

Thermochemistry of polonium evaporation from LBE

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1. Heat Capacity Correlations for $\text{Po}_2(\text{g})$, $\text{PbPo}(\text{g})$ and $\text{BiPo}(\text{g})$

The heat capacity as function of temperature for the gas species $\text{Po}_2(\text{g})$, $\text{PbPo}(\text{g})$ and $\text{BiPo}(\text{g})$, calculated by Mertens [1] was correlated using the equation:

$$C_p(T) = A + 10^{-3} BT + 10^5 CT^2 + 10^{-6} DT^2 + 10^8 ET^{-3} + 10^{-9} FT^3 \quad (\text{S1})$$

The resulting fits are shown in Figures S1 ($\text{Po}_2(\text{g})$) and S2 ($\text{PbPo}(\text{g})$ and $\text{BiPo}(\text{g})$). The relative residuals, (fitted value – data value)/data value, were less than 1% in the considered temperature ranges.

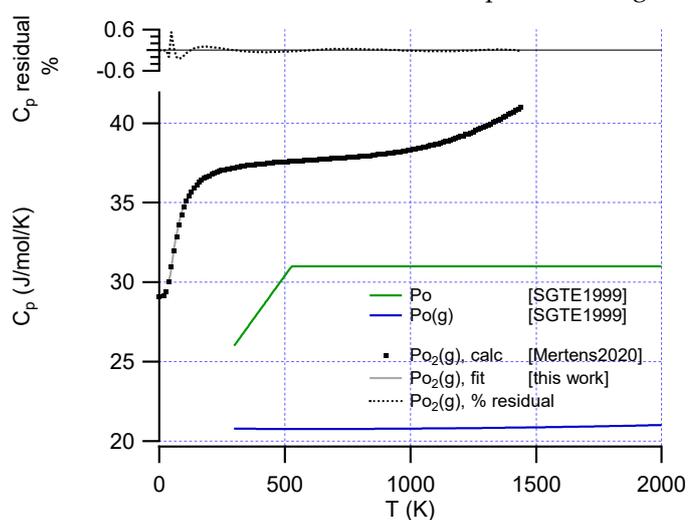


Figure S1. (black markers) Heat capacity of $\text{Po}_2(\text{g})$ by Mertens [1] and (grey line) fit according to Equation (S1) between 48.15 K and 1438.15 K. Top: relative residual for the fit for $\text{Po}_2(\text{g})$. The heat capacity functions for Po and $\text{Po}(\text{g})$ from SGTE [2] are plotted for comparison.

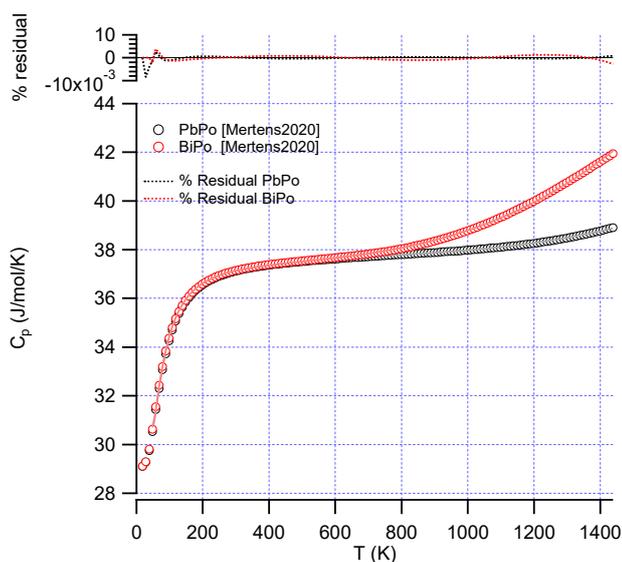


Figure S2. (markers) Heat capacity of PbPo(g) and BiPo(g) by Mertens [1] and (lines) fits according to equation SM1 between 48.15 K and 1438.15 K. Top: relative residuals.

2. PbPo(s)

The temperature dependence of the heat capacity C_p of PbPo(s) was estimated by simultaneously fitting the extrapolated enthalpy (H) and entropy (S) data obtained by Eichler [3] (Figure. S4) according to the correlation for C_p (Eq. S1) with $C=D=E=F=0$ and the following relations:

$$S = S(298.15K) + \int_{298.15}^T \frac{C_p(T)}{T} dT = S(298.15K) + A \ln(T) + 10^{-3}BT \quad (S2)$$

$$H = H(298.15K) + \int_{298.15}^T C_p(T)dT = H(298.15K) + AT + \frac{1}{2}10^{-3}BT^2$$

The optimized coefficients were $A = 47.368$ and $B = 16.164$.

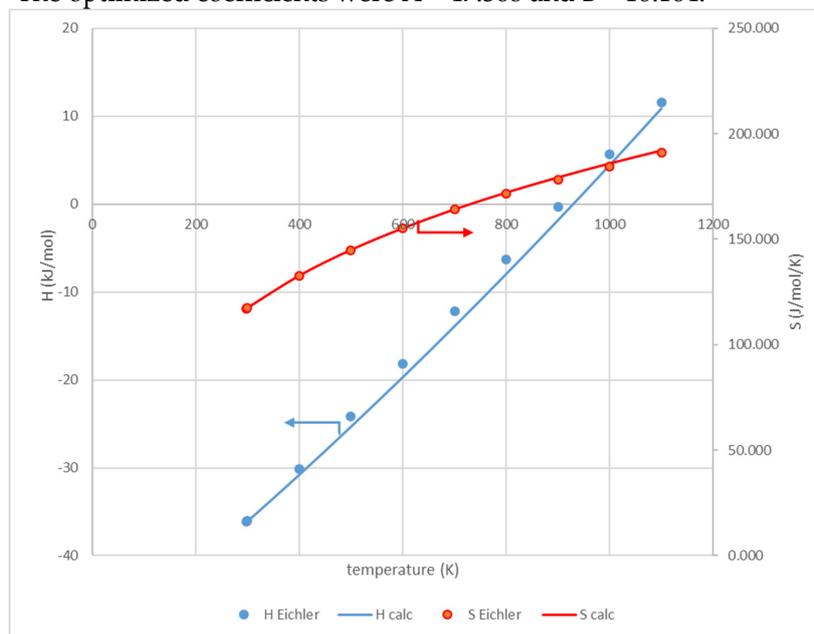


Figure S3. (markers) Enthalpy and entropy of PbPo(s) obtained by extrapolation by Eichler [3]. (curves) fits according to Equation (S2).

The resulting Gibbs energy functions ($G=H-TS$) for PbPo(s) were calculated (Figure S4). The current, optimized data predict a more stable PbPo(s) compound than when Eichler's data are used.

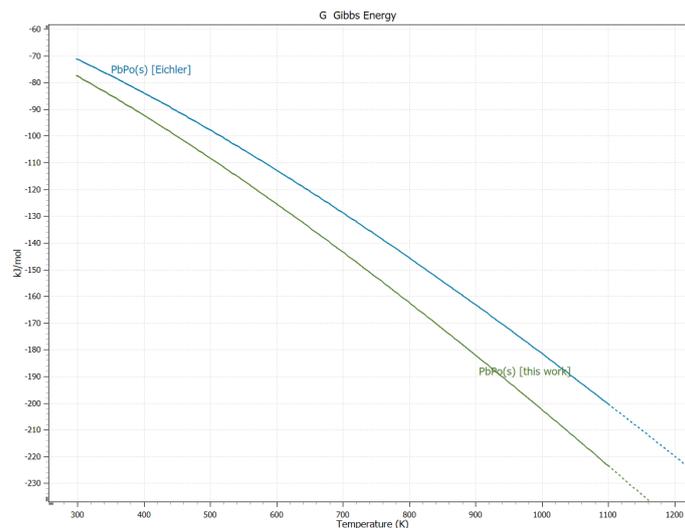


Figure S4. Gibbs energy G of PbPo(s) : comparison of G calculated from Eichler's extrapolation results and from the optimization presented in this work.

References

1. Mertens, M.A.J.; Cottenier, S. *Ab Initio Predicted Binding Energies of Small and Medium-Sized Po-Containing Molecules. Deliverable D4.2 of the European Union H2020 MYRTE Project*; 2019.
2. SGTE Thermodynamic Properties of Elements, Ni to S6. In *Landolt-Börnstein - Group IV Physical Chemistry · Volume 19A1: "Pure Substances. Part 1 Elements and Compounds from AgBr to Ba3N2"*; Lehrstuhl für Werkstoffchemie; Rheinisch-Westfälische Technische Hochschule Aachen (on behalf of SGTE), Ed.; Springer-Verlag Berlin Heidelberg, 1999.
3. Eichler, B.; Neuhausen, J. *Verflüchtigungspfade Des Poloniums Aus Einem Pb-Bi-Spallationstarget. Report of the Paul Scherrer Institut 04-06*; Villigen, Switzerland, 2004.