Mass transport in molten alkali carbonate mixtures

Andreas Bodén and Göran Lindbergh

Department of Chemical Engineering and Technology, Applied Electrochemistry, Royal Institute of Technology, KTH, SE-100 44, Stockholm, Sweden

INTRODUCTION

The Molten Carbonate Fuel Cell is a well-developed fuel cell technology for converting chemical energy to electrical energy. The major limiting factors for longer lifetime are corrosion, nickel shorting and electrolyte losses. All of these are depending on the local composition of the electrolyte. The local composition of the electrolyte can be calculated using the laws of transport phenomena. Pollard and Newman [1] derived transport equations for molten salt in porous electrodes based on the theory for multicomponent diffusion and concentrated electrolytes. A set of important parameters is the transport coefficients for binary interactions between two ions. Tanase et al [2] have made conductivity measurements for binary combinations of Li$_2$CO$_3$, Na$_2$CO$_3$, and K$_2$CO$_3$ salts in a wide range of temperatures. Spedding [3] has made density measurements for the same combinations. In this work, binary interaction coefficients and transport parameters have been calculated for three binary salts using conductivity and density data.

MODEL

To be able to calculate the interaction coefficients from conductivity data a model for the conductivity based on composition, temperature and binary interaction coefficients was derived using the theory for multicomponent diffusion and transport phenomena. The interaction coefficients are assumed to have Arrhenius temperature dependency and to be independent of composition. For each composition and temperature a relative error can be calculated between the measured and the calculated conductivity. The interaction coefficients were calculated using the measured data [2,3] by minimising the sum of all the relative errors. The modeling work was done in MATLAB®.

RESULTS AND DISCUSSION

Six binary interaction coefficients were obtained using the procedure described above. From the calculated values of the interaction coefficients, the following relationship was found: Li$^+$CO$_3^{2-}$ > Na$^+$CO$_3^{2-}$ > K$^+$CO$_3^{2-}$. When comparing the measured and the calculated conductivities it could be seen that the largest deviations were for the Li$_2$CO$_3$/K$_2$CO$_3$ system whilst the smallest were for K$_2$CO$_3$/Na$_2$CO$_3$. For the Li$_2$CO$_3$/Na$_2$CO$_3$ system, the calculated conductivity as a function of both temperature and composition are shown in figure 1. Relative errors for the calculated conductivity using the binary interaction coefficients are shown in figure 2. These results will be used to calculate concentration polarization and the local electrolyte composition for MCFC using the theory from [1].

CONCLUSIONS

The model developed, based on multicomponent diffusion for calculating the conductivity, shows good agreement with measured values in the whole composition and temperature range for all three binary salts.

ACKNOWLEDGEMENTS

The Swedish Energy Agency is acknowledged for financing this study.

REFERENCES