High Temperature Enthalpy and Decomposition Pressures of RuS$_2$

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An adiabatic drop-calorimeter has been used to measure the enthalpy of RuS$_2$(s) in the temperature interval 374 – 1480 K* relative to 298.15 K. The enthalpies obtained can be represented by the equation:

$$[H°(T) - H°(298 \text{ K})]/\text{cal}_{th} \text{ mol}^{-1} =$$
$$19.819(T/\text{K}) - 0.3699 \times 10^{-2}(T/\text{K})^2$$
$$+ 0.77887 \times 10^{-4}(T/\text{K})^{5/2} + 241\,400(\text{K}/T) - 6509.$$

The decomposition pressures according to the reaction

$$\text{RuS}_2(s) = \text{Ru}(s) + \text{S}_2(g)$$

were measured with a silica spiral gauge and Knudsen effusion cells. A second-law treatment of the pressure data gave for the standard enthalpy of formation according to the reaction

$$\text{Ru}(s) + 2\text{S}(rh) = \text{RuS}_2(s)$$
$$\Delta H_f°(298 \text{ K}) = (-47.6 \pm 0.2) \text{ kcal}_{th} \text{ mol}^{-1}$$

and a standard entropy for the compound

$$S°(298 \text{ K}) = (13.2 \pm 0.5) \text{ cal}_{th} \text{ K}^{-1} \text{ mol}^{-1}.$$

The uncertainty is the standard deviation from the mean.

Even if most of the sulfides of the platinum metals have been studied by tensimetric methods, the treatment of the pressure data has been based on estimated high-temperature heat-capacities. This is also the case for RuS$_2$. The decomposition pressures for the compound, according to the reaction: RuS$_2$(s) = Ru(s) + S$_2$(g), were first measured by Juza and Meyer[1] in the temperature interval 1396 – 1481

K. These authors estimated the standard enthalpy of formation for the compound at room temperature to -42 kcal$_{th}$ mol^{-1}. Reviewers have later used different heat-capacity estimates and concluded with more negative enthalpy values: Kelley:[2] -46.99 kcal$_{th}$ mol^{-1}, Rossini et al.:[3] -48.1 kcal$_{th}$ mol^{-1}, Westrum et al.:[4] -53.0 kcal$_{th}$ mol^{-1}. Kelly[2] and Westrum et al.[4] have also estimated the standard entropy for the compound and have proposed 12.5 cal$_{th}$ K^{-1} mol^{-1} and 10.4 cal$_{th}$ K^{-1} mol^{-1}, respectively, as the most probable value at 298 K. The standard enthalpy and entropy estimates have been based on four pairs of pressures which were the only measurements reported by Juza and Meyer.

In the present study, the decomposition pressure measurements were repeated and the new data together with the experimentally determined heat-capacities were employed in the evaluation of the thermodynamic properties of RuS$_2$.

RuS$_2$ is the only compound formed in the ruthenium-sulfur system. It has pyrite-type structure[5] and it has diamagnetic[6] properties.

EXPERIMENTAL

Stoichiometric amounts of ruthenium powder (purity: 99.99 mass %) and sulfur (purity: 99.999 mass %) from Koch-Light Lab. were weighed into a long silica ampoule to form RuS$_2$. After evacuation and sealing of the ampoule, the elements were brought to the reaction temperature. To prevent explosion during the reaction, half of the ampoule was heated to 673 K and the other half where the metal was kept, was maintained at 1173 K. The reaction did not go to completion and weighing of the unreacted sulfur showed that the end product corresponds to the gross composition RuS$_{1.900}$, (0.05 Ru + 0.95 RuS$_2$). For enthalpy measurements by means of the drop-calorimeter, 7.1021 g RuS$_{1.900}$ was sealed in a silica ampoule

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* Throughout this paper temperatures are in IPTS-68; 1 atm = 760 Torr = 101325 Pa, 1 cal$_{th}$ = 4.183 J.

(mass 3.5611 g) under argon at a pressure of 150 Torr. The ampoule was placed in a close-fitting container (mass 24.2866 g) made of Pt + 10 mass % Rh. An identical ampoule without $RuS_{1.900}$ was sealed for empty drops. The calorimeter which is of the aneroid type with adiabatic shields, has been described elsewhere.[7]

The decomposition pressures were measured with two effusion cells and a silica spiral gauge of Bodenstein type.

The effusion cells were made of silica. They were calibrated with silver as a standard.[8,9] During effusion both cells were placed in a horizontal silica tube, contained in a three-zone furnace which has an axial temperature gradient less than 0.5 K along the central zone of 6 cm. The silica tube was connected to vacuum pumps, the system evacuated and the furnace positioned around the tube after having been brought up to the desired temperature. The pressure was always maintained below 5×10^{-6} Torr. The furnace was kept within ± 0.5 K of the set point temperature by means of a Leeds and Northrup M-Line Model C temperature controller. The cell temperature was measured by a calibrated Pt-to-(Pt + 10 mass % Rh) thermocouple placed in a pocket close to the cells in the silica tube. The uncertainty in the temperatures for the effusion-runs is ± 1.0 K. The weight losses of the cells were determined at room temperature by means of a microbalance. Thirteen weight-loss runs with $RuS_{1.900}$ were performed in the temperature interval 1073 – 1150 K.

The experimental arrangement for the Bodenstein manometer mainly follows the description given by Biltz and Juza.[10] The furnace temperature was kept within ± 0.5 K of the set point temperature by means of a Eurotherm LP96-PID controller, and the sample temperature measured by means of a calibrated Pt to (Pt + 10 mass % Rh) thermocouple with an uncertainty of ± 1.0 K. The sample container was charged with two different amounts of $RuS_{1.900}$ and eleven sets of manometer measurements with decreasing sulfur content were performed, giving a total of 113 single measurements in the temperature interval 1373 – 1505 K.

X-Ray diffraction photographs were taken with a 19 cm diameter Unicam high-temperature camera with copper radiation. The samples were sealed in thin-walled silica capillaries. The temperatures given represent the sample temperatures within ± 3 K.

SULFUR VAPOUR

Sulfur vapour can contain all the eight molecular species from S_1 to S_8. The relative amount of the different species is dependent on temperature and pressure. At high temperature and low pressure practically all sulfur gas exists as S_2 molecules. Reactions with sulfur gas will ordinarily be referred to the S_2 molecule as the reacting species. It is therefore necessary to know besides the exact thermodynamic properties of the molecule, the equilibrium constants which connect the molecule with the other species in the vapour, in order to calculate the partial pressures. In recent years new molecular constants for $^{32}S_2$ and $^{34}S_2$ have been published,[11] and Table 1 contains the constants recalculated to natural molecular weight. By means of statistical mechanics calculations,[12] the thermodynamic functions shown in Table 2 were obtained. A least square treatment of the heat-capacities gave the following expression valid in the temperature interval 298 – 1500 K:

$$C_p(S_2,g/cal_{th} \ K^{-1} \ mol^{-1}) = 8.015 + 1.43966 \times 10^{-3}.(T/K) - 1.43237 \times 10^{-5}(T/K)^{3/2} - 52\,858(K/T)^2 \quad (1)$$

aver. real. dev. ± 0.1 %.

By means of the recommended value[9] for standard enthalpy of formation for $S_2(g)$: $\Delta H_f°(298 \ K) = (30710 \pm 70)cal_{th} \ mol^{-1}$, estimated[13] heat-capacities of the molecular species S_3 to S_8, and the saturated vapour pressures of sulfur,[14–22] the equilibrium constants as a function of temperature for the reactions
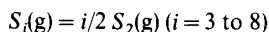
$$S_i(g) = i/2 \ S_2(g) \ (i = 3 \text{ to } 8)$$

Table 1. Molecular constants for $S_2(g)$, M = 64.12.

State	g	$\dfrac{T_e}{cm^{-1}}$	$\dfrac{\omega_e}{cm^{-1}}$	$\dfrac{X_e\omega_e}{cm^{-1}}$	$\dfrac{B_e}{cm^{-1}}$	$\dfrac{\alpha_e}{cm^{-1}}$
$^3\Sigma$ X0	1	0	724.65	2.836	0.2946	0.157×10^{-2}
$^3\Sigma$ X1	2	23.68	724.65	2.836	0.2946	0.157×10^{-2}
$^1\Delta$	2	4500	701.36	3.08	0.2915	0.169×10^{-2}
$^1\Sigma$	1	8500	699.80	3.39	0.2915	0.169×10^{-2}

Table 2. Thermodynamic functions for S$_2$(g).

$\dfrac{T}{K}$	$\dfrac{C_p}{cal_{th} K^{-1} mol^{-1}}$	$\dfrac{H(T)-H^\circ(298\ K)^a}{cal_{th}\ mol^{-1}}$	$\dfrac{S^\circ(T)}{cal_{th}\ K^{-1}\ mol^{-1}}$	$\dfrac{-[G^\circ(T)-H^\circ(298\ K)]/T}{cal_{th}\ K^{-1}\ mol^{-1}}$
298.15	7.765	0	54.507	54.507
300	7.773	14	54.555	54.507
400	8.147	812	56.846	54.817
500	8.391	1639	58.692	55.413
600	8.553	2487	60.237	56.092
700	8.670	3349	61.565	56.781
800	8.763	4220	62.729	57.453
900	8.846	5101	63.766	58.098
1000	8.925	5989	64.702	58.712
1100	9.004	6886	65.556	59.296
1200	9.083	7790	66.343	59.851
1300	9.160	8702	67.073	60.379
1400	9.237	9622	67.755	60.882
1500	9.310	10550	68.395	61.361

^{a} H° (298 K) $- H^\circ$ (OK) $= 2184\ cal_{th}\ mol^{-1}$.

Table 3. Equilibrium constants for the reaction $S_i(g)=i/2\ S_2(g)$ $\log_{10}(K_i/atm^{i/2-1})=-A(K/T)-B\log_{10}(T/K)+C\times 10^{-3}(T/K)+D\times 10^{-5}(T/K)^{3/2}+E(K/T)^2+F$.

i	A	B	C	D	E	F
1	-11026.1	0.851147	0.204410	0.150571	-4899.3	-0.4872
3	3079.9	0.418352	0.122330	0.125211	8317.0	4.5504
4	6417.8	1.54076	0.229068	0.166951	19263.0	11.7034
5	11852.3	2.77790	0.365644	0.208691	26767.0	21.7312
6	15656.6	3.79161	0.458834	0.250430	30752.3	28.5186
7	18588.7	4.52151	0.483621	0.292169	31393.7	33.5440
8	22596.1	5.33896	0.535288	0.333907	32734.7	41.8449

were calculated. A general least squares programme was used which gave the values for the constants in the equation

$$\log_{10}(K_i/atm^{i/2-1}) = \\ -A(K/T)-B\log_{10}(T/K)+C\times 10^{-3}(T/K)+ \\ D\times 10^{-5}(T/K)^{3/2}+E(K/T)^2+F$$

shown in Table 3. For completeness the table also contains the equilibrium data for the dissociation of the S$_2$(g) molecule. The Gibbs energy function for the solid and liquid state of sulfur was calculated from C$_p$ data of West[23] with a minor change by using the CODATA recommended value[9] for the 298 K entropy of solid sulfur (Table 4).

Table 4. Gibbs energy function for solid and liquid sulfur.

$\dfrac{T}{K}$	$\dfrac{-[G^\circ(T)-H^\circ(298\ K)]/T}{cal_{th}\ K^{-1}\ mol^{-1}}$
298.15	7.661
368.54	7.777
374.15	7.798
388.36	7.853
400.	7.933
500.	8.699
600.	9.519
700.	10.302
800.	(11.030)
840.	(11.304)

RESULTS

Table 5 presents the empty ampoule enthalpies which can be expressed by the equation:

$$[H°(T) - H°(298\ \text{K})]/\text{cal}_{\text{th}} =$$
$$1.8046(T/\text{K}) + 9.63341 \times 10^{-5}(T/\text{K})^2$$
$$+ 40\ 795(\text{K}/T) - 677.87 \qquad (2)$$

Average relative deviation from the equation is $\pm 0.15\ \%$, and standard deviation from the mean for a single drop is $\pm 1.6\ \text{cal}_{\text{th}}$. By subtracting the values given by this equation from the gross values of the sample drops at the temperatures of the sample drops, the net enthalpies were obtained. These values were corrected for the enthalpy due to the ruthenium content in the sample by means of Kelley's[24] enthalpy equation for Ru:

$$[H°(T) - H°(298\ \text{K})]/\text{cal}_{\text{th}}\ \text{mol}^{-1} =$$
$$5.25(T/\text{K}) + 0.175 \times 10^{-3}(T/\text{K})^2 - 1632. \qquad (3)$$

This equation has been assumed valid to 1600 K because the alleged allotropy[25,26] for ruthenium has not been confirmed by other investigators.[27]

The enthalpy values per mole for RuS_2 were treated by a least squares computer programme[28] to find the best fit to an equation. The following expression was found:

Table 5. Enthalpies of empty ampoule.

$\dfrac{T}{\text{K}}$	$\dfrac{H(T) - H°(298\ \text{K})}{\text{cal}_{\text{th}}}$	$\dfrac{T}{\text{K}}$	$\dfrac{H(T) - H°(298\ \text{K})}{\text{cal}_{\text{th}}}$	$\dfrac{T}{\text{K}}$	$\dfrac{H(T) - H°(298\ \text{K})}{\text{cal}_{\text{th}}}$
374.5	120.39	675.9	646.05	1072.7	1406.16
374.5	119.90	675.9	645.45	1272.8	1805.01
374.5	120.42	873.6	1018.18	1272.6	1805.61
374.5	120.12	874.7	1020.05	1272.6	1805.56
475.0	285.01	874.6	1019.33	1272.4	1805.76
473.7	286.25	873.8	1018.06	1479.6	2233.19
473.5	285.80	1073.3	1412.47	1481.0	2233.19
473.8	285.49	1073.6	1411.90	1480.6	2232.64
676.5	647.87	1073.1	1408.00	1480.6	2231.93
676.5	647.27				

Table 6. Molar enthalpies of RuS_2. Δ is the percentage relative deviation from eqn. (4).

$\dfrac{T}{\text{K}}$	$H(T) - H°(298\ \text{K})$ $\text{cal}_{\text{th}}\ \text{mol}^{-1}$			$\dfrac{T}{\text{K}}$	$H(T) - H°(298\ \text{K})$ $\text{cal}_{\text{th}}\ \text{mol}^{-1}$			$\dfrac{T}{\text{K}}$	$H(T) - H°(298\ \text{K})$ $\text{cal}_{\text{th}}\ \text{mol}^{-1}$		
	obs.	calc.	Δ		obs.	calc.	Δ		obs.	calc.	Δ
374.5	1257	1250	0.5	774.3	8198	8230	−0.4	1075.1	13704	13699	0.1
374.5	1220	1250	−2.5	774.3	8210	8230	−0.3	1075.2	13659	13701	−0.3
374.5	1254	1250	0.3	774.5	8185	8234	−0.6	1175.7	15552	15576	−0.2
374.5	1235	1250	−1.2	774.7	8211	8237	−0.3	1176.2	15578	15585	−0.1
374.8	1253	1255	−0.2	874.4	10046	10029	0.2	1176.2	15486	15585	−0.6
473.7	2973	2939	1.1	874.5	10064	10031	0.3	1176.4	15549	15589	−0.3
473.7	2931	2939	−0.3	874.8	10037	10037	0	1273.7	17419	17432	−0.1
473.7	2953	2939	0.5	874.8	10037	10037	0	1273.9	17442	17436	0.1
473.8	2932	2941	−0.3	973.6	11904	11832	0.6	1274.1	17443	17440	0.1
575.2	4702	4705	−0.1	973.6	11913	11832	0.7	1276.1	17375	17478	−0.6
575.2	4704	4705	−0.1	973.9	11869	11837	0.3	1386.5	19588	19608	−0.1
575.2	4721	4705	0.4	974.4	11882	11847	0.3	1386.8	19653	19614	0.2
575.7	4704	4713	−0.2	976.7	11929	11889	0.3	1386.9	19625	19616	0.1
676.8	6463	6495	−0.5	1073.6	13698	13671	0.2	1386.9	19708	19616	0.5
676.8	6470	6495	−0.4	1073.9	13696	13677	0.1	1480.7	21497	21460	0.2
678.1	6496	6518	−0.3	1074.9	13700	13695	0.1	1481.4	21425	21474	−0.2

Table 7. Thermodynamic properties of RuS$_2$. M = 165.19.

$\dfrac{T}{K}$	$\dfrac{C_p}{\text{cal}_{th}\,K^{-1}\,mol^{-1}}$	$\dfrac{H°(T) - H°(298\ K)}{\text{cal}_{th}\,mol^{-1}}$	$\dfrac{S°(T) - S°(298\ K)}{\text{cal}_{th}\,K^{-1}\,mol^{-1}}$
298.15	15.90	0	0.00
300	15.93	29	0.10
400	16.91	1679	4.84
500	17.33	3394	8.66
600	17.57	5139	11.84
700	17.75	6906	14.57
800	17.93	8690	16.95
900	18.12	10492	19.07
1000	18.34	12315	20.99
1100	18.59	14161	22.75
1200	18.87	16033	24.38
1300	19.19	17936	25.90
1400	19.54	19871	27.34
1500	19.93	21844	28.70

$$[H°(T) - H°(298\ K)]/\text{cal}_{th}\,mol^{-1} =$$
$$19.819(T/K) - 0.3699 \times 10^{-2}(T/K)^2$$
$$+ 0.7788 \times 10^{-4}(T/K)^{5/2} + 241\,400(K/T) - 6509 \quad (4)$$

The average relative deviation for the experimental values from the equation is $\pm 0.35\ \%$, and the standard deviation from the mean is $\pm 37.3\ \text{cal}_{th}$ which together with the uncertainty in the empty drops, gives an overall standard deviation of $\pm 52\ \text{cal}_{th}\,mol^{-1}$. Table 6 contains the enthalpies found per mole RuS$_2$ together with the calculated values and the percentage relative deviation from the equation. The smoothed enthalpies from the equation and the derived heat capacities and entropy increments at 100 K interval are presented in Table 7.

In Fig. 1 the logarithm of the partial pressure of S$_2$(g) of the decomposition pressures is plotted as a function of reciprocal temperature. The line marked 1 represents the equation from the least squares treatment of the data:

$$\log_{10}[p(S_2)/\text{atm}] = (-16521.7 \pm 29.5)(K/T)$$
$$+ 9.6267 \pm 0.0210$$
$$(1073 - 1505\ K) \quad (5)$$

The points marked 2 in Fig. 1 are effusion pressures and the points marked 3 are silica spiral gauge pressures.

Table 8 presents the lattice constant of RuS$_2$ as a function of temperature. The lattice constant increases almost linearly with temperature and shows no transitions in the compound.
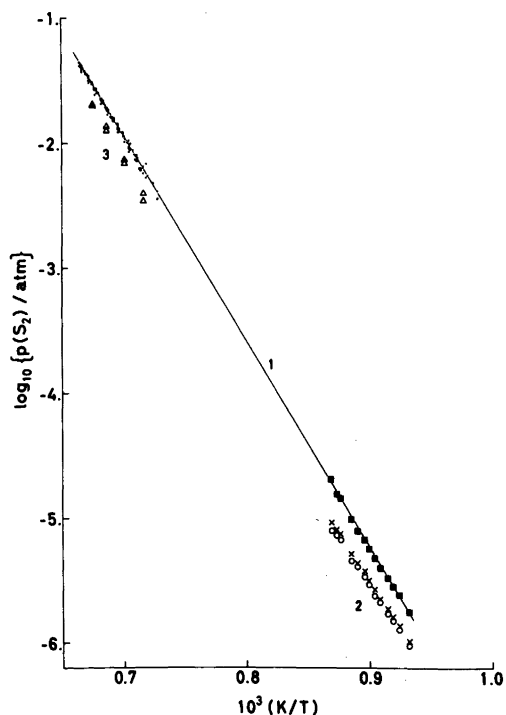
Fig. 1. Logarithm of the partial (S$_2$) decomposition pressure (atm) of RuS$_2$ as a function of inverse temperature. 1, least square line, eqn. (5). 2, uncorrected effusion pressures, cell III ($\times$), cell X (○), corrected effusion pressures (■). 3, silica spiral gauge, pressures from this study (●), pressures from Juza and Meyer (△).

Table 8. Lattice constant of RuS_2.

T/K	293	373	473	573	673	
a/nm	0.56115	0.56122	0.56167	0.56217	0.56254	
T/K	773	873	973	1073	1173	1273
a/nm	0.56290	0.56347	0.56376	0.56415	0.56471	0.56514

SECOND-LAW TREATMENT OF THE PRESSURES

To obtain the enthalpy and entropy change for the decomposition reaction of the compound, a Σ plot calculation was performed. The heat-capacity equation for Ru has been taken from Kelley;[24] for $S_2(g)$ eqn. (1) was used, and for RuS_2 the enthalpy-expression (4) has been differentiated:

$$C_p(RuS_2/cal_{th} \ K^{-1} \ mol^{-1}) =$$
$$19.819 - 0.7398 \times 10^{-2}(T/K) +$$
$$0.1947 \times 10^{-3}(T/K)^{3/2} - 241\,400(K/T)^2$$

The intergration constants ΔH_I and I were determined by the method of least squares:

$$\Delta H_I = (80557 \pm 135) \ cal_{th} \ mol^{-1}$$
$$I = (84.162 \pm 0.097) \ cal_{th} \ K^{-1} \ mol^{-1}$$

The expression for the Σ function equation is:

$$\Delta H_f(K/T) - I = -1.9872 \ln [p(S_2)/atm]$$
$$-6.5539 \ln (T/K) + 5.16883 \times 10^{-3}(T/K)$$
$$-0.557397 \times 10^{-4}(T/K)^{3/2} + 94\,271(K/T)^2 + 6.5539$$

The average relative deviation in Σ is $\pm 0.3 \%$.

By means of the standard enthalpy of formation for $S_2(g,298 \ K)$ and the enthalpy equation for the different species taking part in the reaction, the standard enthalpy of formation for RuS_2 at 298 K according to the reaction: $Ru(s) + 2S(rh) = RuS_2(s)$ is found to be $(-47.6 \pm 0.2) \ kcal_{th} \ mol^{-1}$. In calculating the standard entropy for RuS_2, Clusius and Piesbergen's[29] entropy value for ruthenium, $S°(Ru, s, 298 \ K) = (6.82 \pm 0.05) \ cal_{th} \ K^{-1} \ mol^{-1}$ was used. The derived standard entropy for RuS_2 at 298 K is $(13.2 \pm 0.5) \ cal_{th} \ K^{-1} \ mol^{-1}$. The uncertainty in the values is the standard deviation from the mean.

DISCUSSION

When calculating the constants in the enthalpy equation for the compound, the $C_p(298 \ K)$ value was changed in small steps beginning with the value 16.57 $cal_{th} \ K^{-1} \ mol^{-1}$ deduced from Neumann-Kopp's rule. The value which gives the best fit of the experimental data to the equation, is $C_p(298 \ K) = 15.90 \ cal_{th} \ K^{-1} \ mol^{-1}$. Since the calculated enthalpy values are not very sensitive to moderate changes in the $C_p(298 \ K)$ value, the values of the heat-capacities derived from the equation may be impeded with more uncertainty than the calculation shows, even if the experimentally found enthalpies are satisfactorily reproduced by the equation.

The vapour pressures calculated directly from the effusion cell data showed orifice area dependence and were therefore too low. They were corrected to equilibrium pressures by means of graphical extrapolation. The relation:[30] $p_m = p_e - C(A_o/A_b)p_m$ was used, where p_m is the measured pressure, p_e is the equilibrium pressure, A_o is the effective area of the orifice (determined experimentally), A_b is the inside area of the cell and C is a constant. A plot of $(A_o/A_b)p_m$ against p_m for the cells will when extrapolating to zero opening, give the equilibrium pressure. That the measured pressures tend to be less than the equilibrium pressures, is due to rate-determining step(s) in the decomposition reaction which slow(s) down the evaporation rate. The measured pressures can still often, limited by practical causes, be brought to a close approximation of the equilibrium pressures by using cells with large sample areas and small orifices. In the present case the ratio of the orifice area to the sample area for the cells, is not small enough to fully compensate for the slow rate-determining step(s), and an orifice area dependence is clearly shown.

The decomposition pressures measured with the silica manometer were found to be $40 - 30 \%$ higher than the pressures published by Juza and Meyer (marked 3 in Fig. 1), the difference diminishing with increasing pressure. The two sets of values seem to differ in a systematic way. There can be at least two possible explanations for this: the first is that there still could have been gaseous impurities

present during the measurements, even if precautions were taken in advance to prevent it; the second is that the adjustment of the zero-point on the scale before starting each series of measurements, could be wrong. But in eleven series one would expect this to give pressures both higher and lower than Juza and Meyer's. On the other hand a too-low adjustment of the zero-point could also be the origin of Juza and Meyer's lower pressure values.

A second-law treatment of pressure data may often lead to an improbable entropy value for a compound, because even small systematic errors in the pressures have a relatively large effect on the Σ plot constants. The standard deviations in these constants may well be small, but the numerical value of the constants may nevertheless disagree considerably with the correct one. If the entropy found has a reasonable value, then the standard enthalpy of formation for the compound will also be quite correct. In the present case the entropy found (13.2 ± 0.5) cal$_{th}$ K^{-1} mol^{-1}, has a value which seems probable when it is compared with other compounds of the same type. Grönvold and Westrum[31] have estimated both cationic and anionic entropy contributions in solid transition metal chalcogenides, and have presented a scheme which gives RuS$_2$ a standard entropy at 298 K of 13.5 cal$_{th}$ K^{-1} mol^{-1}.

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