Computational determination of the Pb-Bi phase diagram

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A molten Pb-Bi-alloy will be used as coolant in MYRRHA (Multi-purpose hybrid research reactor for high-tech applications), a prototype of an accelerator-driven nuclear fission reactor (“generation IV reactor”) that will be built at the Belgian Nuclear Research Center (start of construction in 2015, start of operation in 2025). Neutron capture by 209-Bi will inevitably lead to the appearance of 210-Po in the coolant (about 2 kg in the entire coolant volume of MYRRHA). In order to study interactions of Po with the coolant, knowledge of the Pb-Bi-Po phase diagram is required. But, firstly, it is necessary to build a good description of the binary Pb-Bi phase diagram.

Knowledge of binary and ternary phase diagrams is of vital importance to fundamental and applied materials science. They are the basic building blocks on which a lot of research and applications rely. During many decades, the determination of alloy phase diagrams has been a purely experimental activity. This remained the case when around 1980, the CALPHAD methodology became the standard approach: a set of techniques to convert measured formation enthalpies, melting temperatures, etc. at discrete concentration and temperature points into an overall consistent phase diagram. The input to CALPHAD schemes used to be entirely experimental, until around 1995 ab initio solid state calculations became able to provide equivalent input for solids calculated directly from the basic laws of quantum mechanics. Nowadays, newly published ternary phase diagrams are often based on a mixture of ab initio and experimental information. It is even possible to predict ground state phase diagrams based on ab initio calculations only.

The present work aims at determining the binary phase diagram for the Pb-Bi system, both in their ground state (0 K) as well as at elevated temperatures. This will be done by a combination of ab initio calculations and CALPHAD-based schemes. It goes beyond standard approaches because for these heavy elements relativistic effects have to be included, and because temperature dependent properties are explicitly calculated, i.e. we go beyond ground state predictions.