DEVELOPMENT OF NEW MATERIALS USING AN INTEGRATED APPROACH OF DFT CALCULATIONS, CALPHAD MODELING AND KEY EXPERIMENTS

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ICME (Integrated computational materials engineering) is a modern approach to develop new materials with tailored properties for a given application. Chemical thermodynamics is a cornerstone of this method, because it is always the first step to evaluate if new materials and their synthesis routes are feasible. To perform thermodynamic calculations, the Calphad method is generally employed because it is the only method to allow simulations in higher components systems (3 or more) taking into account complex solids, liquids and the gas phase. Calphad thermodynamic modeling is a phenomenological method, i.e. the underlying Gibbs energy functions for the phases are obtained by least square fitting using available theoretical and experimental data. Therefore, the need for the generation of high quality theoretical and experimental data is obvious. This integrated thermodynamic approach will be illustrated in this contribution on three different examples: the solubility limit of Nb in liquid Al and the consequences for the Al-rich equilibria in the Al-Nb binary system, Mg-Al-carbides as new option for grain refinement in magnesium alloys and the development of novel low CO_2 cements.

The solubility and heat of dissolution of Nb in Al was measured by isothermal drop calorimetry at 1126 K. In addition, the heat capacity of $NbAl_3$ was measured and calculated as a function of temperature. The obtained results contradict the peritectic nature of the Al-$NbAl_3$-liquid three phase equilibrium. In the case of Mg-Al-C, ab-initio DFT and phonon calculations were performed and compared to new heat capacity measurements. Based on this new data, a new Calphad modeling of this ternary system was performed and will be discussed. Typical Portland cement is at least a nine component system (CaO-SiO_2-Al_2O_3-Fe_2O_3-MgO-K_2O-Na_2O-SO_3-P_2O_5-TiO_2). Cement clinker, the main component of cements, is obtained by high temperature solid-liquid reaction using natural raw materials. It will be shown how a pragmatic approach to set-up a new multicomponent thermodynamic database using ab-initio calculations and experimental equilibrium data allowed a consistent thermodynamic description of the full clinker system. Based on this database, a simulation model for the clinker burning was set-up and compared to industrially obtained data with excellent agreement. The application of this new model to improve OPC clinker properties will be discussed.