CO_2 reduction under high pressure – Investigation of the solubility of CO_2 and H_2O in electrolyte mixtures and their conductivity

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The electrification of chemistry is an important research area to contribute to the achievement of climate goals. For this purpose, the electrochemical conversion of carbon dioxide seems to be a promising process. Short-chain alcohols, which can be used as commodity chemicals or fuels, can be synthesized electrocatalytically by using carbon dioxide and water. However, two major limiting factors have been identified in past studies.

1. The reaction at ambient pressure mainly produces hydrogen, and
2. the product yield per time is very low.

The first problem can be explained by the low solubility of carbon dioxide in water. Due to the low solubility the mass transport is limited. Therefore, only a few CO_2-molecules interact with the electrode, which results in a limiting carbon dioxide reduction (CO_2RR) and a promotion of hydrogen evolution (HER). To improve the solubility, organic solvents can be applied, as they are miscible with both carbon dioxide and water. [1] Further investigations showed that the solubility of carbon dioxide is still too low even with the addition of organic solvents at ambient pressure. [2] For further improvement, carbon dioxide can be compressed to supercritical CO_2 (scCO_2). Previous work also shows that scCO_2 can shift the product spectrum towards organic components. [2,3]
The other limiting factor is the product yield per time, which depends to a large extent on the current density during the reaction. The current density can generally be increased by smaller electrode distances, higher voltages or higher electrical conductivities of the reaction mixture. However, the voltage is not variable due to the reaction and the distance between the electrodes cannot be chosen infinitely small for constructional reasons. Therefore, the electrical conductivity of the reaction mixture is another decisive factor. Due to the low dielectric constant of carbon dioxide, it is also necessary to use a conducting salt, which must have special properties due to the low polarity of CO_2.

While the solubility of the system H_2O/CO_2 is well documented, the influence of conducting salts together with organic solvents on the phase behavior of the reactants is largely unknown. In this work, first investigations on the phase behavior of conducting salts and solvents with the reactants H_2O and CO_2 are presented. Furthermore, the conductivities of these mixtures are investigated.

References

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