Internally consistent thermodynamic data for rock-forming minerals in the system SiO_2-TiO_2-Al_2O_3-Fe_2O_3-CaO-MgO-FeO-K_2O-Na_2O-H_2O-CO_2

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Abstract: An internally consistent thermodynamic data set has been derived utilizing results from all available experimental phase equilibria in a simultaneous iterative regression approach using $1/T$ *vs.* $\ln K_{red}$ plots. Simultaneous evaluation of all reactions allows the extraction of an internally consistent data set of $\Delta_f H^o$ and S^o values with V^o, c_P, α and β otherwise constrained. An objective function has been defined which requires maximum consistency with available experimental results and minimum deviation from available calorimetric data. One advantage of this procedure is that all available experimental results, compatible or not, are always visible during the calculations. Over 5300 experimental results from 244 contributions for 253 reactions involving 94 phase components have been used. The extracted data set reproduces 92% of the available phase equilibria experiments reported in the literature. It can be shown that for some phases the configurational entropy must be considered (*e.g.* CaAl-pyroxene, clinochlore, mullite, phlogopite). With the addition of a configurational entropy for these phases the reproduction of the available experimental results is significantly improved. The configurational entropy for these phases is chiefly responsible for the observed differences between this and other available internally consistent data sets. In these other data sets non-addition of configurational entropy for these phases is compensated for by adjustments in the enthalpy and/or in other refined thermophysical parameters such as heat capacities. The configurational entropies used here are quite straightforward and involve mostly tetrahedral Si/Al and octahedral Mg/Al disorder.

Key-words: internally consistent set of thermodynamic data, enthalpy of formation, third law entropy.

Introduction

Petrological phase equilibria can be calculated utilizing basic thermodynamic principles, if the required thermodynamic properties and functions for the phases are available. For each phase component these properties are the standard enthalpy of formation $\Delta_f H^o$, the standard entropy S^o, the heat capacities over the relevant range of temperatures (c_P), and the molar volumes as a function of temperature and pressure (V^o, α, β). If a phase transition occurs, the appropriate changes of these properties must be known. Order-disorder transitions and variable compositions of a phase as a function of pressure, temperature and bulk composition must be also taken into account. If the phases are not considered to be pure, activity-composition relations are required. In the case of fluids an equation of state is needed for the calculation of the fugacities. These particular thermodynamic properties and functions are available to varying extents from calorimetry, spectroscopy, X-ray crystallography and the study of experimental phase equilibria.

The available properties and functions for various mineral and fluid components have been collected in a series of thermodynamic data bases/sets. These have been used for geo-thermobarometry, calculations of specific phase equilibria in P-T-*x* space and the kinetic treatment of mineral reactions. The scope, character and extent of these available data bases, however, are quite different.