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Thermodynamic stability of solid $SrThO_3$

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Abstract

The Gibbs energy of formation of strontium thorate was determined by the Knudsen effusion forward collection technique. The evaporation process from a mixture of tungsten and strontium thorate in the Knudsen cell could be represented by the following heterogeneous equilibria: $5SrThO_3(s) + W(s) = Sr_2WO_5(s) + 5ThO_2(s) + 3Sr(g)$, $1670 < T/K < 2040$, $4SrThO_3(s) + W(s) = SrWO_4(l) + 4ThO_2(s) + 3Sr(g)$, $2135 < T/K < 2420$. The measured equilibrium vapor pressures of Sr(g) over the respective temperature regions are given by, $\ln(p/\mathrm{Pa})\ (\pm 0.21) = -44419.9/T + 23.67$, $(1670 < T/K < 2040)$, $\ln(p/\mathrm{Pa})\ (\pm 0.09) = -50009.0/T + 25.71$, $(2135 < T/K < 2420)$. The standard Gibbs energy of formation of $SrThO_3(s)$ derived from the vapor pressure data could be represented by $\Delta_f G^0\ (SrThO_3(s))\ (\pm 5.0\ \mathrm{kJ\ mol^{-1}}) = -1953.6 + 0.367 \cdot T$, $(1670 < T/K < 2040)$ $\Delta_f G^0\ (SrThO_3(s))\ (\pm 7.0\ \mathrm{kJ\ mol^{-1}}) = -1960.2 + 0.369 \cdot T$, $(2135 < T/K < 2420)$. © 2001 Elsevier Science B.V. All rights reserved.

1. Introduction

The third phase of the nuclear energy generation program in India envisages the use of thoria-based fuels [1]. For smooth and safe operation of the nuclear reactors, it is important to look into the chemical reactivity of the thoria matrix with the various fission products as the burn-up progresses. Formation of low-density compounds by interaction of the fission products with the thoria matrix is likely to affect the performance of the fuel due to swelling, changes in properties such as thermal expansion, thermal conductivity, etc. One chemical reactivity of thoria to be considered is that with strontium oxide produced from the fission product strontium. The resultant product strontium thorate, $SrThO_3$ is reported [2] to have quite a high molar volume as compared to that of the thoria matrix. However, the thermodynamic stability data of the compound required for the prediction of its formation in the reactor is not reported. This investigation thus addresses to this problem and reports the stability data obtained by a vaporization study of strontium thorate at high temperatures (1670–2420 K).

2. Experimental

Strontium thorate powder prepared through citrate–nitrate gel combustion was characterized for its purity by chemical assay and X-ray diffraction (XRD) analysis. The XRD pattern (Fig. 1) recorded for the compound agreed well with that reported by Naray-Szabo [3] and Purohit et al. [4]. The XRD data of the compound could be indexed in a monoclinic unit cell with the following lattice parameters: $a = 631.9(4)$ pm, $b = 324.0(1)$ pm, $c = 492.8(3)$ pm, and $\beta = 117.38(5)^0$. The carbon content in the powder was seen to be less than 0.1%. The extent of moisture absorbed by the sample during handling in air was determined by recording several TGA runs on the samples prior to the evaporation study. The nature of phase equilibria between tungsten and strontium thorate was also established in different temperature regions (1670–2420 K). For this, a powder

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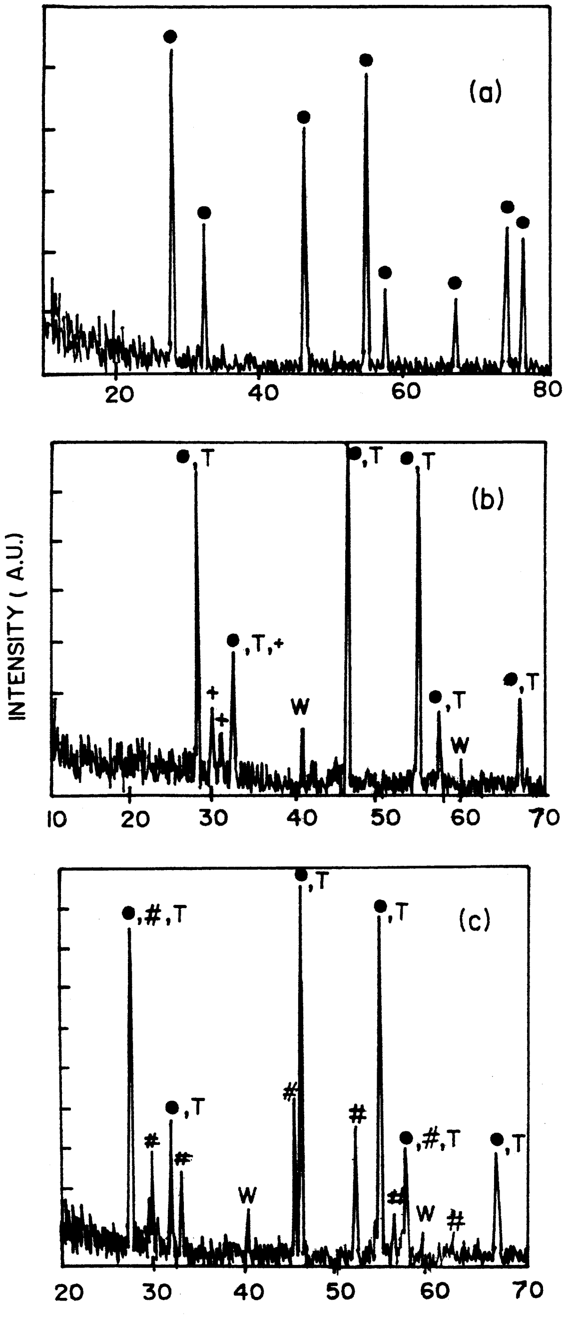

Fig. 1. XRD patterns of solid $SrThO_3$ as prepared (top), of the phase mixture $SrThO_3 + W$ equilibrated at $(1670 < T/K < 2040)$ (middle) and at $(2135 < T/K < 2420)$ (bottom). The patterns are shown as the intensity of peaks (arbitrary units) against the diffraction angle (2θ in degrees). (●) $SrThO_3$ peak, (+) Sr_2WO_5 peak, (#) $SrWO_4$ peak, (**T**) thoria peak and (**W**) tungsten peak.

mixture of $SrThO_3$ and tungsten taken in a tungsten cup within a tungsten Knudsen cell was heated inductively under a dynamic vacuum of 10^{-9} bar for several hours and then quenched under vacuum. After the experiment, the residue in the tungsten cup was analyzed for phases by XRD and EDX techniques.

XRD data of the residue after the partial evaporation of the sample revealed that the nature of condensed phases were different at different temperature regions as described below:

(i) $SrThO_3(s)$, $Sr_2WO_5(s)$, W(s) and $ThO_2(s)$, $1670 < T/K < 2040$,

(ii) $SrThO_3(s)$, $SrWO_4(l)$, W(s) and $ThO_2(s)$, $2135 < T/K < 2420$.

Typical XRD patterns of the phase mixtures for the two temperature regions are given in Fig. 1. SEM and EDX analysis were carried out in case (ii) to find the dissolved components within the frozen melt of the $SrWO_4$ phase (melting point 1808 K). The analysis indicated nearly a pure phase of $SrWO_4$. A few atom per cent (less than 10%) of thorium seen in the EDX spectrum was either from an interference of the neighboring phase of $ThO_2(s)$ or from a truly dissolved chemical component such as $SrThO_3$ within the liquid phase. The $SrWO_4$ phase was approximately treated to be a pure one in the subsequent thermodynamic analysis. Similarly, $ThO_2(s)$ in the above mentioned phase equilibria (i) and (ii) was approximated to be a pure phase though it is reported to have some solubility of SrO [5]. Based on the ideal solubility, this approximation amounts to an error of 2.6 kJ mol^{-1} in the Gibbs energy per mole of ThO_2 at the maximum concentration of 13 mol% SrO reported at 2273 K [5].

It is evident from the phase equilibrium study that the mixture of $SrThO_3(s)$ and W(s) reacted chemically forming thoria and the strontium tungstates in the different temperature regions. In the reactions, a part of strontium thorate underwent reduction to Sr(g) and $ThO_2(s)$ thereby oxidizing W(s) to its hexavalent state in the form of the tungstates, $SrWO_4$ or Sr_2WO_5. Reduction of SrO(s) by tungsten forming Sr(g) and the tungstates has been reported in the literature [6–10]. The reported thermodynamic analysis of the Sr–W–O ternary [9] shows that the tungstates remain in equilibrium with W(s) at a very low pressure of oxygen ($p_{O_2} \leqslant 10^{-10}$ bar) and that the formation of the perovskite $SrWO_3(s)$ is unfavorable in comparison. When the reduction process is carried out in an open system as in Knudsen evaporation the reduction product, strontium, escapes as vapor. Therefore, in the presence of an excess quantity of tungsten the system is driven through the phase fields of $Sr_3WO_6(s)$ and $Sr_2WO_5(s)$ and ultimately, to the two-phase mixture of $SrWO_4(s)$, and W(s) [9]. In the light of the oxidation–reduction behavior of strontium oxide and the tungsten system, the observed phase equilibria in $SrThO_3(s) + W(s)$ system could be explained by the following reactive paths:

$$5SrThO_3(s) + W(s) = Sr_2WO_5(s) + 5ThO_2(s) + 3Sr(g), \quad (1670 < T/K < 2040), \tag{1}$$

$$4SrThO_3(s) + W(s) = SrWO_4(l) + 4ThO_2(s) + 3Sr(g), \quad (2135 < T/K < 2420). \tag{2}$$

The Sr vapor pressure remained invariant at a temperature under any of the above heterogeneous reaction equilibria. The vapor pressure was monitored by the Knudsen effusion forward collection technique. It is to be noted that the phase equilibrium and the vapor pressure study were also attempted below 1670 K, where the reactive path leading to the Sr evaporation was found to be as

$$6SrThO_3(s) + W(s) = Sr_3WO_6(s) + 6ThO_2(s) + 3Sr(g).$$

However, as will be discussed under the results, the vaporization data below 1670 K showed that the solid state reaction is too sluggish to maintain the equilibrium vapor pressure of Sr under the dynamic evaporation.

For the vapor pressure study of the equilibrated mixture of $SrThO_3(s) + W(s)$, the contribution of the other strontium bearing vapor species, e.g., SrO(g), $SrWO_3$ and $SrWO_4$ was obtained by considering mass spectroscopic [7] and other thermodynamic information [11]. It was found that the Sr pressure observed in the exploratory runs of the system was orders of magnitude higher than the summed contributions of the simple and double oxide species irrespective of the temperature regions.

The Sr effusate from the Knudsen cell was received in a defined geometry and condensed on water cooled tantalum collectors. Experimental details involving the measurement of temperature and collection of effusate on targets are given in references [12,13]. The collected strontium was dissolved in 0.1 N hydrochloric acid, the respective solutions were made up to 10 ml and the strontium content of the solutions was determined by using the inductively coupled plasma spectrometric (ICPS) technique. Thus the Sr flux received by the collectors could be determined.

3. Results

The Sr vapor pressure was calculated from the measured vapor flux at a temperature. The vapor pressure expression for Sr(g) in the Knudsen cell was obtained by using the kinetic theory of gases for the effusion from a knife-edged orifice and considering the forward collection of Sr vapor under the given geometry by the relation

$$p_{Sr} = (1/A) \times (w_{Sr}/t) \times [(r^2 + d^2)/r^2] \times [2\pi RT/M_{Sr}]^{1/2}. \tag{3}$$

In the above equation, w_{Sr} is the amount of strontium that effuses out of the orifice of area, A, during time, t. R is the gas constant, T is the temperature of the Knudsen cell monitored during the vapor collection. M_{Sr} is the atomic weight of Sr, $[(r^2 + d^2)/r^2]$ is the geometric factor, r being the radius of the collimator used, and d the orifice to collimator distance. The values of the parameters, A, r and d used in the experiments were typically 5.02×10^{-7} m^2, 0.00875 m and 0.068 m, respectively.

The vapor pressures of Sr(g) calculated from the experimentally determined parameters of Eq. (3) in the temperature regions $(1670 < T/K < 2040)$ and $(2135 < T/K < 2420)$ are presented in Tables 1 and 2, respectively, and plotted in Fig. 2. The tabulated pressures for the second region are those after correction for deviation from Knudsen flow [14] due to high values of p_{Sr} at the high temperatures. The correction involves the use of the collisional diameter σ_c of Sr(g) which was taken to be 430 pm from a consideration of the reported atomic radius of the element. A sim-

Table 1
Vaporization data on reaction (1) and the Gibbs energy of formation of $SrThO_3(s)$

Temperature (K)	Time (s)	Mass collected w_{Sr} (μg)	Pressure p_{Sr} (Pa)	$\Delta_f G^0$ ($SrThO_3$,s) (kJ mol^{-1})
1677	9360	6.0	0.098	−1338.14
1695	6900	3.3	0.074	−1331.54
1740	6600	5.5	0.130	−1315.02
1821	2340	4.4	0.300	−1285.29
1868	900	5.5	0.986	−1268.04
1868	900	4.5	0.807	−1268.04
1950	600	7.0	1.924	−1237.95
1950	600	11.0	3.024	−1237.95
2038	480	20.0	7.025	−1205.65
2047	480	25.0	8.801	−1202.35

Table 2
Vaporization data on reaction (2) and the Gibbs energy of formation of $SrThO_3(s)$

Temperature (K)	Time (s)	Mass collected w_{Sr} (μg)	Pressure p_{Sr} (Pa)	$\Delta_f G^0$ ($SrThO_3$,s) (kJ mol^{-1})
2138	1200	74	2.27	−1171.28
2138	1200	85	2.41	−1171.28
2185	720	90	2.95	−1153.94
2185	720	74	2.75	−1153.94
2216	420	62	3.10	−1142.50
2216	420	60	3.07	−1142.50
2279	241	70	3.78	−1119.25
2333	182	100	4.35	−1099.32
2333	180	95	4.31	−1099.32
2419	144	155	4.99	−1067.59
2419	147	170	5.06	−1067.59

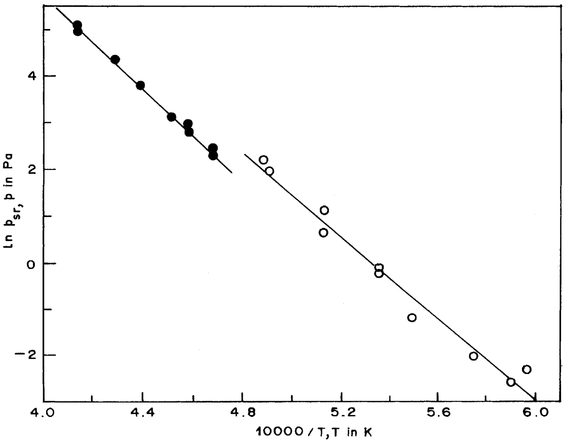

Fig. 2. Plot of $\ln p_{Sr}$ against reciprocal of temperature in the two temperature regions where p_{Sr} is in Pa and temperature in K.

ilar value of σ_c for the atom could be arrived from its Lennard–Jones potential parameter σ by linear interpolation of σ versus σ_c for the known cases. The maximum correction results in about 35% reduction in the pressure value observed at the highest temperature, i.e., at 2420 K. The linear least-square fits of the two sets of data are given by

$$\ln(p_{Sr}/\mathrm{Pa})(\pm 0.30) = -44419.9/T + 23.67, \quad (1670 < T/\mathrm{K} < 2040), \tag{4a}$$

$$\ln(p_{Sr}/\mathrm{Pa})(\pm 0.07) = -50009.0/T + 25.71 \quad (2135 < T/\mathrm{K} < 2420). \tag{5a}$$

The observed data below 1670 K could also be fitted as $\ln(p_{Sr}/\mathrm{Pa})(\pm 0.21) = -67093.8/T + 34.54$, $(1610 < T/\mathrm{K} < 1670)$. However, the sublimation enthalpy of Sr(g) derived from this vapor pressure equation is about 150 kJ mol^{-1} higher as compared to those obtainable from Eqs. (4a) and (5a). This is in contradiction with the nature of the condensed phase equilibrium seen in the lower temperature region. Here, the formation of the higher tungstate, $Sr_3WO_6(s)$ with more enthalpic stability should have occurred with a lower sublimation enthalpy of Sr. The apparently high sublimation enthalpy observed in the lowest temperature region reflects the impediment of the reaction by solid state kinetics at the low temperature. Under the dynamic observation made, the activation enthalpy of the involved kinetics got added to the true enthalpy of the Sr sublimation. The vapor pressure results in the lowest temperature region are, therefore, not included in the presentation of equilibrium data obtained in the other two temperature regions as given in Tables 1 and 2 or in Fig. 2.

4. Discussion

The vapor pressure expressions Eqs. (4a) and (5a) were, respectively, used to calculate the standard Gibbs energy change for the heterogeneous reactions (1) and (2) as given by

$$\Delta G_2^0(\pm 14\ \mathrm{kJ\ mol^{-1}}) \equiv -3RT \ln p_{Sr}/\mathrm{bar} = 1107.9 - 0.303 \cdot T \tag{4b}$$

and

$$\Delta G_3^0(\pm 4\ \mathrm{kJ\ mol^{-1}}) \equiv -3RT \ln p_{Sr}/\mathrm{bar} = 1247.3 - 0.354 \cdot T. \tag{5b}$$

The quantities ΔG_2^0 and ΔG_3^0 can also be expressed in terms of the standard Gibbs energy of formation of the reactants and products of reactions (1) and (2), respectively, as

$$\Delta G_2^0 = \Delta_f G^0(Sr_2WO_5(s)) + 5\Delta_f G^0(ThO_2(s)) - 5\Delta_f G^0(SrThO_3(s)) \tag{4c}$$

and

$$\Delta G_3^0 = \Delta_f G^0(\mathrm{SrWO_4(l)}) + 4\Delta_f G^0(\mathrm{ThO_2(s)}) - 4\Delta_f G^0(\mathrm{SrThO_3(s)}). \tag{5c}$$

It is to be noted that the mean working temperatures of both reactions (1) and (2) being above the reported normal boiling point (1700 K) of Sr(l), the standard state for Sr used in expressions (4c) and (5c) is its gaseous state at 1 bar pressure.

To obtain the standard Gibbs energy of formation of $\mathrm{SrThO_3(s)}$ from the vapor pressure data of reaction (1), the pair of Eqs. (4b) and (4c) were combined as

$$\Delta_f G^0(\mathrm{SrThO_3(s)}) = \frac{1}{5}[\Delta_f G^0(\mathrm{Sr_2WO_5(s)}) + 5\Delta_f G^0(\mathrm{ThO_2(s)}) - (1107.9 - 0.303T)]. \tag{4d}$$

Thus considering the reported thermodynamic data of the compounds $\mathrm{Sr_2WO_5(s)}$ [10] and $\mathrm{ThO_2(s)}$ [11], the following expression for $\Delta_f G^0(\mathrm{SrThO_3(s)})$ was obtained from Eq. (4d):

$$\Delta_f G^0(\mathrm{SrThO_3(s)})(\pm 4.0\ \mathrm{kJ\ mol^{-1}}) = -1953.6 + 0.367 \cdot T \quad (1670 < T/\mathrm{K} < 2040). \tag{6}$$

Eq. (6) can be used to express the Gibbs energy of formation $\Delta_m G^0$ of $\mathrm{SrThO_3(s)}$ from the constituent oxides in their standard states as

$$\Delta_m G^0(\mathrm{SrThO_3(s)})\ \mathrm{kJ\ mol^{-1}} = -4.9 + 0.003 \cdot T \quad (1670 < T/\mathrm{K} < 2040). \tag{6a}$$

The standard Gibbs energy of formation of $\mathrm{SrThO_3(s)}$ in the temperature region of $2135 < T/\mathrm{K} < 2420$ can be derived considering Eqs. (5b) and (5c) together with the reported thermodynamic data of $\mathrm{SrWO_4(s)}$ [9,10] and is given by

$$\Delta_f G^0(\mathrm{SrThO_3(s)})(\pm 4.0\ \mathrm{kJ\ mol^{-1}}) = -1967.7 + 0.373 \cdot T + 0.25\Delta G_{\mathrm{fusion}}(\mathrm{SrWO_4(s)}) \quad (2135 < T/\mathrm{K} < 2420). \tag{7}$$

In the above derivation, the extent of deviation from stoichiometric $\mathrm{SrWO_4}$ in the liquid phase was not considered, as the preferential evaporation of $\mathrm{WO_3(g)}$ was estimated to be at least three orders of magnitude lower than the observed evaporation of Sr. The estimation considers the equilibrium reaction

$$\mathrm{WO_3(g) + 3Sr(g) + 3ThO_2(s) = W(s) + 3SrThO_3(s)},$$

with $a_{\mathrm{SrO}} < 1$ in $\mathrm{SrThO_3(s)}$, occurring inside the W Knudsen cell. In the absence of any reported thermodynamic data on melting of $\mathrm{SrWO_4(s)}$ as required in Eq. (7) the enthalpy and entropy of fusion of the tungstate were estimated. By considering the reported enthalpy of fusion of sulfates, chromates, molybdates and tungstates of alkali and alkaline earth metals [11] a value of (30 ± 10) kJ mol^{-1} was chosen for $\Delta H^0_{\mathrm{fusion}}(\mathrm{SrWO_4, s})$. By using the fact that $\Delta H^0_{\mathrm{fusion}}/T$ and $\Delta S^0_{\mathrm{fusion}}$ are equal at the melting temperature for a substance, the $\Delta G_{\mathrm{fusion}}(\mathrm{SrWO_4(s)})$ term appearing in Eq. (7) was approximately expressed as $(30 - 0.016 \cdot T)$ kJ mol^{-1}. Eq. (7) therefore leads to

$$\Delta_f G^0(\mathrm{SrThO_3(s)})(\pm 10.0\ \mathrm{kJ\ mol^{-1}}) = -1960.2 + 0.369 \cdot T \quad (2135 < T/\mathrm{K} < 2420). \tag{8}$$

Eq. (8) can be used to express the Gibbs energy of formation of $\mathrm{SrThO_3(s)}$ from the constituent oxides as

$$\Delta_m G^0(\mathrm{SrThO_3(s)})\ \mathrm{kJ\ mol^{-1}} = -11.5 + 0.004 \cdot T \quad (2135 < T/\mathrm{K} < 2420). \tag{8a}$$

From Eqs. (6) and (8), it is evident that the two values of $\Delta_f G^0(\mathrm{SrThO_3(s)})$ obtained independently from the two reactions, namely reactions (1) and (2), are in good agreement considering the overall uncertainties. At the respective mean working temperatures of 1855 and 2275 K of the two reactions, the values of $\Delta_f H^0$ $(\mathrm{SrThO_3(s)})$ were obtained as -1953.6 and -1960.2 kJ mol^{-1}. In the absence of thermal function data of $\mathrm{SrThO_3(s)}$ no attempt was made to get the corresponding values of $\Delta_f H^0$ (298.15 K, $\mathrm{SrThO_3(s)}$) from the present result.

There are no reported data on the thermodynamic properties of $\mathrm{SrThO_3(s)}$ to compare with our result. However, one can make a fairly good estimate for its enthalpy and entropy of formation from considerations of available thermodynamic data of $\mathrm{A^{II}B^{IV}O_3}$ perovskites. In the synthesis of the perovskites from their constituent oxides, the entropy change $(\Delta_m S^0)$ is generally very small and the enthalpy change $(\Delta_m H^0)$ bears a linear relationship with the ionic radius r_B of the six-coordinated $\mathrm{B^{IV}}$ ion [15]. The linearity relation reported among the Sr-based perovskites of actinide and rare earth elements leads to the value of $\Delta_m H^0$ of (1 ± 20) kJ mol^{-1} for $\mathrm{SrThO_3(s)}$. On the other hand, the reported statistical analysis method of evaluating $\Delta_m H^0$ for the double oxides [16] when applied considering the thermodynamic data of other thorates and Sr compounds yielded a value of $\Delta_m H^0$ of -11 kJ mol^{-1} for $\mathrm{SrThO_3(s)}$. It may be noted that these estimates are in conformity with the results given in Eqs. (6a) and (8a). The very small values of $\Delta_m S^0$ and $\Delta_m H^0$ arrived in this study reveal that $\mathrm{SrThO_3(s)}$ has a marginal stability over its constituent oxides.

5. Conclusion

The experimentally derived thermodynamic properties of $\mathrm{SrThO_3(s)}$ suggest that the formation of strontium thorate from the constituent oxides is thermodynamically feasible at the normal working temperatures of reactors based on thoria fuel. However, because of the marginal stability of the compound there is a high possibility that strontium oxide will be fixed within the fuel pin by formation of a more stable perovskite such as $SrZrO_3$.

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