

The thermochemical and thermophysical properties of Cs_2RuO_4 and Cs_2MnO_4 at temperatures from 5 K to 1000 K

E. H. P. CORDFUNKE, R. R. VAN DER LAAN,

*Netherlands Energy Research Foundation ECN,
Petten, The Netherlands*

and EDGAR F. WESTRUM, JR.

*Department of Chemistry,
University of Michigan, Ann Arbor, MI 48109, U.S.A.*

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Low-temperature heat capacities from 5 K to 350 K by adiabatic calorimetry and high-temperature enthalpy increments above $T = 450$ K to 800 K by drop calorimetry of Cs_2RuO_4 and Cs_2MnO_4 have been measured. The two compounds exhibit solid-to-solid phase transitions at $T = 906.8$ K and $T = 1051.9$ K, respectively, and melt at $T = 1211.8$ K and $T = 1175.5$ K, respectively. The enthalpies of transition and the melting temperatures have been determined by d.s.c. measurements. From the results, smoothed thermochemical and thermophysical functions have been tabulated at selected temperatures up to 1000 K. For the standard molar entropies of Cs_2RuO_4 and Cs_2MnO_4 at $T = 298.15$ K the values $S_m^\circ/R = 31.64$ and 28.77, respectively, have been found.

1. Introduction

The fission products caesium and ruthenium play an important role in the accident analysis of nuclear reactors because of their radiological hazard. Recently, the possibility of the formation of a volatile caesium ruthenate has been considered. Such an enhancement of ruthenium transport makes knowledge of its thermochemical properties of particular interest. In a previous paper⁽¹⁾ we described the preparation and determination of its standard enthalpy of formation. In this paper we report measurements of the low-temperature heat capacity and the high-temperature enthalpy increment of Cs_2RuO_4 .

For isostructural Cs_2MnO_4 a different situation exists. Manganese is, as a constituent of stainless steel, present in large amounts in the core of a nuclear reactor, and formation of Cs_2MnO_4 can considerably lower the volatility of caesium. Since its high-temperature thermodynamic properties are virtually unknown, we here also report the low-temperature heat-capacity and high-temperature enthalpy-increment measurements.

2. Experimental

The preparation and characterization of Cs_2RuO_4 was described in a previous paper.⁽¹⁾ In this investigation two different samples, designated "32" and "36", were used; both contain some elemental silver as a contaminant arising from the preparation (table 1).

Cs_2MnO_4 was prepared in an analogous way by the reaction of Cs_2O with MnO_2 in purified oxygen in a silver boat at temperatures which were stepwise increased from 500 K to 775 K; the samples were homogenized between each step. Cs_2O was prepared by the oxidation of a known amount of caesium metal in oxygen at $T \approx 475$ K, after which the stoichiometric amount of MnO_2 was added. Since all preparations are very sensitive to moisture, the handling of the samples was done in an argon-filled dry glove box.

The Cs_2MnO_4 sample was established to be phase-pure by X-ray diffraction analysis. The caesium content was determined gravimetrically with kalignost, the manganese content complexometrically with EDTA, and the silver content after dissolution in HNO_3 (+ H_2O_2) by Atomic Absorption Spectroscopy (AAS). The results of the analyses are listed in table 1.

The low-temperature heat capacities were measured in an adiabatic calorimetric cryostat (laboratory designation Mark XIII) over the temperature range 5 K to 350 K using a gold-plated calorimeter made of high-conductivity oxygen-free copper. The temperature of the calorimeter was measured with a Leeds & Northrup platinum-encapsulated platinum resistance thermometer in an entrant well. The thermometer was calibrated by the U.S. National Bureau of Standards (NBS) against IPTS-48,⁽²⁾ and is judged to reproduce thermodynamic temperatures within 0.03 K from 5 K to 300 K. Determinations of mass, current, potential differences, and time were referred to standardizations and calibrations performed at NBS. For a more detailed description of the technique and apparatus, see reference 2. For the measurements, 5.6127 g of Cs_2RuO_4 "32" or 5.3246 g of Cs_2MnO_4 was loaded into the calorimeter (laboratory designation W-99). The loading and unloading of the calorimeters were performed in an argon-filled glove box. To facilitate thermal contact between sample and calorimeter, a pressure 2.7 kPa (at 300 K) of helium was added after evacuation.

The enthalpy increments above $T = 298.15$ K were measured in an isothermal diphenyl-ether drop calorimeter, which is a modified version of the Bunsen-type ice

TABLE 1. Analytical results for Cs_2RuO_4 and Cs_2MnO_4 ; molar mass M and mass fraction w

Compound	$M/(\text{g} \cdot \text{mol}^{-1})$	$10^2 \cdot w(\text{Cs})^a$		$10^2 \cdot w(\text{Mn})$		$10^2 \cdot w(\text{Ag})$
		found	calc.	found	calc.	
Cs_2RuO_4 "32"	430.878	61.55 ± 0.08	61.69	—	—	0.03
Cs_2RuO_6 "36"	430.878	61.69 ± 0.05	61.69	—	—	1.06
Cs_2MnO_4	384.746	69.11 ± 0.10	69.09	14.05 ± 0.04	14.28	0.33

^a Corrected for the presence of silver in the sample.

calorimeter. For the measurements, the samples were enclosed in spherical silver ampoules with a volume of approximately 4 cm³. The ampoules were heated in a furnace the temperature of which was measured with a calibrated platinum-to-(platinum + 10 mass per cent of rhodium) thermocouple to ± 0.1 K. After thermal equilibration the ampoule was dropped into the calorimeter, where the energy of (the ampoule + the sample) melted diphenyl ether in equilibrium with its liquid in a closed system. The resulting volume increase of the ether was determined by weighing the displaced mercury. The ratio of energy input to mass of mercury making up the volume increase was a constant for the apparatus, and was obtained by calibration with NIST (formerly NBS) standard reference material (No. 720) synthetic sapphire, Al₂O₃. Details of the calorimeter, calibration, and performance are given in reference 3. For the present measurements, 7.30260 g of Cs₂RuO₄ "36" or 5.68311 g of Cs₂MnO₄ was enclosed in ampoules of mass 4.45812 g and 4.56170 g, respectively. Loading of the ampoules was performed in an argon-filled glove box, and all masses were corrected for the silver content, and for weighing in argon. A correction was made to account for the difference in enthalpy between the final calorimeter temperature (300.06 K) and the standard reference temperature, using $C_{p,m}^{\circ}$ (298.15 K) values from the present low-temperature measurements.

Phase transitions were measured in a d.s.c. apparatus (Mettler TA 2000) in oxygen with a heating rate of 0.083 K·s⁻¹, using the melting temperatures and molar enthalpies of fusion of Al and Ag as references.

3. Results and discussion

The results of the low-temperature heat-capacity measurements of Cs₂RuO₄ "32" and Cs₂MnO₄ in the temperature range 5 K to 350 K are given in table 2. The latter results have been corrected for the silver content in the samples. No phase transitions were observed in the heat capacity of either compound: the heat-capacity curves rise monotonically to $T = 350$ K. Smoothing and integration of the experimental results using the FITAB computer programme⁽⁴⁾ give the values at 298.15 K (table 3): for Cs₂RuO₄: $C_{p,m}^{\circ}/R = (18.78 \pm 0.036)$, $S_m^{\circ}/R = (31.64 \pm 0.031)$; and for Cs₂MnO₄: $C_{p,m}^{\circ}/R = (17.95 \pm 0.036)$; $S_m^{\circ}/R = (28.77 \pm 0.031)$.

Cs₂RuO₄ and Cs₂MnO₄ are isostructural with Cs₂MoO₄ (orthorhombic, space group Pnma) and have, like the latter compound⁽⁵⁾ a phase transition to a hexagonal structure.⁽⁶⁾ The transition temperatures and molar enthalpies of transition have been measured in the d.s.c. apparatus. We thus found for Cs₂RuO₄: $T_{\text{trs}} = (906.8 \pm 0.5)$ K and $\Delta_{\text{trs}}H_m^{\circ} = (2.78 \pm 0.13)$ kJ·mol⁻¹ as the mean of 12 runs. For Cs₂MnO₄ the values $T_{\text{trs}} = (1051.9 \pm 5.2)$ K and (0.59 ± 0.08) kJ·mol⁻¹ were obtained as the mean of 8 runs. We have also determined the melting temperatures of Cs₂RuO₄ and Cs₂MnO₄, which were found to be at (1211.8 ± 0.3) K (8 runs) and (1175.5 ± 0.5) K (3 runs), respectively. The molar enthalpy of fusion of Cs₂RuO₄ is (27.4 ± 0.5) kJ·mol⁻¹. The enthalpy of fusion of Cs₂MnO₄ could not be determined since the solid decomposes during melting.

The high-temperature molar enthalpy increments are given in table 4. The values have been fitted to polynomials by the least-squares method, using as boundary

conditions $\{H_m^\circ(T) - H_m^\circ(298.15 \text{ K})\} = 0$ at $T = 298.15 \text{ K}$ and the $C_{p,m}^\circ(298.15 \text{ K})$ values given above. We thus obtain for Cs_2RuO_4 (298.15 K to 693 K):

$$\{H_m^\circ(T) - H_m^\circ(298.15 \text{ K})\}/(R \cdot \text{K}) = 23.181(T/\text{K}) - 7.5297 \cdot 10^{-3}(T/\text{K})^2 + 0.79012 \cdot 10^{-5}(T/\text{K})^3 + 1.7947 \cdot 10^5(T/\text{K})^{-1} - 7053.4.$$

This function is, due to its curvature, applicable only within the temperature bounds indicated. To extrapolate the thermodynamic properties to 906.8 K, the temperature of the solid-to-solid phase transition, the function:

$$\{H_m^\circ(T) - H_m^\circ(298.15 \text{ K})\}/(R \cdot \text{K}) = 14.114(T/\text{K}) + 6.9564 \cdot 10^{-3}(T/\text{K})^2 - 4838.0,$$

can be used (693 K to 906.8 K).

TABLE 2. Experimental molar heat capacities of Cs_2RuO_4 and Cs_2MnO_4 ($R = 8.3145 \text{ J} \cdot \text{K}^{-1} \cdot \text{mol}^{-1}$)

T/K	$C_{p,m}^\circ/R$	T/K	$C_{p,m}^\circ/R$	T/K	$C_{p,m}^\circ/R$	T/K	$C_{p,m}^\circ/R$	T/K	$C_{p,m}^\circ/R$	T/K	$C_{p,m}^\circ/R$
Cs_2RuO_4 "32"											
5.36	0.340	27.35	4.579	60.86	9.804	128.62	14.57	210.79	17.12	293.59	18.68
6.05	0.450	29.10	4.967	63.83	10.24	133.70	14.78	215.92	17.17	298.77	18.78
7.46	0.740	30.85	5.379	66.96	10.43	138.80	14.96	221.09	17.38	303.95	18.75
9.02	0.780	32.61	5.774	70.26	10.82	143.92	15.07	226.25	17.48	309.11	18.99
10.28	0.880	34.39	6.162	73.72	11.05	149.03	15.31	231.46	17.66	314.27	18.98
11.62	1.080	36.18	6.451	77.34	11.38	154.13	15.48	236.66	17.62	319.42	19.04
12.89	1.251	37.97	6.845	81.09	11.75	159.28	15.60	241.86	17.82	324.57	19.30
14.18	1.488	39.78	7.160	85.04	12.13	164.41	15.88	247.03	17.87	329.72	19.47
15.44	1.696	41.68	7.435	89.27	12.40	169.54	15.93	252.18	17.98	334.89	19.48
16.75	1.927	43.69	7.681	93.74	12.71	174.71	16.15	257.36	18.16	340.04	19.70
18.14	2.369	45.79	8.085	98.42	12.96	179.87	16.22	262.54	18.14	345.19	19.57
19.63	2.710	48.00	8.332	103.34	13.21	185.01	16.41	267.71	18.32		
21.13	3.031	50.14	8.670	108.37	13.49	190.16	16.51	272.88	18.43		
22.64	3.415	52.76	9.039	113.40	13.93	195.30	16.68	278.07	18.59		
24.17	3.805	55.32	9.299	118.47	14.00	200.46	16.77	283.24	18.61		
25.71	4.197	58.03	9.732	123.53	14.36	205.63	17.00	288.41	18.66		
Cs_2MnO_4											
Series I		30.96	4.862	69.60	9.599	110.34	12.28	192.87	15.50	278.76	17.58
6.26	0.247	32.82	5.189	73.05	9.974	115.37	12.60	196.26	15.70	283.95	17.69
7.94	0.301	34.36	5.431	76.65	10.23	120.46	12.90	201.39	15.82	289.10	17.79
9.07	0.531	35.99	5.711	80.44	10.45	125.54	13.00	206.54	15.98	294.20	17.87
10.51	0.590	37.70	6.031	84.42	10.77	130.61	13.38	211.70	16.09	299.00	17.97
12.05	0.926	39.49	6.293	88.61	11.05	135.71	13.44	216.87	16.24	304.41	18.08
13.49	1.057	41.37	6.551	93.01	11.32	140.81	13.80	222.02	16.34	309.51	18.16
14.78	1.424	43.34	6.937	97.68	11.59	145.92	13.93	227.18	16.47	314.60	18.25
16.02	1.587	45.41	7.211	102.52	12.02	151.05	14.16	232.36	16.61	319.74	18.34
17.37	1.882	47.60	7.446			156.13	14.36	237.51	16.71	325.04	18.46
18.68	2.125	49.88	7.822			161.26	14.48	242.60	16.82	330.25	18.53
20.00	2.451	52.30	8.121	Series II		166.38	14.71	247.82	16.94	335.63	18.62
21.63	2.585	54.83	8.360	83.66	10.99	171.53	14.85	253.01	17.04	340.93	18.71
23.31	3.297	57.50	8.609	86.65	11.01	176.67	15.05	258.18	17.18	346.25	18.79
24.79	3.490	60.30	8.860	91.01	11.24	177.00	15.07	263.35	17.27		
26.54	3.885	63.23	9.161	95.57	11.45	181.00	15.24	268.51	17.38		
28.76	4.318	66.32	9.405	100.34	11.72	187.27	15.43	273.67	17.49		

For Cs₂MnO₄ the molar enthalpy increment in the temperature region 298.15 K to 798 K can be described by

$$\{H_m^\circ(T) - H_m^\circ(298.15 \text{ K})\}/(R \cdot K) = 14.274(T/K) + 6.7333 \cdot 10^{-3}(T/K)^2 + 0.30210 \cdot 10^5(T/K)^{-1} - 4955.7.$$

There is a good agreement between the low- and high-temperature results presented here. This is shown in figure 1, in which $\{H_m^\circ(T) - H_m^\circ(298.15 \text{ K})\}/(T - 298.15 \text{ K})$ is

TABLE 3. Thermodynamic properties at selected temperatures for Cs₂RuO₄ "32" and Cs₂MnO₄ (R = 8.31451 J · K⁻¹ · mol⁻¹)

T/K	C _{p,m} /R	Δ ₀ ^T S _m ^o /R	Δ ₀ ^T H _m ^o /(R · K)	Φ _m ^o /R	T/K	C _{p,m} /R	Δ ₀ ^T S _m ^o /R	Δ ₀ ^T H _m ^o /(R · K)	Φ _m ^o /R
Cs ₂ RuO ₄									
0	0	0	0	0	150	15.33	19.89	1472.5	10.071
5	(0.172)	(0.061)	(0.228)	(0.016)	160	15.66	20.89	1627.5	10.716
10	0.843	0.368	2.634	0.105	170	15.98	21.85	1785.7	11.343
15	1.609	0.853	8.745	0.270	180	16.28	22.77	1947.0	11.952
20	2.714	1.454	19.323	0.488	190	16.57	23.66	2111.2	12.545
25	4.055	2.208	36.332	0.755	200	16.84	24.51	2278.2	13.122
30	5.146	3.044	59.35	1.066	210	17.09	25.34	2447.9	13.684
35	6.300	3.926	88.04	1.411	220	17.33	26.14	2620.0	14.232
40	7.170	4.823	121.68	1.781	230	17.55	26.92	2794.4	14.767
45	7.878	5.710	159.36	2.169	240	17.77	27.67	2971.0	15.29
50	8.680	6.588	201.02	2.567	250	17.98	28.40	3149.7	15.80
60	9.813	8.275	293.73	3.379	260	18.18	29.11	3330.6	16.30
70	10.758	9.860	396.69	4.193	270	18.37	29.80	3513.4	16.78
80	11.613	11.353	508.6	4.996	280	18.54	30.47	3697.9	17.26
90	12.389	12.767	628.7	5.781	290	18.68	31.12	3884.0	17.73
100	13.075	14.108	756.1	6.548	298.15	18.78	31.64	4036.6	18.10
110	13.666	15.38	889.9	7.293	300	18.80	31.76	4071.4	18.18
120	14.169	16.59	1029.1	8.018	325	19.21	33.27	4543.8	19.29
130	14.601	17.75	1173.0	8.723	350	19.59	34.73	5022	20.38
140	14.981	18.84	1321.0	9.407					
Cs ₂ MnO ₄									
0	0	0	0	0	150	14.116	17.73	1328.9	8.871
5	(0.089)	(0.031)	(0.115)	(0.008)	160	14.490	18.65	1471.9	9.454
10	0.575	0.216	1.586	0.057	170	14.843	19.54	1618.6	10.022
15	1.415	0.600	6.467	0.169	180	15.18	20.40	1768.7	10.574
20	2.406	1.140	15.971	0.342	190	15.50	21.23	1922.1	11.114
25	3.530	1.795	30.756	0.565	200	15.80	22.03	2078.6	11.640
30	4.648	2.540	51.27	0.831	210	16.08	22.81	2238.0	12.153
35	5.546	3.326	76.82	1.131	220	16.34	23.56	2400.0	12.655
40	6.386	4.123	106.70	1.455	230	16.58	24.30	2564.6	13.145
45	7.158	4.922	140.64	1.796	240	16.80	25.01	2731.5	13.624
50	7.804	5.710	178.08	2.148	250	17.01	25.70	2900.6	14.094
60	8.855	7.230	261.59	2.870	260	17.21	26.37	3071.7	14.553
70	9.713	8.661	354.55	3.596	270	17.40	27.02	3244.8	15.00
80	10.461	10.008	455.49	4.314	280	17.60	27.66	3419.8	15.44
90	11.135	11.279	563.5	5.018	290	17.79	28.28	3596.8	15.88
100	11.749	12.485	678.0	5.705	298.15	17.95	28.77	3742.4	16.22
110	12.310	13.631	798.3	6.374	300	17.99	28.88	3775.7	16.30
120	12.821	14.725	924.0	7.024	325	18.45	30.34	4231.2	17.32
130	13.288	15.77	1054.6	7.657	350	18.86	31.72	4697.5	18.30
140	13.718	16.77	1189.7	8.273					

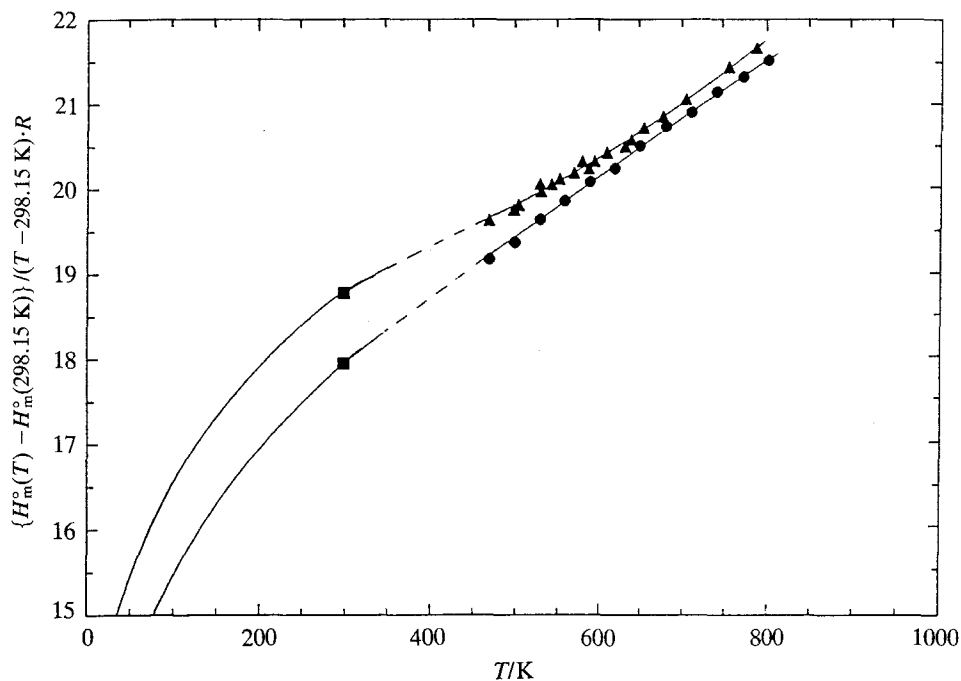


FIGURE 1. Values of $[\{H_m^o(T) - H_m^o(298.15 \text{ K})\} / (T - 298.15 \text{ K})] \cdot R$ plotted against T for Cs_2RuO_4 and Cs_2MnO_4 . \blacktriangle , Cs_2RuO_4 ; \bullet , Cs_2MnO_4 ; \blacksquare , $C_{p,m}^o(298.15 \text{ K})$.

TABLE 4. Experimental molar enthalpy increments of Cs_2RuO_4 "36" and Cs_2MnO_4

T K	$\{H_m^o(T) - H_m^o(298.15 \text{ K})\}$ ($R \cdot K$)		$10^2 \cdot \delta \Delta H_m^o$ ΔH_m^o	T K	$\{H_m^o(T) - H_m^o(298.15 \text{ K})\}$ ($R \cdot K$)		$10^2 \cdot \delta \Delta H_m^o$ ΔH_m^o
	expt	calc.			expt	calc.	
Cs_2RuO_4 "36"							
468.4	3343.0	3347.7	-0.14	586.9	5845.8	5861.0	-0.26
497.7	3943.7	3953.3	-0.24	594.0	6015.3	6017.5	-0.04
497.8	3950.6	3955.4	-0.12	608.9	6350.1	6348.3	0.03
502.9	4057.6	4061.8	-0.10	630.8	6820.3	6840.7	-0.30
528.3	4617.7	4596.3	0.46	638.4	7003.5	7013.5	-0.14
529.5	4621.2	4621.7	-0.01	652.7	7347.8	7341.0	0.09
542.0	4891.6	4887.9	0.08	675.5	7870.6	7870.6	0.00
552.0	5108.7	5102.2	0.13	702.4	8513.9	8507.6	0.07
569.4	5478.0	5478.4	-0.01	753.0	9751.2	9744.3	0.07
579.1	5712.2	5689.9	0.39	784.7	10541.7	10546.7	-0.05
Cs_2MnO_4							
469.5	3286.1	3294.6	-0.26	648.6	7188.2	7181.7	0.09
499.2	3895.7	3908.4	-0.33	678.9	7896.2	7882.9	0.17
528.4	4523.3	4523.9	-0.01	708.9	8587.3	8589.6	-0.03
558.2	5166.2	5164.2	0.04	739.0	9322.1	9311.0	0.12
588.3	5829.3	5823.5	0.10	769.5	10049.4	10054.5	-0.05
618.5	6486.0	6497.5	-0.18	798.9	10775.0	10783.2	-0.08

TABLE 5. Standard molar thermodynamic functions of Cs₂RuO₄ and Cs₂MnO₄ ($R = 8.31451 \text{ J} \cdot \text{K}^{-1} \cdot \text{mol}^{-1}$)

$\frac{T}{\text{K}}$	$\frac{C_{p,m}^\circ}{R}$	$\frac{\Delta_0^{\ddagger} S_m^\circ}{R}$	$\frac{\Phi_m^\circ}{R}$	$\frac{\Delta_{298.15 \text{ K}}^{\ddagger} H_m^\circ}{R \cdot \text{K}}$	$\frac{\Delta_f H_m^\circ}{R \cdot \text{K}}$	$\frac{\Delta_f G_m^\circ}{R \cdot \text{K}}$
Cs ₂ RuO ₄						
298.15	18.779	31.640	31.640	0	-116014	-103604
300	18.802	31.756	31.640	35	-116012	-103527
400	19.828	37.312	32.391	1969	-116371	-99220
500	20.859	41.844	33.841	4001	-116137	-94957
600	22.180	45.758	35.508	6150	-115796	-90751
700	23.852	49.300	37.229	8450	-115318	-86613
800	25.244	52.576	38.945	10905	-114705	-82553
900	26.635	55.630	40.631	13499	-113978	-78576
906.8	26.730	55.830	40.744	13680	-115034	-78365
906.8	27.730	56.199	40.744	14015	-114699	-78365
1000	28.026	58.877	42.310	16566	-129105	-73857
1100	29.418	61.613	43.942	19438	-127944	-68387
1200	30.809	64.232	45.524	22450	-126664	-63029
1211.8	30.973	64.535	45.708	22814	-126506	-62411
Cs ₂ MnO ₄						
298.15	17.949	28.770	28.770	0	-142883	-129495
300	17.978	28.881	28.771	33	-142883	-129412
400	19.472	34.261	29.494	1907	-143336	-124762
500	20.887	38.759	30.909	3925	-143162	-120135
600	22.270	42.690	32.551	6083	-142870	-115556
700	23.639	46.225	34.256	8379	-142465	-111035
800	25.000	49.471	35.958	10811	-141950	-106580
900	26.357	52.494	37.629	13379	-141331	-102196
1000	27.710	55.341	39.259	16082	-157176	-97015
1051.9	28.412	56.761	40.088	17538	-156653	-93925
1051.9	28.412	56.828	40.088	17609	-156582	-93925
1100	29.062	58.113	40.848	18992	-156066	-91048
1176.7	30.098	60.106	42.038	21260	-155189	-86561

plotted against T ; the sets of results join smoothly at $T = 298.15 \text{ K}$. No previous measurements on the two compounds are found in the literature.

The smoothed thermodynamic quantities of Cs₂RuO₄ and Cs₂MnO₄ at selected temperatures from 298.15 K to the solid-to-liquid phase-transition temperature are listed in table 5. The formation properties were calculated from the enthalpies of formation of Cs₂RuO₄ and Cs₂MnO₄: $\Delta_f H_m^\circ(298.15 \text{ K}) = -(964.6 \pm 5.3) \text{ kJ} \cdot \text{mol}^{-1}$,⁽¹⁾ and $-(1188.0 \pm 2.0) \text{ kJ} \cdot \text{mol}^{-1}$,⁽⁷⁾ respectively. Auxiliary quantities for the reference states of caesium, ruthenium, and oxygen were taken from Cordfunke and Konings,⁽⁸⁾ and of manganese from reference 9.

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