeared more frequently around the Ta_2O_5 support than on carbon. The selection of magnification was fundamental to the information that could be extracted. The agglomeration of metal oxides may be due to the nature of the original material and the preparation method used for these catalysts. Crystallisation of Pt particles was likely to be due to chemical reduction (in the preparation method).

5.1.2 X-ray diffraction

With the use of XRD, we were able to detect the existence of different phases in ZrO_2 , Ta_2O_5 and Nb_2O_5 . It also disclosed the coexistence of Pt and metal oxides because there was no shift in the diffraction peaks hence any change in crystalline lattice. The $\text{Pt}/\text{Ta}_2\text{O}_5\text{-C}$ catalyst had the largest crystal size. However, the smaller crystal size of $\text{Pt}/\text{ZrO}_2\text{-C}$ did not result in superior performance when compared all other prepared catalysts.

5.1.3 Electrochemical active surface area

The loss of electrochemical active surface area caused severe performance degradation. The $\text{Pt}/\text{ZrO}_2\text{-C}$ had a higher EAS than $\text{Pt}/\text{Ta}_2\text{O}_5\text{-C}$, $\text{Pt}/\text{Nb}_2\text{O}_5\text{-C}$ and Pt/C . The high EAS of $\text{Pt}/\text{ZrO}_2\text{-C}$ can be explained by the low crystal size. However, after a

series of linear polarisation scans Pt/ZrO₂-C experienced a much greater area loss than all other catalysts.

5.1.4 Catalyst activity

The linear polarisation scans for each of the catalysts revealed that the influence of temperature and sulphuric acid concentration were positive, meaning the current density increased with the increase of temperature and sulphuric acid concentration. The Levich and Koutecky-Levich plots revealed that the SO₂ oxidation was a multistep reaction on all the prepared catalysts and that there were regions which were kinetic and diffusion controlled and others which were diffusion controlled only.

5.1.5 Catalyst stability

Catalyst stability tests were not performed for this study. However, conclusions were drawn from electrochemical active surface area studies and Pt dissolution. Pt dissolution results from ICP-OES (iCAP 6000 series Thermo scientific Duo utilising iTEVA) and the calculation made from EAS prior to and after the linear polarisation scans showed gradual loss of the Pt area and detectable Pt concentration. The result indicated that the Pt/ZrO₂-C catalysts showed the least favourable results.

5.1.6 Main conclusion

Pt/Ta₂O₅-C catalysts showed superior catalytic activity and stability amongst, the Pt/Nb₂O₅-C, Pt/ZrO₂-C and Pt/C. The Pt/ZrO₂-C showed the most inferior catalytic activity and stability.

5.2 Recommendations

- There seems to be a minimal effect of additions of tantalum pentoxide, niobium pentoxide and zirconium dioxide on the catalytic activity of Pt/C. Different additions of the metal oxides need to be investigated to determine the influence of different metal oxides on the catalytic activity.
- Alloying platinum with zirconium, tantalum and niobium in the metallic state can be further investigated.

- The catalyst preparation method, reduction temperature and catalyst loading need to be further investigated to optimise the electrode surface area for better catalytic activity in the electrolyser.
- A study in increased acid concentration (from 50 to 70 wt %) and elevated temperatures (80-100°C) should be done in order to emulate the electrochemical conditions in the SDE to test these prepared electrocatalysts. Higher molar concentrations of H_2SO_4 i.e. greater than 1.5M should also be investigated as it is known that SO_2 solubility decrease with increased acid concentration.