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HEAT CAPACITY AND ENTHALPY OF UO_2 AND GADOLINIA-DOPED UO_2 *

K.C. MILLS, F.H. PONSFORD and M.J. RICHARDSON

Division of Materials Applications, National Physical Laboratory, Teddington, Middlesex TW11 0LW (Gt. Britain)

N. ZAGHINI

AGIP, Medicina, Bologna (Italy)

P. FASSINA

Fabbricazioni Nucleari, Milan (Italy)

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ABSTRACT

The effect of additions of 3 mass% and 5 mass% Gd_2O_3 on the heat capacity (C_p) and enthalpy of urania for temperatures between 298 and 1800 K has been determined using differential-scanning calorimetry and drop calorimetry. Heat capacity values of UO_2 and $U_{1-x}Gd_xO_2$ obtained in the present investigation are in good agreement with data recorded in the literature. Additions of Gd_2O_3 to UO_2 appear to lower the temperature for the commencement of the upturn in the $C_p(UO_2)$–temperature curve. Estimated heat capacities for the gadolinium-doped samples were found to be within ±3% of the measured values.

INTRODUCTION

Gadolinia-doped urania has been used as a burnable poison to control the reactivity of fuels in boiling-water reactors. It also has a potential application in pressurized-water reactors to attain extended fuel cycles at higher target burnup. Heat capacity and enthalpy data are needed for the design of fuel assemblies for reactors and this measurement programme was initiated because there are no published data available for these materials. Subsequently, Inaba et al. [1,2] have reported heat capacity data for UO_2 with Gd_2O_3 additions of 5–10% ** for temperatures between 300 and 1370 K using direct heat-pulse calorimetry. The objectives of the present investiga-

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* Dedicated to Professor E.F. Westrum, Jr., on the occasion of his 70th birthday and in honour of his contribution to calorimetry and thermal analysis.

** Throughout this paper, % refers to mass per cent.

tion were to determine the heat capacities and enthalpies of UO_2, $UO_2 + 3\%Gd_2O_3$ and $UO_2 + 5\%Gd_2O_3$ and to evaluate the reliability of the Kopp–Neumann rule for the prediction of C_p values for gadolinia-doped urania samples.

The temperature dependence of $C_p(UO_2)$ has attracted considerable attention in recent years because the slope of the C_p–T curve increases progressively above 1200 K, and is particularly marked above 1500 K. This behaviour contrasts sharply with that exhibited by most oxides in which the heat capacity approaches a constant value at high temperatures. One consequence of the anomalous behaviour of UO_2 is that the heat capacity at the melting point has a value approximately twice that of the Dulong–Petit value, which is the characteristic value for most oxides. The progressive increase in the slope of the C_p–T curve and the consequent high values of $C_p(UO_2)$ at high temperatures have been variously attributed to heat capacity contributions from (i) the formation of oxygen Frenkel pairs [3], (ii) the formation of an electron-hole pair by the reaction $2U^{4+} \rightarrow U^{5+} + U^{3+}$ [4], (iii) other forms of electronic excitation [5] and (iv) the occurrence of a Bredig-type transition at 2610 K [6–8] similar to those observed for CaF_2 and $SrCl_2$ which have similar structures to UO_2. It was anticipated that monitoring the effects on $C_p(UO_2)$ of vacancy formation (produced by Gd_2O_3 additions) might provide a valuable insight into the origin of the enhanced (or excess) heat capacity of urania.

EXPERIMENTAL

Sample preparation

The samples were prepared from weighed amounts of UO_2 and Gd_2O_3 powders, the latter having been predried for 3 h at 120 °C and screened

TABLE 1

The chemical composition and density of the samples used in enthalpy determination

Composition				Density (g cm^{-3})	Theoretical density (%)
Nominal	Actual				
	$\%Gd_2O_3$	O/U ratio			
		Before [a]	After [a]		
$UO_{2.00}$	0	2.001	2.0167	10.71	97.81
$UO_2 + 3\%Gd_2O_3$	2.96	2.0419	2.0355	10.33	95.24
	2.96	—	2.0407		
	2.98	—	2.0395		
$UO_2 + 5\%Gd_2O_3$	4.80	2.0313		9.61	89.19

[a] As determined before and after the enthalpy measurements.

through a 325 mesh sieve. The weighed powders were then mixed for 3 h in a Turbula blender and the homogeneity of the blend was checked by sampling from the top, centre and bottom and analysing for gadolinia (2.96, 2.98 and 2.98%, respectively). The samples were then pressed at a pressure of 0.9 ton cm^{-2} and granulated to a size of < 20 mesh sieve. These granules were pressed into cylindrical pellets of appropriate dimensions using a hydraulic, double-acting press at a pressure of 2.2 ton cm^{-2}. The 'green' pellets were then sintered for 4 h at 1650 °C in an atmosphere of wet Ar + 5%H_2 and specimens were cut from the sintered cylinders using a diamond wheel. The specimens for the differential-scanning calorimeter were 5 mm diam. × 1 mm and those used in drop calorimetry were 11 mm diam. × 12 mm. Details of the chemical composition and the density of the samples are given in Table 1.

Measurements

Heat capacities (C_p) were measured directly using a Perkin–Elmer differential-scanning calorimeter (DSC) Model 2 for temperatures between 298 and 1000 K. The procedure and analysis of the data obtained have been described in detail elsewhere [9]. The samples were placed in platinum crucibles and an atmosphere of argon was maintained in the apparatus. Measurements were made over temperature spans of 200 K using a heating rate of 10 K min^{-1}, and a period of 80 s was allowed for the sample to come to thermal equilibrium at the upper temperature. Heat capacity values obtained for silver with this method were within 1% and 2% of the values obtained from the $C_p(T)$ values recommended by Hultgren et al. [10] for the temperature ranges 298–800 K and 800–1000 K, respectively.

For temperatures above 1000 K it was necessary to derive C_p values by differentiation of the $(H_T - H_{298})$–T curve with respect to temperature. Enthalpy values were obtained by drop calorimetry for temperatures between 1073 and 1775 K and experimental details of the method have been reported by Andon et al. [11]. The samples were placed in crucibles of platinum with 10% rhodium and sealed under argon to prevent further oxidation. The crucibles were examined after the first "drop" to check the integrity of the weld since the gas pressure in the crucible increased considerably during heating and caused distortion of the crucible lid. Visual examinations of the welds for the formation of "pin-holes" were also carried out after every drop. The performance of the drop calorimeter was checked at periodic intervals throughout the measurement programme by determining the enthalpies of Calorimetry Conference Sapphire supplied by the National Bureau of Standards (NBS); the measured enthalpies were always within 1% of those recommended by NBS. All measurements of temperature were based on IPTS-68.

Enthalpy values, obtained by drop calorimetry, were combined with C_p values determined by DSC using a computer program and the equations giving the best fit with the experimental data were derived. The C_p–T and $(H_T–H_{298})$–T relationships were assumed to have the form shown in equations (1) and (2), respectively.

$$C_p = \mathrm{a} + \mathrm{b}T + \mathrm{c}T^2 - \mathrm{d}T^{-2} \tag{1}$$

$$(H_T - H_{298}) = \mathrm{a}(T - 298) + \frac{\mathrm{b}}{2}(T^2 - 298^2) + \frac{\mathrm{c}}{3}(T^3 - 298^3) + \frac{\mathrm{d}}{T} - \frac{\mathrm{d}}{298} \tag{2}$$

RESULTS AND DISCUSSION

Experimental details of previous studies involving the measurement of C_p and $(H_T - H_{298})$ of UO_2 and $U_{1-x}Gd_xO_2$ are summarized in Table 2.

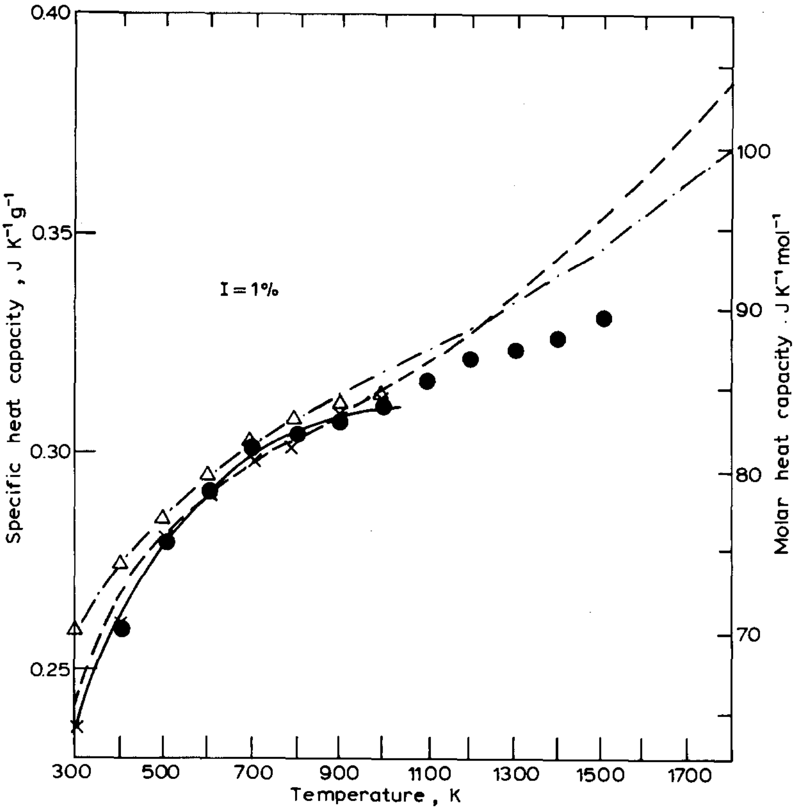

Fig. 1. Specific heat capacity for UO_2 as a function of temperature: ———, experimental (DSC) results, this investigation; - — - — -, best fit [A, see Table 4] curve; — — —, 'best fit' [B, see Table 4] curve; △, Engel [13]; ×, Gronvold [14]; ●, Inaba [2].

TABLE 2

Experimental details of the various investigations carried out on UO_2 and $U_{1-x}Gd_xO_2$, M in column three refers to metal, i.e. $(1-x)U+xGd$

Sample	Reference	Stoichiometry MO_x	Experimental details	Temp. range (K)
UO_2	Popov et al. [12]	?	C_p; dynamic pulse heating	420– 880
	Engel [13]	?	C_p; dynamic pulse heating	300–1000
	Gronvold et al. [14]	2.017	C_p; adiabatic calorimetry; sealed SiO_2 cell	300–1000
	Moore and Kelley [15]	1.98?	(H_T-H_{298}) drop calorimetry; sealed Pt + Rh crucible	470–1460
	Conway et al. [16]	?	(H_T-H_{298}) drop calorimetry; sealed crucible	1180–1175
	Hein and Flagella [17]	2.003	(H_T-H_{298}) drop calorimetry; sealed W crucible	1200–3260
	Fredrickson and Chasanov [18]	2.005	(H_T-H_{298}) drop calorimetry; sealed Ta crucible	675–1450
	Ogard and Leary [19]	?	(H_T-H_{298}) drop calorimetry; sealed crucible	1230–2630
	Affortit and Marcon [20]	(2.00–2.01)	C_p; dynamic pulse heating	1560–3075
	Leibowitz et al. [21]	2.015	(H_T-H_{298}) drop calorimetry	2550–3080
	Inaba et al. [2]	2.004	C_p; direct heating pulse calorimetry	380–1500
	This investigation	2.00	C_p; DSC (H_T-H_{298}) drop calorimetry sealed Pt crucible	298–1500 1000–1800
$U_{1-x}Gd_xO_2$	Inaba et al. [1,2]	1.998	C_p; direct heating pulse calorimetry	310–1500
	This investigation	1.953	C_p; DSC (H_T-H_{298}) drop calorimetry, sealed Pt crucible	298–1000 1000–1800

$UO_{2.00}$

The C_p results obtained in this investigation using DSC are shown as a solid line in Fig. 1 and it can be seen that they are in excellent agreement

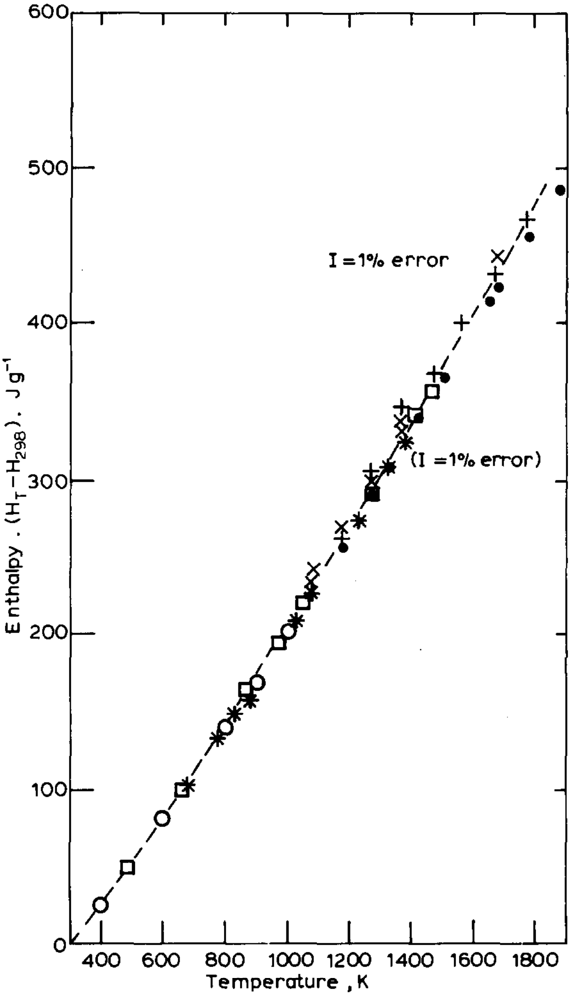

Fig. 2. The enthalpy of UO_2 as a function of temperature: this investigation, ○, DSC; +, ×, drop calorimetry, samples 1 and 2, respectively; — — —, fitted curve (B, see Table 4); □, Moore [15]; ●, Hein [17]; *, Fredrickson [18].

with the values reported by Gronvold et al. [14]. The agreement is improved when the results of Gronvold et al. are reduced by ca. 0.4% to account for differences in the ratio of oxygen to uranium in the samples. The C_p data reported by Engel [13] are some 1–2% higher than the present results: however the recent data reported by Inaba et al. [1,2] for $UO_{2.004}$ are in excellent agreement with those reported here.

The experimental enthalpy values obtained using DSC and drop calorimetry are given in Fig. 2 and Table 3. The scatter in the experimental values of ($H_T - H_{298}$) for UO_2 was greater than that usually encountered with the apparatus. This was attributed to the failure of the weld of the crucible at high temperatures which would result in oxidation of the sample and a

TABLE 3

Experimental enthalpy values obtained from drop calorimetry measurements for UO_2 and gadolinia-doped UO_2

$UO_{2.00}$		$U_{0.956}Gd_{0.044}O_{2.00}$		$U_{0.927}Gd_{0.073}O_{2.00}$	
T(K)	$(H_T - H_{298})$ (J g^{-1})	T(K)	$(H_T - H_{298})$ (J g^{-1})	T(K)	$(H_T - H_{298})$ (J g^{-1})
Sample 1		Sample 1		Sample 1	
1080.54	229.71	1061.04	233.3	1080.68	239.21
1176.02	263.03	1079.73	245.18	1171.28	262.96
1275.18	306.21	1160.33	263.76	1280.20	300.13
1370.49	346.49	1261.92	297.56	1379.00	330.57
1471.53	366.36	1362.4	333.38	1478.49	366.65
1570.03	399.95	1470.71	373.14	1579.47	401.65
1667.86	432.58	1570.03	401.25	1587.98	415.83
1773.52	466.82	1673.52	451.61	1686.27	452.17
		1775.66	487.3	1769.19	473.99
Sample 2		Sample 2			
1059.40	229.12	1064.3	231.2		
1061.04	230.71	—	—		
1079.73	243.69	1174.45	267.32		
1080.52	235.94	1275.18	301.31		
1082.16	243.93	1375.63	339.07		
1172.1	269.54	1474.31	373.55		
1270.64	299.29	1573.58	410.07		
1369.75	332.19	1672.82	439.85		
1371.22	337.53	1774.95	480.79		
1573.58	402.85				
1673.52	444.15				
		Sample 3			
		1043.49	223.4		
		—	—		
		1156.47	262.83		
		1258.38	297.71		
		1358.53	328.53		
		1459.12	364.69		
		1559.6	402.35		
		1660.07	435.95		
		1761.36	474.01		

consequent increase in both the ratio of oxygen to uranium and the heat capacity. This view was confirmed in the post-measurement analysis which indicated an increase in the oxygen to uranium ratio from 2.001 to 2.0167. Therefore, further measurements were carried out on a second specimen which was unchanged in composition after the measurements. However, there were no significant differences in the enthalpy results obtained for the two specimens because the change in composition of the first specimen would be expected to produce * an increase of c. 0.5% which is appreciably less than the experimental scatter of the measurements (±1%). Consequently it is difficult to account for the relatively large scatter in the enthalpy data obtained for UO_2, although changes in the chemical composition of the sample may have been a contributory factor. The experimental results are compared with the enthalpies recorded by Moore and Kelley [15], Hein and Flagella [17] and Fredrickson and Chasanov [18] in Fig. 2. It can be seen that the present results are in essential agreement with those reported by the other investigators, despite the fact that they are higher than those reported by Fredrickson and Chasanov [18], and Hein and Flagella [17] by 1% and 2% respectively. These are acceptable differences and are within the bounds of the combined uncertainties, namely ±1% for each set of measurements.

Heat capacities for the temperature range from 298 to 1800 K were calculated from the equation providing the 'best fit' of the temperature-dependence of the experimental C_p $(H_T - H_{298})$ data. Constants for equations (1) and (2) derived in this manner are given in Table 4. The initial fit (denoted A in Table 4) was found to overestimate the heat capacity between 298 and 600 K and consequently a second fit (denoted B) was derived by weighting in favour of the lower-temperature C_p data. It can be seen from Fig. 1 that this relationship (B) produced a good fit for the C_p data in the range from 298 to 1000 K but gave C_p values at 1500 K which are ca. 5% higher than those reported by Inaba et al. [2]. Furthermore, there were significant differences in the C_p values at high temperatures calculated using relationships A and B, whereas the corresponding differences in the predicted enthalpies (Fig. 2) were negligible. This highlights the problems encountered in deriving heat capacities by differentiation of enthalpy data. Hyland and Ohse [22] have discussed this problem in some detail and have shown that the form of expression used to describe the temperature dependence of the enthalpy can have a significant influence on the C_p values obtained **. Ralph and Hyland recommended an alternative (numerical) method for treatment of the enthalpy data [7] and this has been applied to the present results. Unfortunately, the data were too few to produce reliable values of C_p and there was an appreciable scatter in the calculated values. It was found

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* Corrections for non-stoichiometry can be made in two ways: (i) by assuming that $C_p(UO_{2+x}) = yC_p(UO_2) + (1-y)C_p(UO_{2.2})$ and using the C_p data for UO_2 and $UO_{2.2}$ reported by Gronvold et al. [14]; (ii) by assuming that C_p per g atom (where UO_{2+x} would contain $(3+x)$ g atom) is constant, thus

$$C_p(UO_2) = \left[\left(\frac{3}{3+x}\right)C_p(UO_{2+x})\right].$$

** The temperature dependence of $(H_T - H_{298})$ calculated by relationship A (see Table 4) is not shown in Fig. 2 because it was too close to curve (B) to be readily discernible.

TABLE 4

'Best fit' values for the constants a, b, c and d for the calculation of C_p and $(H_T - H_{298})$ derived from eqns. (1) and (2)

Sample	Mol. mass (g mol^{-1})	Eqn No.	C_p(J K^{-1} g^{-1})				$(H_T - H_{298})$ (J K^{-1} mol^{-1})			
			a	$-b \times 10^5$	$c \times 10^8$	$-d$	a	$-b \times 10^3$	$c \times 10^6$	$-d \times 10^3$
$UO_{2.00}$	270.1	A	0.2975	0.220	0.692	4458	80.35	0.594	1.87	12.04
		B	0.3233	4.97	4.82	6512	87.32	13.42	13.02	17.59
$U_{0.956}Gd_{0.044}O_{2.00}$	266.48 [a]	C	0.3497	9.30	7.27	7453	93.19	24.78	19.37	19.86
$U_{0.927}Gd_{0.073}O_{2.00}$	264.1 [a]	D	0.3621	12.14	8.436	8063	95.63	32.25	22.28	21.29

[a] Values have been calculated for the formula $U_{1-x}Gd_xO_2$ to aid comparison with the data of Inaba [2]; molar C_p values for $U_{0.956}Gd_{0.044}O_{1.952}$ and $U_{0.927}Gd_{0.073}O_{1.882}$ are lower than those for $U_{1-x}Gd_xO_{2.00}$ by 0.28 and 0.70% respectively.

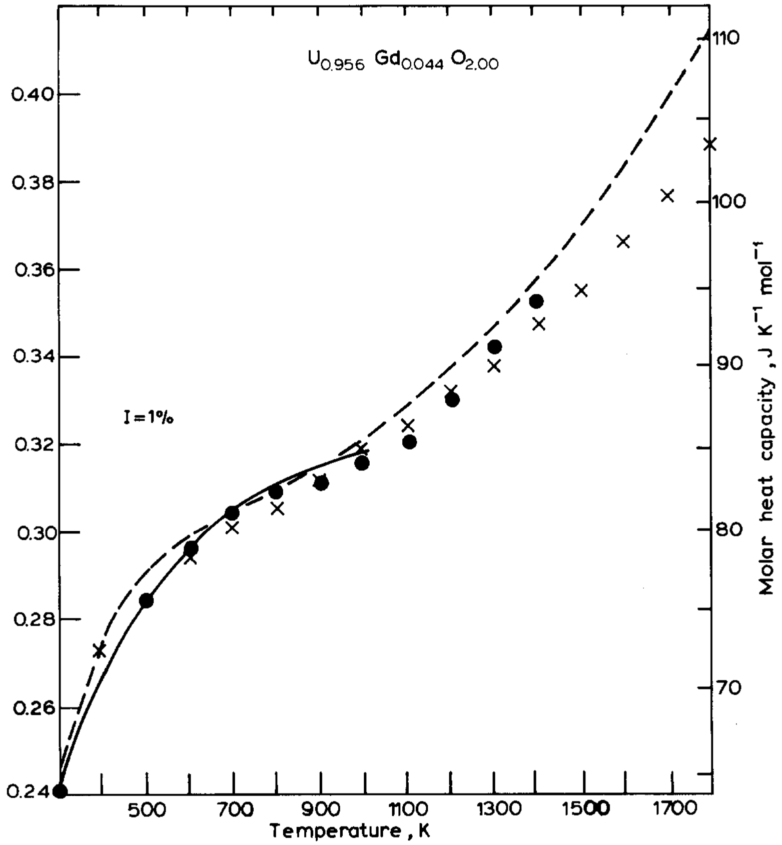

Fig. 3. The heat capacity of $U_{0.956}Gd_{0.044}O_{2.00}$ as a function of temperature; ———, experimental (DSC) results, this investigation; — — —, 'best fit' [C, see Table 4] curve; ●, Inaba [2]; ×, estimated values.

that, despite this scatter, the C_p values followed the general form of the C_p–T curve obtained with equation B (Table 4) *.

$U_{0.956}Gd_{0.044}O_{1.952}$ $(UO_{1.972} + 3\% Gd_2O_3)$

Heat capacity results obtained by DSC are shown as a solid line in Fig. 3. Inaba et al. [2] have recently reported values for $C_p(U_{0.956}Gd_{0.044}O_{2.00})$ for temperatures between 350 and 1500 K, and it can be seen that these are in excellent agreement with the DSC results obtained in the present investigation for $U_{0.956}Gd_{0.044}O_{2.00}$.

Enthalpy values obtained in the drop calorimetry experiments on three

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* $C_p(U_{0.956}Gd_{0.044}O_2 = 0.956\ C_p(UO_{2.024})+0.022\ C_p(Gd_2O_3)$

where $C_p(UO_{2.023})$ was assumed equal to (3.023/3.005) $C_p(UO_{2.005})$.

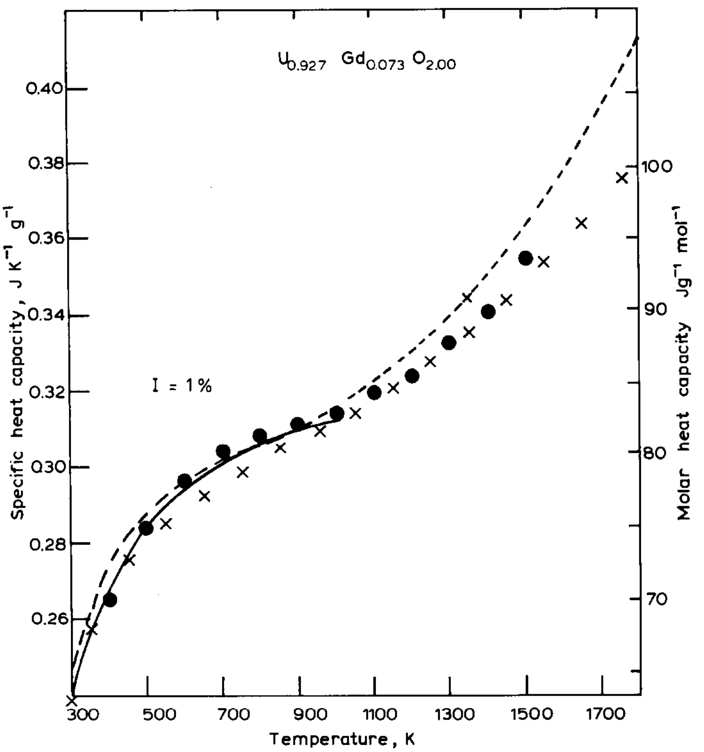

Fig. 4. The heat capacity of $U_{0.927}Gd_{0.073}O_{2.00}$ as a function of temperature: ———, experimental (DSC) results, this investigation; — — —, 'best fit' [D, see Table 4] curve; ●, Inaba [2]; ×, estimated values.

different specimens are given in Table 3 and the 'best fit' relationship (C) obtained from a fit of the enthalpy and the low temperature (DSC) C_p data is given in Table 4. It can be seen from Fig. 3 that the C_p values calculated from this relationship are in good agreement with the experimental C_p values. However, calculated C_p values are c. 2% higher than experimental values for the 400–600 K and 950–1250 K temperature ranges and these differences probably arise over the entire temperature range.

The Kopp–Neumann rule has been found to provide reliable estimates of the heat capacities of mixed oxides and therefore estimated values were calculated * from $C_p(UO_{2.00})$ given in Table 4 and the recommended values [23] for $C_p(Gd_2O_3)$. It can be seen from Fig. 3 that the estimated values are lower than the experimental values but are nevertheless within 3% of the latter.

$U_{0.927}Gd_{0.073}O_{1.883}$ ($UO_{1.922}$ + 5% Gd_2O_3)

Experimental heat capacity values for $U_{0.927}Gd_{0.073}O_{2.00}$ are given in Fig. 4 and it can be seen that these differ by less than 1% from the C_p data reported by Inaba et al. [1,2] for this composition. The enthalpy results obtained in drop calorimetry experiments are given in Table 3 and the 'best fit' relationship (D) derived from these data and the C_p values are given in Table 4. It can be seen from Fig. 4 that C_p values calculated from this relationship are in good overall agreement with the experimental C_p data although the calculated values are c. 2% higher in the temperature ranges 400–600 K and 1100–1500 K; this is believed to be due to the inadequacies of equation (1) to provide an accurate fit of the C_p data, especially for temperatures above 1000 K. Values estimated in a similar manner to those for $U_{0.956}Gd_{0.044}O_{2.00}$ were found to be slightly lower than the experimental values but were within 3% of the latter for the entire temperature range.

The effect of Gd_2O_3 upon the C_p and enthalpy of UO_2

The experimental values of C_p and $(H_T - H_{298})$ of the two doped materials, when calculated on a mass basis, were about 2% higher than those of UO_2. However, there is little difference in the heat capacity and enthalpy of the three samples when they are compared on a molar basis because the molecular masses of $U_{0.956}Gd_{0.044}O_2$ and $U_{0.927}Gd_{0.073}O_2$ are some 1.24 and 2.2%, respectively, lower than that of UO_2. However, it should be noted that these differences lie well within the combined uncertainties arising from the experimental determination of enthalpy ($\pm 1\%$) and the differentiation of the parameter. It can be seen from Fig. 5, where the molar heat capacities of the three materials are compared, that the upturn in the C_p–T curves occurred at progressively lower temperatures, T_u as x increased from zero to 0.073. This trend can also be seen in the direct measurement of C_p reported by Inaba et al. [2] for samples with similar compositions. Inaba et al. [2] derived values for the "excess" heat capacity (ΔC_p) which was defined as the difference between the experimental values at $T > T_u$ and the values obtained by extrapolation of the fit to the C_p data for temperatures below T_u. The fitted values of $\Delta C_p(T)$ were used to derive activation energies for a process which they subsequently identified as the formation of (oxygen) Frenkel defects. Gadolinia additions apparently decreased the enthalpy of formation of these Frenkel defects to a value below that derived for $UO_{2.00}$. This treatment has not been carried out in the present investigation as it was considered that the ΔC_p values could be prone to appreciable errors arising from uncertainties in C_p values derived from fits of enthalpy data.

Furthermore, the mechanisms, such as the formation of Frenkel effects, which do occur in pure, stoichiometric UO_2, may not occur or may be modified in $U_{1-x}Gd_xO_2$. It would be expected on the basis of the Law of Mass Action that the interstitial oxygen atoms present in the ternary compound would reduce the number of interstitial oxygens formed by Frenkel defects. It is possible that differences in the stoichiometry of the $U_{1-x}Gd_xO_y$ samples used in the two studies could have a significant effect

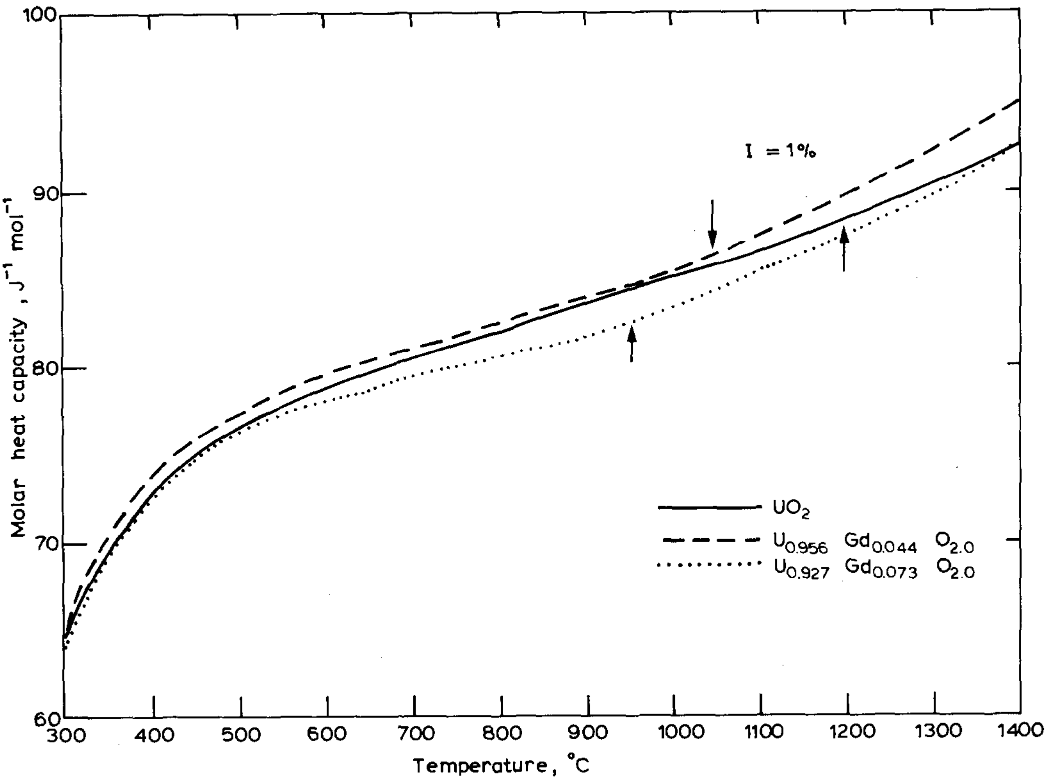

Fig. 5. Comparison of the temperature dependencies of the molar heat capacities of UO_2, $U_{0.956}Gd_{0.044}O_2$ and $U_{0.927}Gd_{0.073}O_2$ using best fit relationships B, C and D respectively (Table 4).

on the C_p–T relationships, particularly at high temperatures. In the previous study [2], the sample was stoichiometric ($y \simeq 2.00$) and consequently must contain a significant concentration of U^{5+} ions, if electrical neutrality is to be maintained in the presence of Gd^{3+} ions. In contrast to this, the sub-stoichiometric samples ($y = 1.95$–1.88) used in this investigation would contain no U^{5+} ions and, because the latter are 12% smaller than U^{4+} ions, it is possible that the concentration of U^{5+} ions could have a significant effect on the formation of Frenkel effects and consequently on the heat capacity at high temperatures.

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