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Journal of Nuclear Materials 247 (1997) 293–300

journal of nuclear materials

Section 8: Properties of other compounds

Phase diagram calculations of the U–Pu–N system with carbon and oxygen impurities

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Abstract

The most common method for the preparation of mixed nitride fuels is the carbothermic reduction of a $UO_2 + PuO_2 + C$ mixture in a nitrogen atmosphere. A mixed nitride fuel thus formed has carbon and oxygen impurities which are kept well below 5000 ppm. For a given overall composition of the nitride fuel and the temperature, the present work calculates the stable phases in equilibrium and the amounts of those phases. These calculations are based on the principle of mass balance and minimization of the Gibbs free energy for the system. The Gibbs free energy of formation of the binary compounds UN, PuN, UO, PuO, UC, PuC, UO_2, PuO_2 and $UN_{1.5}$ are used for the calculations. In general, the binary compounds are assumed to form ideal solid solutions, but in certain cases, available or estimated interaction parameters were used to see the effect of deviation from the ideal solution assumption on the phase diagram. Partial phase diagrams of (U, Pu)–C–N were drawn for different oxygen impurity concentrations at various temperatures. Comparison with the experimental data available in the literature is carried out. Based on these calculations it is suggested that a separate oxide phase has to be left deliberately in the mixed nitride fuel matrix during preparation to ensure fuel clad chemical compatibility. © 1997 Elsevier Science B.V.

1. Introduction

Mixed (uranium, plutonium) nitride is being investigated as a fuel for fast breeder reactors in order to improve the breeding performance of these reactors [1–4]. However, limited data exists on the thermophysical properties of this fuel, particularly in the composition range of interest to the reactor. A mixed mononitride fuel prepared by carbothermic reduction usually has small quantities (< 5000 wt. ppm) of carbon and oxygen impurities. Following the brief review of the relevant binary phase relations, phase diagrams of the nitride fuels, in the composition range of interest for fuel manufacture, have been calculated. Thermodynamic properties of the constituent binary compounds, available in the literature, were used to derive information on this multicomponent system.

2. Method of calculation

The general approach of the present calculation is similar to that of Potter [5], Sekar [6] and Srivastava et al. [7]. These investigators have calculated the thermodynamic properties of the U–Pu–C–N system with oxygen impurity, assuming various phase fields given in the tentative phase diagram shown in Fig. 1 [5]. In this figure, (U, Pu)CNO is a mononitride compound with dissolved carbon and oxygen impurities. Fig. 1 shows a broad composition range of stability for the phase field (U, Pu)CNO + $UN_{1.5}$. However, it does not reflect the phase field (U, Pu)CNO + (U, Pu)O_2 detected during the preparation of mixed nitride fuels [8,9]. Previous authors did not specifically calculate the phase diagrams. They first assumed a stable phase field and then calculated its thermodynamic parameters for different compositions. In the present study, the overall composition of the mixed nitride fuel was used for determining the stable phase field. The calculations were focused near the mononitride composition range of the system with

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PII S0022-3115(97)00077-9

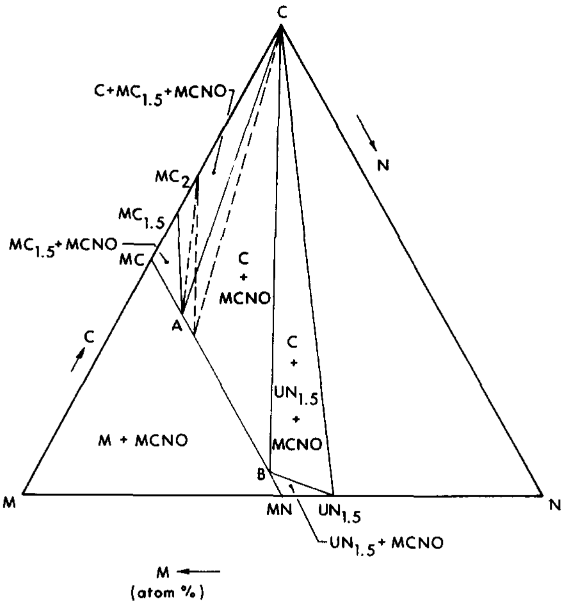


Fig. 1. Tentative phase diagram.

limited carbon and oxygen impurities. The multicomponent mixed nitride fuel, represented as (U, Pu)N(C, O), consists mainly of a mononitride phase containing dissolved oxygen and carbon impurities, (U, Pu)CNO, in equilibrium with small amounts of other phases.

2.1. *Thermodynamic data base*

2.1.1. *Solid solution behaviour*

Matsui and Ohse [10], in their study on UN–PuN, compared the calculated partial pressures based on the ideal solution assumption with experimental results and concluded that the UN–PuN system behaves like an ideal solution. Potter and Spear [11], based on the experimental work of Leitnaker [12], reported an ideal solid-solution behaviour of UC and UN over the complete composition and temperature range. For the PuC–PuN solid solution, Potter and Spear [11] expressed difficulty in assuming any particular model. Nitrogen pressures over the PuC–PuN solid solution, measured by de Franco [13], are more positive than the calculated partial pressures using an ideal solution model. However, no miscibility gap is reported in the system. Therefore, PuC–PuN is also assumed to be an ideal solid solution. Introduction of a small interaction parameter of -20.9 kJ mol^{-1} for the PuC–PuN solution, as suggested by Potter and Spear [11], was also tried. UN dissolves only a limited amount of oxygen [14–16]. The solid solution of UO and UN is ideal, or very close to ideal, within the solubility limit of 3 mol% [11]. PuN dissolves up to 14 mol% PuO [17] and shows a positive deviation from ideal solution behaviour [11]. Due to a lack of sufficient experimental data to determine the interaction parameter, the PuN–PuO system is also assumed to be an ideal solution. This assumption should be reasonably valid for low concentrations of oxygen normally encountered in the nitride fuel. The calculations were repeated using an approximate interaction parameter of 46.0 kJ mol^{-1} for the PuN–PuO solution as suggested by Potter and Spear [11].

For low concentrations of UO, PuO, UC and PuC, these compounds can be reasonably assumed to exhibit an ideal solution behaviour in a (U, Pu)N lattice. The molar Gibbs free energies of formation of the relevant binary compounds are given in Table 1 [5,11,18,19].

2.1.2. *Stoichiometry of binary compounds*

Uranium nitride exists over a narrow composition range of N:U = 0.995 to 0.997 below 1100 K [20]. Lapat et al. [21] reported a hyperstoichiometric composition of $UN_{1.03}$ at 1346 K and Benn and Bowman [22] reported a hypostoichiometric composition of $UN_{0.96}$ at 1973 K. Plutonium nitride is isomorphous with uranium nitride with a very narrow stoichiometric range [11]. Carbides of uranium and plutonium exist over a definite composition range. In the case of nitride fuel, carbon substitutes nitrogen in the lattice, therefore, these carbides can be considered as stoichiometric compounds in solution with the mononitride. The stable plutonium carbide is a nonstoichiometric compound with the upper limit of hypostoichiometry being $PuC_{0.88}$. The Gibbs free energy of formation of hypothetical PuC was derived by extrapolating the enthalpy of formation of the hypostoichiometric compound with respect to composition and using an estimated entropy value. Mono-oxides of plutonium and uranium are hypothetical compounds with oxygen present only in solution form in nitrides. For the present study, UN, UC, UO, UO_2, $UN_{1.5}$, PuN, PuC, PuO and PuO_2 have been assumed to be stoichiometric compounds.

2.1.3. *Mutual solubility of phases*

The solubility of UO in $UN_{1.5}$, is reported to be small [23]. Martin [24] reported an increase in the solubility of

Table 1
The molar Gibbs free energies of formation of the binary compounds used for the calculations as a function of temperature $\Delta G_f{}^{\circ} = A + BT + CT\ln(T)$ (J mol^{-1})

Compound	A	B	C	Ref.
PuN	−302825.0	97.28	0.0	[5]
UN	−297356.0	86.19	0.0	[5]
PuC	−57990.0	−0.75	0.0	[11]
UC	−110085.0	1.17	0.0	[18]
UO	−505758.0	94.18	0.0	[11]
PuO	−557250.0	92.34	0.0	[11]
$UN_{1.5}$	−367326.0	133.05	0.0	[5]
PuO_2	−1060560.0	219.60	−14.43	[19]
UO_2	−1082225.0	170.92	0.0	[19]

UN in UO_2 from 5 to 13 mol% as the temperature increases from 1773 to 2273 K, but below this temperature, it is considerably low. Ermolaev et al. [25] carried out a neutron diffraction analysis of uraniumoxycarbonitrides and reported a negligible solubility of nitrogen and carbon in uranium dioxide. As plutonium dioxide behaves similar to uranium dioxide, it can be assumed that the (U, Pu)O_2 phase also has a negligible solubility for (U, Pu)C or (U, Pu)N. Blum et al. [26] carried out an X-ray diffraction analysis of the U–O–N system and observed a mutual solubility of UO_2 and $UN_{1.5}$ with an eutectoid at 1343 K. For simplicity all the phases were considered as pure phases in the present calculation, i.e., the binary compound of one phase does not dissolve in the other phase.

2.1.4. Sesquinitride phase

Plutonium sesquinitride is an unstable compound and is reported to be stabilized in a $UN_{1.5}$ matrix by dissolution up to 15 mol% $PuN_{1.5}$ [27,28]. However, the thermodynamic properties of $PuN_{1.5}$ are not available and its stabilization in solution with $UN_{1.5}$ at high temperatures, particularly in the absence of high nitrogen pressure, appears doubtful. Therefore, the sesquinitride phase was assumed to be a pure uranium sesquinitride phase.

2.1.5. Ternary compound

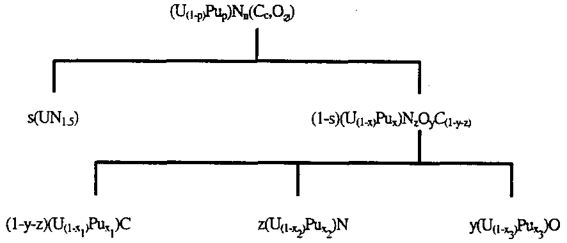
There are no ternary compounds reported for the system. Therefore, the calculation involved only the binary compounds discussed above.

2.2. Procedure

The first step in the calculation on the U–Pu–C–N–O system is to define the phase fields relevant for nitride fuel with small impurities of carbon and oxygen. These phase fields are: (U, Pu)CNO + C; (U, Pu)CNO + (U, Pu); (U, Pu)CNO + (U, Pu)O_2; (U, Pu)CNO + $UN_{1.5}$ and (U, Pu)CNO + $UN_{1.5}$ + C.

The composition of the total fuel is presented as $(U_{(1-p)}Pu_p)N_n(C_c, O_\phi)$. For each mole of the metal present in the fuel, the number of moles of uranium and plutonium are $(1-p)$ and p, respectively. The mole ratio, C/(U, Pu), N/(U, Pu) and O/(U, Pu) in this multicomponent fuel are c, n and ϕ, respectively. The mononitride phase (U, Pu)CNO can be expressed in full as $(U_{(1-x)}Pu_x)N_zO_yC_{(1-y-z)}$. The distribution of various binary compounds in a phase field was calculated using mass-balance equations and chemical equilibria. The calculation was carried out by a FORTRAN program written on the basis of these equations.

A typical example of a phase field, (U, Pu)CNO + $UN_{1.5}$, is used here to describe the present method of calculation. A similar procedure was applied for other phase fields. The mass distribution of the elements can be understood using a composition tree.



The number of moles of the sesquinitride phase in the fuel is shown as s, therefore, $(1-s)$ is number of moles of monoxycarbonitride solid-solution phase. Each mole of the monoxycarbonitride solid-solution consists of z moles of mononitride $(U_{(1-x_2)}Pu_{x_2})N$, y moles of monoxide $(U_{(1-x_3)}Pu_{x_3})O$ and $(1-y-z)$ moles of monocarbide $(U_{(1-x_1)}Pu_{x_1})C$. The overall fuel composition parameters, i.e., p, n, c and ϕ are known. To determine the phase compositions, the unknown parameters to be calculated are s, y, z, x_1, x_2 and x_3. To determine these unknown parameters, independent relations were defined using mass balance and chemical equilibria. The mass balance relation for the elements can be expressed as

$$c=(1-s)(1-y-z) \quad \text{for carbon,} \tag{1}$$

$$y=\phi/(1-s) \quad \text{for oxygen,} \tag{2}$$

$$z=(n-1.5s)/(1-s) \quad \text{for nitrogen,} \tag{3}$$

$$p=(1-s)(x_1(1-y-z)+x_2z+x_3y) \quad \text{for plutonium.} \tag{4}$$

For the known non-metals to metal ratios, Eqs. (1)–(3) were solved together to calculate the number of moles of uranium sesquinitride 's' in the fuel. This value was substituted into Eqs. (2) and (3) to calculate the non-metal to metal ratio in the monoxycarbonitride solid-solution (U, Pu)CNO.

Two more relations were derived from the following independent chemical equilibria:

$$[[UC]_{PuC}]_{UNO}+[[PuN]_{UN}]_{PuCO} \Leftrightarrow [[UN]_{PuN}]_{UCO}+[[PuC]_{UC}]_{PuNO}, \tag{5}$$

$$[[PuC]_{UC}]_{PuNO}+[[UO]_{PuO}]_{UCN} \Leftrightarrow [[UC]_{PuC}]_{UNO}+[[PuO]_{UO}]_{PuCN}. \tag{6}$$

The square bracket indicates the solid solution, e.g., $[[PuC]_{UC}]_{PuNO}$ means the compound PuC dissolved in UC which is in solution with plutonium monoxynitride. At equilibrium the molar Gibbs free energy of the reaction is zero,

$$\Delta\overline{G}_{UN}+\Delta\overline{G}_{PuC}-\Delta\overline{G}_{UC}-\Delta\overline{G}_{PuN}=0, \tag{7}$$

$$\Delta\overline{G}_{UC}+\Delta\overline{G}_{PuO}-\Delta\overline{G}_{PuC}-\Delta\overline{G}_{UO}=0. \tag{8}$$

The partial molar Gibbs free energy of a species in solution is related to its standard molar Gibbs free energy of formation, ΔG_f°. For example, the partial molar Gibbs free energy of uranium nitride in monoxycarbonitride solid solution can be expressed as follows:

$$\Delta \overline{G}_{UN} = \Delta G_f^\circ(UN) + RT \ln(a_{UN}), \tag{9}$$

where, under ideal solution assumption,

$$a_{UN} = (1 - x_2)z. \tag{10}$$

Similarly, the Gibbs free energies of formation of other binary compounds were expressed in terms of their standard molar Gibbs free energies of formation and composition. Substituting the values into Eqs. (7) and (8) gives the following expressions:

$$\Delta G_f^\circ(UN) + \Delta G_f^\circ(PuC) - \Delta G_f^\circ(UC) - \Delta G_f^\circ(PuN) + RT \ln((1 - x_2)x_1/(1 - x_1)x_2) = 0, \tag{11}$$

$$\Delta G_f^\circ(UC) + \Delta G_f^\circ(PuO) - \Delta G_f^\circ(PuC) - \Delta G_f^\circ(UO) + RT \ln((1 - x_1)x_3/(1 - x_3)x_1) = 0. \tag{12}$$

The equilibrium constants of the two chemical equilibria (Eqs. (5) and (6)) can be expressed as

$$\exp[\{\Delta G_f^\circ(UC) + \Delta G_f^\circ(PuN) - \Delta G_f^\circ(UN) - \Delta G_f^\circ(PuC)\}/RT] = A, \tag{13}$$

$$\exp(\{\Delta G_f^\circ(PuC) + \Delta G_f^\circ(UO) - \Delta G_f^\circ(UC) - \Delta G_f^\circ(PuO)\}/RT) = B. \tag{14}$$

By substituting the equilibrium constants into Eqs. (11) and (12), the following two relations were derived:

$$x_2 = x_1/(A - Ax_1 + x_1), \tag{15}$$

$$x_3 = Bx_1/(1 + Bx_1 - x_1). \tag{16}$$

From Eqs. (4), (15) and (16), the following equation containing a single unknown parameter x_1, was derived:

$$(1 - s)\{x_1(1 - y - z) + zx_1/(A - Ax_1 + x_1) + Byx_1/(1 + Bx_1 - x_1)\} = p. \tag{17}$$

The value of x_1 was calculated by the false position iteration technique (FPIT). The calculated value was substituted into Eqs. (15) and (16) to obtain x_2 and x_3. In certain cases where deviation from the ideal solution behaviour was considered, e.g., in the PuN–PuO solution, the activity of PuO was calculated as follows:

$$a_{PuO} = x_{PuO}\gamma_{PuO} = x_3 y \gamma_{PuO}, \tag{18}$$

where γ_{PuO} is the activity coefficient of PuO in solid solution. It is related to the interaction parameter $\chi_{PuN-PuO}$ by the following relationship:

$$\chi_{PuN-PuO} = RT \ln(\gamma_{PuO})/(x_{PuN})^2. \tag{19}$$

Similar relationships were derived for other phase fields, based on the mass balance and chemical equilibria, to calculate the composition of respective phases. For a given total composition of the fuel $(U_{(1-p)}Pu_p)N_n(C_c, O_\phi)$, the stable phase field was decided by mass distribution. If any

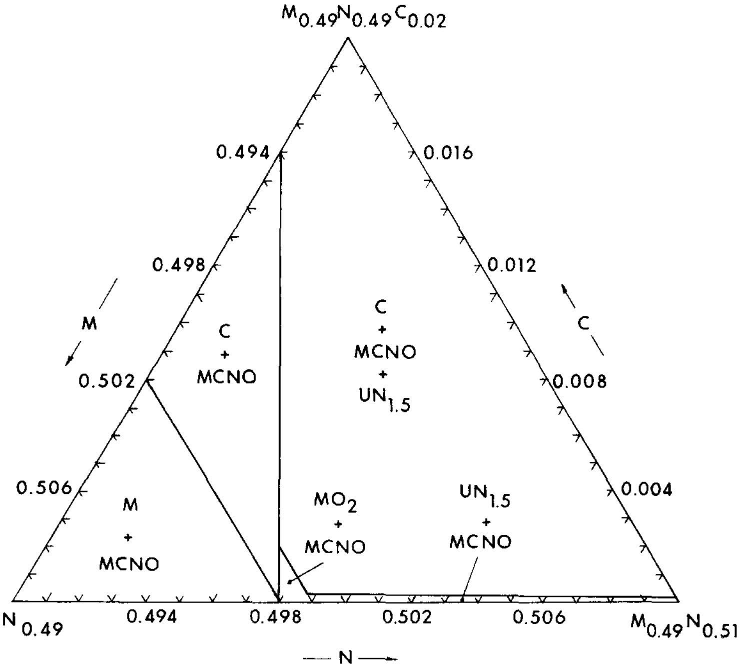


Fig. 2. Partial phase diagram of the M–C–N system at 1000 K; Pu/M = 0.2; [O] = 500 ppm.

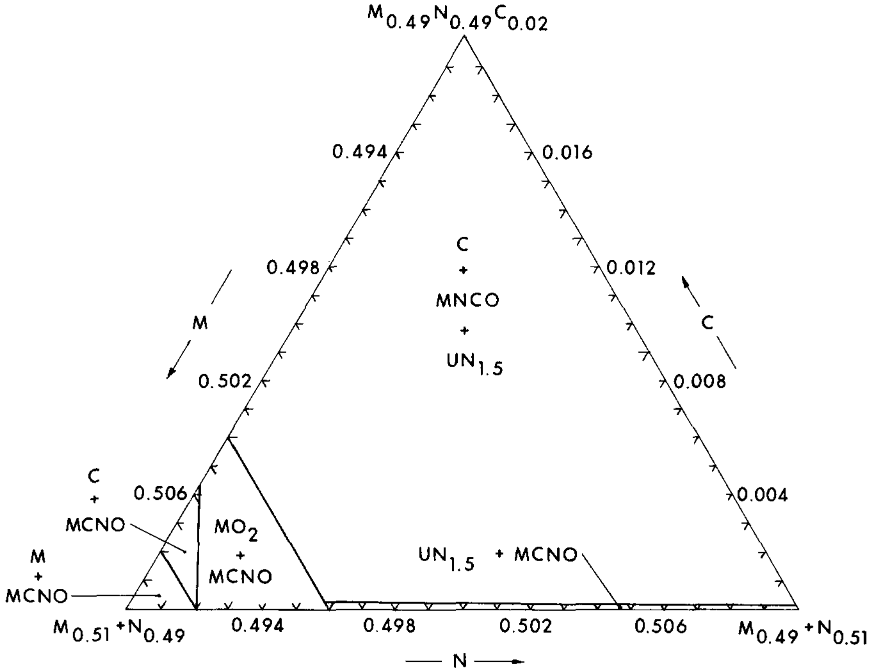


Fig. 3. Partial phase diagram of the M–C–N system at 1000 K; Pu/M = 0.2; [O] = 2000 ppm.

one of the calculated compositional parameters s, y, z, $(1-y-z)$, x_1, x_2 or x_3 was less than 0.0 or more than 1.0 then that particular phase field was considered unstable for the given composition of the fuel by the mass balance law. When more than one phase fields satisfy the mass balance relation then the total Gibbs free energies, ΔG_{tot},

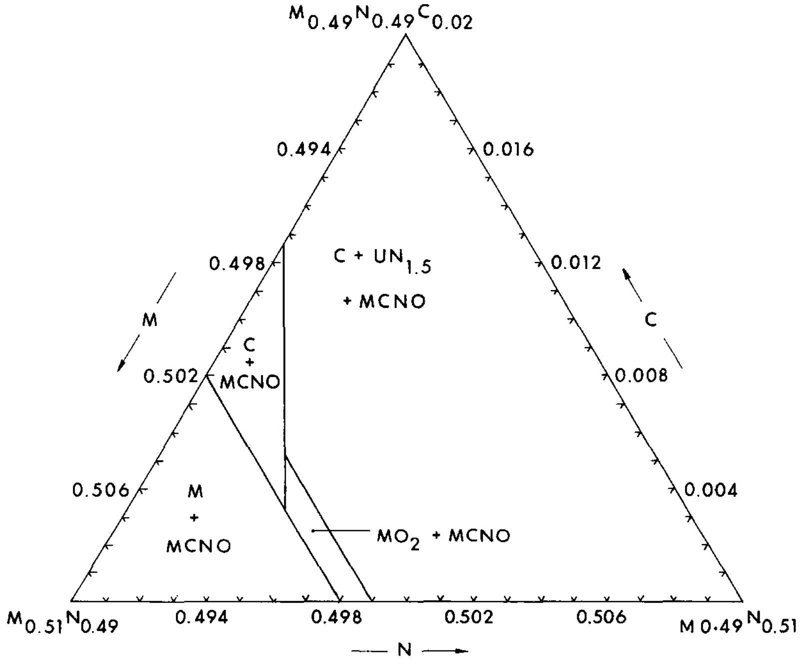


Fig. 4. Partial phase diagram of the M–C–N system at 1500 K; Pu/M = 0.2; [O] = 500 ppm.

of those phase fields were compared. The phase field with the minimum Gibbs free energy was considered stable. The total Gibbs free energy, ΔG_{tot}, of a phase field was calculated from the standard molar Gibbs free energy of formation of the binary compounds and the number of moles of that compound in the fuel. For example, in the (U, Pu)CNO + $UN_{1.5}$ phase field, it was calculated from the following:

$$\Delta G_{tot} = s\Delta G_f^{\circ}(UN_{1.5}) + (1-s) \times \left\{ x_{UC}\Delta\bar{G}_{UC} + x_{PuC}\Delta\bar{G}_{PuC} + x_{UN}\Delta\bar{G}_{UN} + x_{PuN}\Delta\bar{G}_{PuN} + x_{UO}\Delta\bar{G}_{UO} + x_{PuO}\Delta\bar{G}_{PuO} \right\}. \quad (20)$$

3. Results and discussion

$(U_{(1-p)}Pu_p)N_n(C_c, O_\phi)$ is a five component system. Therefore, even at constant temperature and pressure, a five coordinate figure is required to draw the phase diagram of this system. As this is very difficult on a two-dimensional surface, pseudo ternary phase diagrams, (U, Pu)–C–N, are drawn for simplicity by fixing the oxygen impurity level and the value of 'p'. However, Pu/(U + Pu) for various coexisting phases is different and is governed by chemical equilibria. For convenience (U, Pu) is substituted by 'M' in Figs. 1–5.

In the present work, phase diagrams were calculated for metal/nonmetal molar ratio close to unity. Some typical partial phase diagrams of the (U, Pu)–C–N system based on these calculations are given in Figs. 2–5. In these phase diagrams, oxygen impurity was varied from 500 to 2000 wt. ppm at 1000 and 1500 K. The total metal composition of the system was $U_{0.8}Pu_{0.2}$. The major phase fields in these diagrams are (U, Pu)CNO + C, (U, Pu)CNO + (U, Pu) and (U, Pu)CNO + $UN_{1.5}$ + C. Phase fields (U, Pu)CNO + $UN_{1.5}$ and (U, Pu)CNO + (U, Pu)O_2 are observed to be stable over a limited composition range. None of the major phase fields are suitable as reactor fuels as they have either free metal or free carbon, which is not desirable. Comparison of Figs. 2 and 3 and Figs. 4 and 5 indicates that the composition range of stability of (U, Pu)CNO + (U, Pu)O_2 can be increased by increasing the oxygen impurity level of the fuel. From Figs. 2 and 3 it can be seen that the carbon solubility limit in (U, Pu)CNO phase is very low in the presence of the $UN_{1.5}$ phase and carbon precipitation occurs at a very low carbon content (50 to 100 wt. ppm). It is seen from Figs. 4 and 5 that at 1500 K the phase field having the $UN_{1.5}$ phase is not stable. Introduction of the suggested interaction parameters of −20.9 kJ mol^{-1} for the PuC–PuN system and of 46.0 kJ mol^{-1} for the PuN–PuO system did not have any noticeable effect on the phase diagrams.

These phase diagram calculations were limited to low oxygen and carbon impurity ranges of a mixed nitride fuel. In these ranges the basic assumptions of present calculations were valid. The calculation of phase diagrams for a wider composition range requires much more experimental

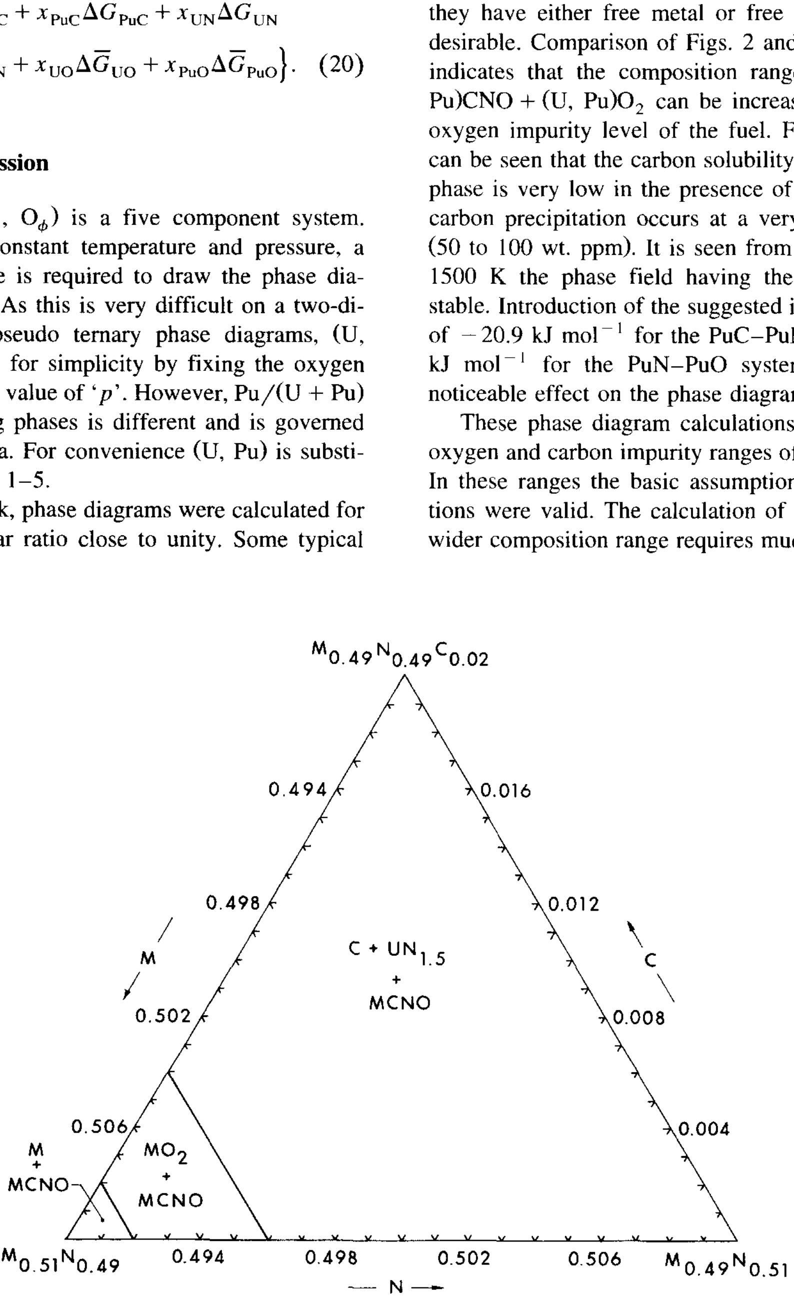


Fig. 5. Partial phase diagram of the M–C–N system at 1500 K; Pu/M = 0.2; [O] = 2000 ppm.

Table 2
Calculated compositions of the uranium nitride solid solutions for the given experimentally determined overall composition by Jain and Ganguly [29]

Total non-metal (wt%)			Phase field		Calculated composition	
[O]	[C]	N/M	exp. (a)	calc. (b)	S.S. (a)	S.S. (b)
0.28	0.123	0.96	UO_2 + S.S.	C + $UN_{1.5}$ + S.S.	$UN_{0.965}C_{0.026}O_{0.03}$	$UN_{0.952}C_{0.003}O_{0.045}$
0.45	0.127	0.95	UO_2 + S.S.	C + $UN_{1.5}$ + S.S.	$UN_{0.97}C_{0.0265}O_{0.03}$	$UN_{0.922}C_{0.003}O_{0.075}$
0.38	0.140	0.94	UO_2 + S.S.	C + S.S.	$UN_{0.954}C_{0.03}O_{0.03}$	$UN_{0.94}O_{0.06}$

S.S. (a): For the experimentally determined phase field, considering 3 at.% oxygen solubility in S.S.
S.S. (b): For the calculated phase field, considering (N + C + O)/(U, Pu) = 1.0.

data on stoichiometry, solubility and stability of the various phases. Therefore, available experimental results were evaluated and compared with each other. In Section 4 certain inconsistencies observed during these evaluations are discussed.

No experimental data on the phase diagram of (U, Pu)CNO system is available in the literature even in the limited region relevant to the use of mixed nitride fuel. The present calculation is an attempt to provide information which can be confirmed by experimental data. Jain and Ganguly [29] have carried out measurements on oxygen solubility in uranium nitride, plutonium nitride and mixed nitrides. The samples used also had a carbon impurity. In the composition studied in the present investigation, only some data on UNCO and PuCNO could be compared using the calculation method described under Section 2.2. The calculated phase fields are compared with the experimentally observed phase fields in Table 2. It can be seen that the calculated and experimental data are not in agreement. In the calculated phase field, the phases C and $UN_{1.5}$ are indicated, but the quantities may be small for phase determination by X-ray analysis. The presence of UO_2, observed experimentally, can be explained either on the basis of non-equilibrium or uncertainty in the thermodynamic data used for the calculation.

For the PuCNO system, Jain and Ganguly have confirmed the solubility of 14 mol% PuO in PuN as reported by Lorenzelli et al. [17]. However, the data reported by Jain and Ganguly could not be rationalised. For example, for a composition of $PuN_{0.23}C_{0.01}O_{0.093}$, reported by them, if apportionment of the metal and the non-metal are carried out assuming 14 mol% solubility of PuO in PuN, then 28 mol% of the PuCNO phase having a composition of $PuN_{0.82}C_{0.04}O_{0.14}$ would be present by mass law (assuming all N and C to be in this phase). Out of the total oxygen content of 0.93 mol, the oxygen present in the mononitride phase is only 0.0392 mol. The remaining 0.89 mol of oxygen should form 44 mol% of PuO_2. But then the total number of moles of plutonium present in both the phases adds up to only 0.72. Even considering the narrow nonstoichiometry of the mononitride phase, one can not account for the remaining 0.28 mol of plutonium. The absence of mass balance is difficult to explain. Hence more experimental phase diagram data are required on this system. For a mixed nitride system, the compositions studied by Jain and Ganguly had a very high oxygen content and comparison with the present calculation was not possible.

4. Conclusion

The mononitrides of plutonium and uranium are known to be completely soluble but with a very limited non-stoichiometry. To avoid metal precipitation in the nuclear fuel during the reactor operation, it is desirable to have a second phase with a higher nonmetallic content. The calculations show that the phase field (U, Pu)CNO + $UN_{1.5}$ is stable over a very narrow composition range. The threshold carbon content for the (U, Pu)CNO + $UN_{1.5}$ phase field is in the region of 50–100 wt. ppm. Uranium sesquinitride is stable only in a limited temperature range. At high temperatures it may decompose to give a high nitrogen pressure that could thermodynamically result in nitriding of the clad material. However, the present calculations indicate that a nitride fuel with (U, Pu)O_2 as second phase is relatively stable over a limited composition but wider temperature range. Therefore, in-pile thermodynamic behaviour of the nitride fuel with the (U, Pu)CNO + (U, Pu)O_2 phase field is expected to be superior to (U, Pu)CNO + $UN_{1.5}$.

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