Comparing thermodynamic data for monatomic and diatomic gases from ab-initio and CALPHAD data

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Every advancement in technology such as energy generation, storage and conversion, requires or would benefit from new components. The objective is to reduce cost, increase performance or replace rare elements with more sustainable earth-abundant alternatives. To design or develop such new energy efficient materials, accurate thermochemical data is a key. Thermochemical quantities such as enthalpy of formation or entropy are among the most important chemical data. Large-scale experimentation to determine those quantities would defy the above-mentioned objective. Therefore, accurate computational chemistry methods, which allow a reliable prediction of thermochemical data, would be highly desirable.

The aim of this work is to compare and understand the thermodynamic properties of monatomic and diatomic gases calculated using ab-initio based approach (Density Functional Theory - DFT) and experimental based approach (CALculation of PHAse Diagrams - CALPHAD). This work focuses mainly on FCC elements, which are nonmagnetic in nature. For monatomic gases, elements Ag, Al, Au, Ca, Cu, Pb, Pd, Pt, Rh and Sr were considered. For diatomic gases all the above mentioned elements were considered except Pd, Pt and Rh. All CALPHAD calculations have been done using FactSage 7.1 (Database: FactPS). The source for FactPS database is NIST JANAF thermochemical data. To confirm the same, JANAF data for available elements have also been added to the results along with DFT and FactPS calculations. Here, the first-principles calculations were performed within the DFT framework as implemented in the Vienna Ab-initio Simulation Package (VASP).