Thermoelectric Properties of Molten Carbonate Mixtures

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A thermoelectric energy converter is able to convert excess heat from various sources at various temperatures to electric energy. The efficiency of this conversion is low, and research has so far focused on the use of semiconductor materials. Seebeck coefficients of these materials amount to 0.3 mV/K. Theoretical descriptions of the thermoelectric cell [1] predict that molten salts (ionic liquids) and gas electrodes can increase this value several times. We report measurements of Seebeck coefficients for molten alkali carbonate mixtures with carbon dioxide and oxygen gas on gold or platinum electrodes, confirming a first study from 1977 of this cell [2]. Results are presented for pure (Li2CO3) and mixed carbonates (e.g. Li2CO3, K2CO3(l)) at stationary state, in the absence and presence of MgO(s) and in the temperature range 550 - 750 C. Data are reduced according to the theory of non-equilibrium thermodynamics for heterogeneous systems. The total gas pressure was 1 bar. The composition of oxygen, carbon dioxide and helium were systematically varied and shown to confirm the theoretically expected stoichiometry of the electrode reaction. The Seebeck coefficient varied between 1.2 and 1.4 mV/K depending on the electrolyte composition; the highest values were typical for mixtures of alkaline carbonates. The temperature dependence of the coefficient was negligible, as expected at the high temperature used. The transported entropy of the carbonate ion of in pure molten Li2CO3 was calculated to 281 ± 3 J/mol.K, increasing to 297 ± 1 J/mol.K when the melt was saturated with MgO(s). This value is more than one order of magnitude larger than the transported entropy in a semiconductor. The data enable us to prescribe operating conditions for thermoelectric converters with higher Seebeck coefficients. This work supports the idea that the technology could benefit from systematic studies of complex-formers and gas reactions in the electrolyte.

References

[1] S. Kjelstrup and D. Bedeaux, Non-equilibrium thermodynamics of heterogeneous systems, World Scientific, 2008
[2] T. Jakobsen and G.H.J Broers, J. Electrochem. Soc. **124** (1977) 210-214.