Thermodynamic Optimization of the Systems K_2O-Al_2O_3-SiO_2 and Na_2O-Al_2O_3-SiO_2

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ABSTRACT

Complex oxide systems containing high amounts of silica and alumina and alkali oxides are important in many scientific and industrial fields, e.g. in coal combustion and gasification processes where alkali release and behaviour of slags are among the main problems. 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 the various phases. The available databases are not sufficient to model the complete ash (slag) and gas systems. Therefore, a new database for the slag relevant oxide system is necessary. The present assessment continues our work on the development of a database for the system containing silica, alumina and alkali oxides.

In the present work, two ternary systems K_2O-Al_2O_3-SiO_2 and Na_2O-Al_2O_3-SiO_2 are considered. The binary Alk_2O-SiO_2, Alk_2O-Al_2O_3 (Alk=Na, K), Al_2O_3-SiO_2 and ternary Na_2O-K_2O-SiO_2 systems have already been successfully evaluated. The phase equilibria calculated using the new optimized solution data show good agreement with the experimental data. In contrast to other available databases the new dataset allows the description of the whole composition range including the alkali rich parts of the corresponding subsystems. The associate species model is applied for the description of the liquid and solid phases in the systems under consideration. The available phase diagrams were collected and evaluated for the purpose of improving the solution database. Data on the solution components and interaction parameters were optimized to represent the phase relationships in the systems under consideration.

In the system K_2O-Al_2O_3-SiO_2, the solution based on $KAlO_2$ is described in the framework of the associated species model. Solid solution components were selected and their thermodynamic data were assessed in order to obtain the best agreement with the experimental data.

In the system Na_2O-Al_2O_3-SiO_2, similar solutions with Na instead of K are introduced using the sublattice model for both crystalline modifications of $NaAlO_2$, low- and high-temperatures. Two other solid phases, based on modifications of $NaAlSiO_4$, nepheline and carnegieite, are also described using the sublattice approach. The appropriate model parameters for liquid and solid phases are adjusted for the preliminary representation of the phase equlibria taking into account the lack of experimental data on this system, especially for the Na_2O-rich part of the ternary diagram.