Including SO_3 into the HotVeGas Oxide Database

ELENA YAZHENSKIKH[1], TATJANA JANTZEN[2], KLAUS HACK[2], MICHAEL MÜLLER[1]

[1]*Institute of Energy and Climate Research, Microstructure and Properties of Materials (IEK-2), Forschungszentrum Jülich, Jülich, 52425, Germany*
[2]*GTT-Technologies, Kaiserstr. 100, Herzogenrath, D-52134, Germany*

ABSTRACT

Complex oxide systems containing sulphur along with silica, alumina, alkali-earth and alkali oxides are important in many scientific and industrial fields. Thermodynamic properties of such systems for which the measurements are experimentally difficult can be described and predicted by thermodynamic modelling on the basis of reliable experimental data and appropriate Gibbs energy models for various phases. In the framework of the HotVeGas project the oxide database containing SiO_2, Al_2O_3, alkali, earth-alkali, iron oxides, MnO_x has been created. As a sulphur-containing species, SO_3 is considered as the next important slag component to be added into the database, because sulphates widely occurre in nature as well as in industrial processes. For instance, the presence of S in coals and biomass leads to formation of deposits on heat exchange surfaces containing appreciable amounts of alkali metal sulphates and pyrosulphates, which could destroy the normally protective oxide on the metal surface. A knowledge of the phase equilibria in the system is important for process control and simulation, particularly a knowledge of the liquidus temperatures and the solubility limits of sulphates.

The aim of the present work is the development of a database for the slag relevant oxide system containing SO_3 for the modelling of a complete coal ash (slag) and gas system. These systems should be studied depending on temperature and composition, the latter being directly related to the partial pressure of SO_3.

In the presentation it will be shown that similar to the phosphates which have been incorporated into the database as compounds between the oxides and P_2O_5, the sulphates can be handled as compounds between the oxides and SO_3.

The Gibbs energy of the liquid phase has been modelled using a non-ideal associate solution approach. The Gibbs energy of the stoichiometric compounds have been presented in form of a simple G(T) function. The solid solutions have been described using the sublattice approach.

Firstly, the corresponding M_xO_y-SO_3 systems (M=Na, K, Ca, Mg) have been considered, then the phase equilibria between Na, K, Ca and Mg sulphates are studied. The DTA-measurements which have been carried out in the lab of IKE2, FZJ, were also used for the assessment. In the next step, ternary systems containing the different sulphates can be calculated.

The solubilities among alkali sulphates as well as those between alkali and alkali- earth sulphates are also incorporated into the database. Solid solutions based on Na_2SO_4 and K_2SO_4 are described using the sublattice model, and the corresponding solid solution parameters have been found to give adequate representations of the available experimental data. Generally, the thermodynamic assessment of further systems with SO_3 is continued, and alkali as well as alkali- earth sulphates have to be taken into account.