

Note

THE VAPOUR PRESSURE OF DI-CAESIUM TELLURIDE (Cs_2Te)

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In the system Cs–Te several compounds have been identified [1]. Of these Cs_2Te is of importance in a study of the safety aspects of water-cooled nuclear reactors, tellurium being one of the main volatile fission products of the “source term”. Although it is known that Cs_2Te is rather volatile, quantitative data are completely lacking. As part of our work on a study of the thermodynamic properties of the “source term” we here present measurements of the vapour pressure of Cs_2Te .

EXPERIMENTAL

Sample preparation and characterization

Cs_2Te was prepared by direct reaction of caesium vapour with solid tellurium at about 220°C, in a closed, evacuated pyrex capsule. Before reaction tellurium was purified by melting in a hydrogen stream at 500°C for 3 h. After the experiment TeO_2 can be removed from the surface.

Powdered tellurium is placed in a pyrex capsule together with three smaller pyrex capsules containing the caesium. The large capsule is then evacuated, sealed and slowly heated. Above 130°C the reaction starts; caesium vapour escaping from the small capsules reacts with tellurium in the bottom of the large capsule. The temperature is gradually raised to 220°C and maintained at this level for 3 days, before being raised to 280°C and maintained for 5 days. Finally, maintaining the temperature at 300°C for 5 days completes the reaction. The resulting product is mostly black due to the presence of Cs_3Te_2 . To convert this into Cs_2Te , the sample is thoroughly powdered in the glove box and heated with some extra caesium at 300°C. Excess caesium is removed by distillation at ca. 350°C. The resulting product is yellow green, and is shown by X-ray diffraction to be pure Cs_2Te .

Cs_2Te is extremely sensitive to oxygen and water vapour, and was always handled in a glove box filled with dry argon.

Vapour pressure measurements

The vapour pressure of Cs_2Te was measured using the transportation method. The principle of this method is that the saturated vapour of the

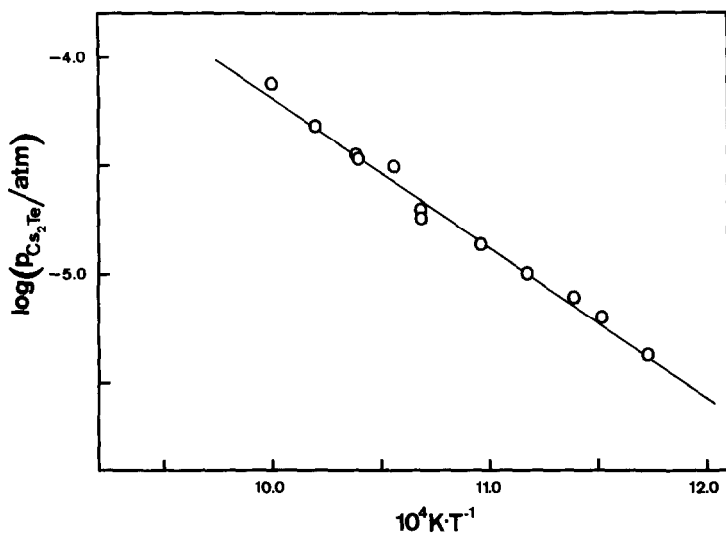


Fig. 1. Graphical results of the transportation measurements.

sample is condensed in a tube and determined after transportation with a known amount of inert carrier gas. The apparatus and the experimental conditions have been described previously [2]. After the experiment the amount of condensate in the condenser tube was determined by dissolving it in a mixture of 0.04 mol HNO_3 and 0.032 mol HCl per litre of water. Because some evolution of H_2Te was always observed during the dissolution, caesium only in the solution was determined (by atomic absorption spectrophotometry).

To study the experimental conditions, runs with different gas flows were carried out and the surface area of the sample was varied. It was found very difficult to obtain saturation of the carrier gas with the heavy Cs_2Te vapour. A large surface area, a short distance between the surface and the opening of the condenser tube, and a gas flow of ca. $50\text{--}70 \text{ ml min}^{-1}$ are necessary. Under these conditions reproducible values of the vapour pressure were obtained which were assumed to be equilibrium values. The results of the experiments are listed in Table 1, and shown in Fig. 1.

RESULTS

Equilibrium vapour pressures of Cs_2Te have been calculated from the data in Table 1, taking $\text{Cs}_2\text{Te}(\text{g})$ as the only gaseous species *. Quenching experiments, carried out at the highest temperature of vapour pressure

* 1 atm. = 1.013 bar.

TABLE 1
Numerical results of the transportation measurements

| Furnace temp. (K) | Volume of bottle (10^{-6} m ³) | Temperature (K) | | Pressure (mmHg) | | Amount of Cs transported (mg) | $P_{\text{Cs}_2\text{Te}}$ (atm.) |
|-------------------|---|-----------------|-----------------|-----------------|------------------------|-------------------------------|-----------------------------------|
| | | Lab. | Water in bottle | Atm. | ΔP (manometer) | | |
| 961.3 | 6194.3 | 296.4 | 294.7 | 757.0 | 3.6 | 2.268 | 3.443×10^{-5} |
| 962.2 | 6194.3 | 295.4 | 293.2 | 753.9 | 2.5 | 2.349 | 3.541×10^{-5} |
| 894.3 | 6194.3 | 295.3 | 292.5 | 756.0 | 3.0 | 0.673 | 1.013×10^{-5} |
| 912.0 | 6194.3 | 297.8 | 296.0 | 763.9 | 2.1 | 0.885 | 1.350×10^{-5} |
| 935.2 | 6194.3 | 297.8 | 293.1 | 770.3 | 1.9 | 1.156 | 1.758×10^{-5} |
| 935.1 | 6194.3 | 297.5 | 292.8 | 770.9 | 1.4 | 1.296 | 1.967×10^{-5} |
| 868.4 | 6194.3 | 298.6 | 296.0 | 766.0 | 2.6 | 0.411 | 6.289×10^{-6} |
| 999.8 | 6194.3 | 299.5 | 296.8 | 760.5 | 2.4 | 4.850 | 7.445×10^{-5} |
| 946.8 | 6194.3 | 298.2 | 296.9 | 756.4 | 3.2 | 2.033 | 3.118×10^{-5} |
| 877.9 | 6194.3 | 296.3 | 294.0 | 758.3 | 1.7 | 0.516 | 7.803×10^{-6} |
| 980.4 | 6194.3 | 296.0 | 293.4 | 758.8 | 1.6 | 3.151 | 4.753×10^{-5} |
| 852.3 | 6194.3 | 295.8 | 293.7 | 761.4 | 2.3 | 0.281 | 4.237×10^{-6} |

measurements, showed that Cs_2Te vaporizes congruently. Although the quenched samples were poorly crystalline, the X-ray diffraction pattern of Cs_2Te was clearly visible. Moreover, when the quenching experiments were carried out in quartz glass, no indications for dissociation in the vapour (e.g. $2\text{Cs}_2\text{Te}(\text{g}) \rightarrow \text{Cs}_3\text{Te}_2(\text{s}) + \text{Cs}(\text{g})$) were obtained, i.e. no attack on the glass was observed.

A linear pressure function is calculated with the least-squares method to be

$$\log(\text{Cs}_2\text{Te}/\text{atm})[852-1000 \text{ K}] = -(6904 \pm 249)/T + (2.72 \pm 0.27)$$

Any influence of the stoichiometry of the solid phase (which had the composition $\text{Cs}/\text{Te} = 2.00 \pm 0.03$) has not been observed.

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