ERRATUM

Erratum: Corrigendum to "Thermochemistry of glass forming Y-substituted Sr-analogues of titanite ($SrTiSiO_5$)" [J. Mater. Res. 24(11), 3380 (2009)]

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This recently published paper[1] reported the thermodynamic stability of Sr-loaded titanium silicate waste form and their potential beta-decay product series with charge balance in the titanite composition ($Sr_{1-x}Y_{0.67x}TiSiO_5$). The crystallization behavior and glass stability of our samples were studied using x-ray diffraction (XRD) and thermogravimetry and differential scanning calorimetry (TG-DSC). $SrTiSiO_5$ undergoes bulk crystallization, while potential decay products undergo surface crystallization. With proper thermochemical cycle, the enthalpies of formation from oxides ($\Delta H^o_{f,ox}$) were obtained from drop solution calorimetry in a molten lead borate ($2PbO{\cdot}B_2O_3$) solvent at 702 °C. $\Delta H^o_{f,ox}$ were exothermic but became less so with increasing Y substitution. The destabilizing effect in titanite compositions with Y substitution was discussed in terms of the basicity difference between SrO and $YO_{1.5}$ and the ratios of non-tetravalent cations to tetravalent cations [$R_{cat} = (Sr + Y)/(Ti + Si)$]. $\Delta H^o_{f,ox}$ of $SrTiSiO_5$ and $CaTiSiO_5$ glasses were compared.

While our scientific reasoning and conclusions remain valid, unfortunately we found a calculation mistake in our thermochemical cycle. The reference enthalpy of drop solution (ΔH_{ds}) value of SrO is wrong. Instead of using the ΔH_{ds} value of SrO in lead borate solvent, we used that in sodium molybdate. Therefore, the purpose of this corrigendum is to report the correct enthalpies of formation and further compare the glass stability related to the titanite analogues.

The ΔH_{ds} of SrO in $2PbO{\cdot}B_2O_3$ solvent at 702 °C is -56.6 ± 2.2 kJ/mol and can be derived from the thermodynamic cycle listed in Table I. Thus, the enthalpies of formation from the constituent oxides ($\Delta H^o_{f,ox}$) for $SrTiSiO_5$, $Sr_{0.75}Y_{0.17}TiSiO_5$, and $Sr_{0.5}Y_{0.33}TiSiO_5$ glasses are -88.1 ± 2.7, -63.4 ± 2.5, and -38.9 ± 1.6 kJ/mol, respectively. The values reported in the original publication were -162.9 ± 2.4, -119.5 ± 2.4, and -76.3 ± 1.9 kJ/mol, respectively. The corrected enthalpy values are shown in Table II. The linear relationship between $\Delta H^o_{f,ox}$ and x ($YO_{1.5}$ content) and between $\Delta H^o_{f,ox}$ and R_{cat} are observed.

We now compare the enthalpy of formation of the analogues (Ca, Sr, Ba) of the titanite composition. $\Delta H^o_{f,ox}$ of $CaTiSiO_5$, $SrTiSiO_5$, and $BaTiSiO_5$ glass are -38.8 ± 3.4, -88.1 ± 2.7 (this work), and -127.6 ± 3.0 kJ/mol, respectively. Among titanite analogues, $BaTiSiO_5$ glass is most stable, $SrTiSiO_5$ is intermediate,

TABLE I. Thermochemical cycle used for calculation of the enthalpy of drop solution of SrO in lead borate solvent ($2PbO{\cdot}B_2O_3$) at 702 °C.

	Reaction	Enthalpy
1	$SrCO_3$ (solid, 25 °C) → SrO (dissolved, 702 °C) + CO_2 (gas, 702 °C)	$\Delta H_1 = \Delta H_{ds}$ ($SrCO_3$)[2]
2	SrO (solid, 25 °C) + CO_2 (gas, 25 °C) → $SrCO_3$ (solid, 25 °C)	$\Delta H_2 = \Delta H^o_{f,ox}$ ($SrCO_3$)[3]
3	CO_2 (gas, 702 °C) → CO_2 (gas, 25 °C)	$\Delta H_3 = -32.0$ kJ/mol
4	SrO (solid, 25 °C) → SrO (dissolved, 702 °C)	$\Delta H_4 = \Sigma\Delta H_i$ (i = 1–3)

$\Delta H_1 = 209.3 \pm 1.3$ kJ/mol[2]; $\Delta H_2 = -233.9 \pm 1.8$ kJ/mol[3]; $\Delta H_4 = -56.6 \pm 2.2$ kJ/mol.

TABLE II. Enthalpies of drop solution in lead borate at 702 °C (ΔH_{ds}) and enthalpies of formation from the oxides ($\Delta H^o_{f,ox}$) and from the elements ($\Delta H^o_{f,el}$) at 25 °C for Y-substituted Sr-analogue of titanite samples $Sr_{1-x}Y_{0.67x}TiSiO_5$ (x = 0, 0.25, 0.5), and reference values of binary oxides.

x	ΔH_{ds} (Jg^{-1})	ΔH_{ds} ($kJmol^{-1}$)	$\Delta H^o_{f,ox}$ ($kJmol^{-1}$)	$\Delta H^o_{f,el}$ ($kJmol^{-1}$)
0	517.2 ± 5.5	126.0 ± 0.8	−88.1 ± 2.7	−2608.1 ± 2.9
0.25	496.6 ± 5.6	117.4 ± 1.4	−63.4 ± 2.5	−2575.9 ± 2.8
0.5	475.4 ± 2.5	109.1 ± 0.6	−38.9 ± 1.6	−2543.8 ± 2.1
SrO	...	−56.6 ± 2.2	...	−591.3 ± 1.0
$YO_{1.5}$	...	12.0 ± 0.6	...	−952.7 ± 1.1
TiO_2	...	55.4 ± 1.2	...	−944.0 ± 0.8
SiO_2	...	39.1 ± 0.3	...	−910.7 ± 1.0

Uncertainty is two standard deviations of the mean.

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and $CaTiSiO_5$ is least stable. This reflects the basicity: BaO > SrO > CaO.

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