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THERMODYNAMIC PROPERTIES OF LIZARDITE, DETERMINED BY CALORIMETRY

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In the context of radioactive waste disposal, the long term stability of the natural and engineered clay barriers must be assessed over hundreds of thousands of years. In particular, in the framework of glass/iron/clay interactions, recent experimental works have brought evidences of the precipitation of serpentine-like mineral. Until now, very few thermodynamic data are available for the serpentine minerals. The most common polymorphs are lizardite, chrysotile and antigorite. This study aims to determine a consistent thermodynamic dataset for a natural lizardite, in order to be integrated in thermodynamic databases (e.g., THERMODDEM (Blanc et al., 2012), ThermoChimie (Giffaut et al., 2014)) for geochemical modelling.

The lizardite sample, highly crystallized, was coming from Monte-Fico (Elba, Italy). The mean structural formula of the lizardite mineral was determined from microprobe analyses and ^{77}Fe Mössbauer spectrometry: $(Si_{1.908}Al_{0.074}Fe^{3+}_{0.017})(Mg_{2.722}Fe^{2+}_{0.094}Fe^{3+}_{0.048}Al_{0.105}Mn_{0.002})O_5(OH)_4$. The sample contains 25 wt.% of chrysotile according to Viti et al. (2011). The methodology used to determine the thermodynamic properties of the lizardite was described in Gailhanou et al. (2013). The standard enthalpy of formation at 298.15 K was obtained by acid solution calorimetry in HF-HNO_3 solution. The heat capacity were measured from 2 K to 300 K by PPMS and between 298 K and 520 K by DSC. The third-law entropy was determined by adding the measured calorimetric entropy and estimated residual entropy terms (configurational and magnetic). The Gibbs free energy of formation was then calculated from the standard enthalpy and entropy of formation.

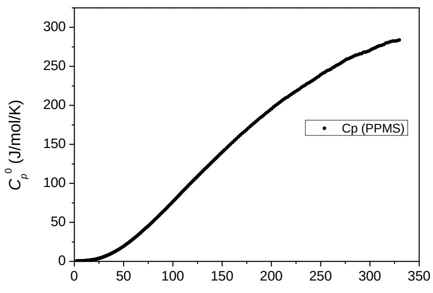

Figure 1. Heat capacity measurements for lizardite.

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