KERNFORSCHUNGSZENTRUM

KARLSRUHE

September 1974 KFK 2049

Institut für Material- und Festkörperforschung
Projekt Schneller Brüter

Quantitative Phase Analysis in the U,Pu-C-System by X-Ray Diffraction

C. Ganguly, D. Vollath

GESELLSCHAFT
FÜR
KERNFORSCHUNG M.B.H.

KARLSRUHE

GESELLSCHAFT FÜR KERNFORSCHUNG M. B. H.
KARLSRUHE

KERNFORSCHUNGSZENTRUM KARLSRUHE

KFK 2049

Institut für Material- und Festkörperforschung
Projekt Schneller Brüter

Quantitative Phase Analysis in the U,Pu-C-System by X-Ray Diffraction

C. Ganguly[x)]
D. Vollath

Gesellschaft für Kernforschung mbH., Karlsruhe

[x)] B.A.R.C.-Bombay, India

ABSTRACT

Quantitative Phase Analysis in the U,Pu-C-System by X-Ray Diffraction

An absolute method is proposed in the following investigation for the quantitative phase analysis of mono-,di-and sesquicarbide and dioxide in the U-Pu-C-O system by X-ray diffraction.

Theoretically, the integrated intensity for the different selected lattice planes of the above phases are calculated taking into consideration the crystal structure, Lorentz Polarisation Factor, multiplicity factor, temperature factor, absorption factor and unit cell volume. For the pure phase, the intensity ratios between different planes were found comparable with the corresponding measured integrated intensity ratios, thus confirming the basis of calculation.

Using these calculated values and experimentally determining the corresponding integrated intensities of the X-ray diffraction pattern any unknown carbide pellet of 'U', 'Pu' or 'U-Pu' can be quantitatively analysed directly for mono, di, sesquicarbide and oxide phases.

KURZFASSUNG

Quantitative Phasenanalyse im U,Pu-C System mit Hilfe der Röntgenbeugung

Der vorliegende Bericht beschreibt eine absolute Methode zur quantitativen Phasenanalyse in Uran-Plutonium-Mischkarbiden durch Röntgenbeugung.

Über die Berechnung der Intensität für verschiedene Gitterebenen der interessierenden Phasen UC, U_2C_3, UC_2 und UO_2 bzw. der entsprechenden Plutoniumverbindungen, bei der die Kristallstruktur, der Lorentz-Polarisationsfaktor, der Flächenhäufigkeits- sowie der Temperatur- und Absorptionsfaktor berücksichtigt wurden, konnten Formeln für die relativen Intensitäten der einzelnen Beugungslinien angegeben werden.

Auf der Basis dieser Formeln kann nach der experimentellen Ermittlung der integrierten Intensitäten einzelner Beugungslinien der interessierenden Phasen die Konzentration dieser Gefügebestandteile ermittelt werden. Die Übereinstimmung des aus diesen Analysenwerten berechneten Äquivalentkohlenstoffgehaltes mit den chemisch ermittelten Werten ist gut.

Content

1. Introduction

2. Background of the X-ray methods for quantitative phase analysis
    - 2.1 Direct Comparison Method
    - 2.2 Internal Standard Method
    - 2.3 Active Dilution Method

3. Method Used in the Present Investigation
    - 3.1 Theoretical Calculation of the "K"-Values
        - 3.1.1 Structure Factor $/F/^2$
        - 3.1.2 Structure Factor of "UC" and (U,Pu)C
        - 3.1.3 Structure Factor of U_2C_3 and $(U,Pu)_2C_3$
        - 3.1.4 Structure Factor of UC_2 and $(U,Pu)C_2$
        - 3.1.5 Structure Factor of UO_2 and $(U,Pu)O_2$
    - 3.2 Temperature Factor
    - 3.3 Lorentz Polarisation Factor
    - 3.4 Multiplicity Factor
    - 3.5 Absorption Factor
    - 3.6 Unit Cell Volume
    - 3.7 K-Values as a Function of Plutonium Concentration

4. Experimentally Determining the Intensity Values for the Selected Lines
    - 4.1 Checking the Closeness of "K"-Ratio with the Corresponding Intensity Ratio for $(U_{.85}Pu_{.15})C$ Lines

5. Analysis of the Mixed Carbide Pellets

Table 1 - 15

1. Introduction

High metal atom density and thermal conductivity together with good irradiation stability led the physicists and engineers to consider uranium monocarbide and uranium plutonium mixed monocarbide as fuels for use in thermal and fast reactors respectively.

However, producing pure single phase (UC) or (U,Pu)C is somewhat difficult and the higher carbides UC_2, $(U,Pu)C_2$, U_2C_3, $(U,Pu)_2C_3$ are nearly always present specially in the hyperstoichiometric fuel. If too much of these phases are present then there is **possibility** of a transfer of carbon from fuel to clad giving rise to compatibility problems.

So one of the most important task in the carbide fuel quality control is to determine the exact amount of the phases present. Chemical analysis gives the amount of U, Pu free and total carbon but no information about the phases. Metallography allows the different phases present to be detected but cannot throw much light on the extent to which they are present.

Only by the X-ray diffraction method it is possible to get the precise quantitative phase analysis data.

2. Background of the X-ray methods for quantitative phase analysis /1/

The intensity of the diffraction pattern of a particular phase in a mixture of phases depends on the concentration of that phase in the mixture. To find a relation between diffracted intensity and concentration we must use the following basic equation which gives the integrated intensity of the (hkl) reflection from the phase "X"

$$I^{x}_{(hkl)} = I_o \cdot C' \cdot \{ /F/^2_{hkl} \cdot p \cdot LP \}_x \cdot \frac{T'}{V^2} \cdot A \cdot C_x \qquad (1)$$

where

$I^{x}_{(hkl)}$ = The integrated intensity of the (hkl) reflection from component "X"

(hkl) = Miller's indices of a **lattice plane**

I_o = Intensity of the incident X-ray beam

C' = A constant which is independent of the kind and amount of the diffracting substance and is equal to

$$C' = \frac{e^4 \ \lambda^3 \ A}{m^2 \ C^4 \cdot 32 \pi r} \quad \text{where}$$

e, m = charge and mass of the electron

c = velocity of light

λ = wavelength of the incident radiation

A = cross sectional area of the incident beam

r = radius of diffractometer circle

F = crystal structure amplitude and $/F/^2$ is the crystal structure factor

LP = angular or Lorentz polarisation factor

T' = temperature factor

V = unit cell volume

A = absorption factor

C_X = volume fraction of component "X"

p = multiplicity factor

For brevity we write

$$K_x = \{ /F/^2_{hkl} \cdot p \ . \ LP \ . \}_x \cdot \frac{T'}{V^2} \ . \ A$$

and

$$C = C' . \ I_o$$

Doing this we get a short and handy form of equation (1) as

$$I^x_{(hkl)} = C \ . \ K_x \ . \ C_x \qquad (2)$$

Equation (1) and (2) are the basic equations for the following analytical diffraction methods -

- direct comparison method
- internal standard technique
- active dilution method

2.1 Direct Comparison Method

If any two components 'X' and 'Y' of a multi-component mixture are considered then from equation (2) for a particular reflexion (hkl) of the 'X'-phase and (h'k'l) of the 'Y'-phase we can calculate the ratio of the intensities

$$\frac{I^{x}_{(hkl)}}{I^{y}_{(h'k'l)}} = \frac{K_{x(hkl)}}{K_{y(h'k'l)}} \cdot \frac{C_x}{C_y} \tag{3}$$

The lines (hkl) of 'X' and (h'k'l) of 'Y' should be of relatively stronger intensity and should not coincide or overlap with any line of other phases present. Further on they are called 'analytical' lines.

Thus in a mixture where 'X' and 'Y' are present, the ratio of their portion by volume is proportional to the ratio of the intensity of the reflexions chosen as analytical lines. Now if we can supply the proportionality constant 'M' given by

$$M = \frac{K_{x(khl)}}{K_{y(h'k'l)}}$$

by any suitable method we can calculate the ratio of their portion by volume. Earlier Conti, Toussaint and Vos /6/ used equation (3) for analysing a two component mixture of UC and UC_2 respectively. The ratio 'M' equal to $K_{x(hkl)} / K_{y(h'k'l)}$, they determined experimentally with mixtures of known concentrations of UC and UC_2. The relative error of their analysis for synthetic samples were two percent.

Another direct method applied by Conti and others /6/ was by measuring the intensity of a reflexion of component 'X' say UC, in the mixture and the same reflexion of the pure component 'X' i.e. pure 'UC' and then using the following equation

$$\frac{I_{UC(hkl)_{mixture}}}{I_{UC(hkl)_{pure}}} = \frac{\alpha \ \mu_1}{\{ \alpha(\mu_1 - \mu_2) + \mu_2\}}$$

where

α = weight fraction of 'UC'

μ_1 and μ_2 are the mass absorption coefficient of the component UC and UC_2 respectively

2.2 Internal Standard Method

In this method a diffraction line from the phase being determined is compared with a line from a standard substance mixed with the sample, in the form of powder, in known proportions by weight.

Next a calibration curve is made from measurements on a set of synthetic samples, containing known concentration of a particular component say 'X' and a constant concentration of a suitable standard.

After this the concentration of 'X' in an unknown sample is obtained simply by measuring the ratio $I_x/I_{standard}$ for a composite sample containing the unknown component 'X' and the same proportion of standard as was used in calibration.

For analysing the bi-component mixture of 'UC' and UC_2 Conti and others /6/ selected 'Thoria' as an internal standard. The relative error in their analysis were three percent.

However the disadvantage of this method is that it is restricted to samples in powder form. Moreover dilution of the sample takes place and it is often difficult to find a suitable internal standard, which must be free of interferences from the constituents of the sample.

2.3 Active Dilution Method

Here the sample is diluted with a known amount of the component of interest. If 'a' is the weight percentage added and I_x and I_{x+a} are respectively the intensities of a particular reflexion of component 'X' before and after adding the portion a respectively then

$$(I_x/\ I_{x+a})_{(hkl)} = \frac{x}{x+a} \tag{5}$$

This method was applied earlier /6/ for determining small amounts of UC in UC_2 samples.

This is specially suitable in the absence of a proper internal standard and for a system where all the components present have mass absorption coefficients which are close together. The disadvantage is also that it is necessary to use samples in the form of powders. The relative precision obtained by this method was three to five percent for zero to ten percent concentration range and two to three percent for concentration range from ten to hundred.

3. Method Used in the Present Investigation

In this paper the "Direct Method" is utilised. The "K" value, however is calculated from theory and not determined experimentally like earlier workers using synthetic samples. Because it is difficult to get synthetic samples of well known phase composition this particular approach to the analysis was preferred. An additional advantage of this method is that cut and ground specimen are directly used and there is no need of powdering the pellets.

The entire work can be classified under three headings.

- Theoretically calculating the "K" values of selected lines of UC, UC_2, U_2C_3, UO_2, (U,Pu)C, $(U,Pu)C_2$, $(U,Pu)_2C_3$, and $(U,Pu)O_2$. In the mixed U,Pu carbides and oxide the "K" values were calculated for uranium and plutonium in the ratio by weight of 70/30, 75/25, 80/20 and 85/15 respectively.
- Experimentally determining the intensities for these selected lines for UC, UC_2, U_2C_3 and UO_2. Since nearly pure specimen were used, the intensity ratio between two lines of a particular phase must be equal to their corresponding "K" ratio. The closeness of these ratio values were compared and used as parameters to judge the quality of the theoretical calculation.

- Using the theoretically calculated "K" values analysing quantitatively the (U,Pu)C, $(U,Pu)C_2$, $(U,Pu)_2C_3$ content of an unknown mixed carbide fuel. The "K"-ratio between different lines of the (U,Pu)C phase were compared with the corresponding intensity ratios.

3.1 Theoretical Calculation of the "K"-Values

3.1.1 Structure Factor $/F/^2$

Assuming that all the atoms in the crystal vibrate with the same amplitude the structure amplitude which is the resultant X-ray wave scattered by all the atoms of the unit cell may be written as /5/

$$F_{hkl} = \sum_{n=1}^{n=N} f_n \cdot e^{2\pi i (h U_n + k V_n + l W_n)}$$

where the summation over "n" goes over all the N atoms in the unit cell with fractional coordinates U_n, V_n, W_n and atomic scattering factors "f_n".
"F" is in general a complex number and so $/F/^2$ which comes in the intensity equation (1) is obtained by multiplying by its complex conjugate.

3.1.2 Structure Factor of "UC" and (U,Pu)C

The monocarbide phase has a NaCl type F.C.C. structure with the position of carbon and uranium atoms given in Table 1. For the mixed monocarbide the uranium position is assumed to be replaced by an average (U,Pu) atom proportional to their composition by weight. That is the distribution of the uranium and plutonium atoms are assumed to be homogeneous. The atom postion in this unit cell may be written in short, as

4 "U" atoms at 000 + face centering translations
4 "C" atoms at $\frac{1}{2}\frac{1}{2}\frac{1}{2}$ + face centering translations

For (111), which is incidentally the analytical line selected for the monocarbide.

$$F_{(111)} = 4\{f_U.e^{2\pi i(1.0+1.0+1.0)} + f_C.e^{2\pi i(1.\frac{1}{2}+1.\frac{1}{2}+1.\frac{1}{2})}\}$$

$$= 4\{f_U - f_C\}$$

Therefore F^2_{111} (monocarbide) = $16 \; (f_U - f_C)^2$

Similarly, calculating for (200), (220), (311), (422) and (511, 333) planes we get

$$F^2_{(200)} = 16\;(f_U + f_C)^2$$

$$F^2_{(220)} = 16\;(f_U + f_C)^2$$

$$F^2_{(311)} = 16\;(f_U - f_C)^2$$

$$F^2_{(422)} = 16\;(f_U + f_C)^2$$

$$F^2_{(511,333)} = 16\;(f_U - f_C)^2$$

For the mixed uranium plutonium monocarbide "f_U" is replaced by f_{U-Pu} where

$$f_{U+Pu} = f_U \cdot \text{(relative amount of "U")} + f_{Pu} \cdot \text{(relative amount of "Pu")}$$

The values of "f"$_{\text{Uranium}}$, "f"$_{\text{Plutonium}}$, "f"$_{\text{Carbon}}$ and "f"$_{\text{Oxygen}}$ are obtained from standard tables /2/ where "f" of any atom is given against its corresponding sin θ / λ values.

Details of the $/F/^2$ values thus obtained for the monocarbide phase is given in Table 2.

In choosing the atomic scattering factor value of "carbon" for all the carbides the values found by Heorniand Ibers /5/ are taken. It is assumed that in none of the carbides the carbon exist as C^{+2} or C^{+4} but as carbon atom in "UC" and as molecule C_2 in U_2C_3 and UC_2.

3.1.3 Structure factor of U_2C_3 and $(U,Pu)_2C_3$

The sesquicarbide phase is a relatively complicated B.C.C. phase with eight molecules per unit cell. The space group is $I\bar{4}3d\text{-}T_d^6$. The uranium atoms are in 16-fold positions with $\chi = 0.05$ as given in Table I . The carbon atoms are in 24-fold positions with $y = 0.295$.
That is the atomic positions of U and C are /4/

16 U atoms - $(.05, .05, .05), (.55, .45, .\overline{05}), (.\overline{05}, .55, .45),$
$(.45, .\overline{05}, .55), (.30, .30, .30), (.80, .20, .70),$
$(.70, .80, .20), (.20, .70, .80)$

24 C atoms - $(.295, 0\ \ .25), (.25, 295, 0), (0, .25, .295),$
$(.\overline{295}, .50, .25), (.25, .\overline{295}, .5), (.5, .25, .\overline{295}),$
$(.545, .5, .25), (.25, .545, .5), (.5, .25, .545),$
$(.455, .0, .25), (.25, .455, .0), (0, .25, .455)$

For (310), the selected analytical line we get for the structure amplitude

$$
\begin{aligned}
F = 2\ f_U \Big[\ & e^{2\pi i(3\cdot .05 + 1\cdot .05 + 0\cdot .05)} + e^{2\pi\, i(3\cdot .55 + 1\cdot .45 +} \\
& {}^{+\,0\cdot(-.05))} + e^{2\,\pi\, i(3\cdot(-.05) + 1\cdot .55 + 0\cdot .45)} \\
& + e^{2\pi\, i(3\cdot .45 + 1\cdot(-.05) + 0\cdot .55)} \\
& + e^{2\pi\, i(3\cdot .30 + 1\cdot .30 + 0\cdot .30)} \\
& + e^{2\pi\, i(3\cdot .80 + 1\cdot .20 + 0\cdot .70)} \\
& + e^{2\pi\, i(3\cdot .70 + 1\cdot .80 + 0\cdot .20)} \\
& + e^{2\pi\, i(3\cdot .20 + 1\cdot .70 + 0\cdot .80)}\ \Big]
\end{aligned}
$$

$$
\begin{aligned}
+\ f_C \Big[\ & e^{2\pi\, i(3\cdot .295 + 1\cdot 0 + 0\cdot .25)} \\
& + e^{2\pi\, i(3\cdot .25\ \ \ 1\cdot .295 + 0\cdot\ 0)} \\
& + e^{2\pi\, i(3\cdot\ 0 + 1\cdot\ .25 + 0\cdot .295)} \\
& + e^{2\pi\, i(3\cdot(-.295) + 1\cdot .50 + 0\cdot .25)}
\end{aligned}
$$

$$+ e^{2\pi i(3\cdot .25 + 1\cdot(-.295) + 0\cdot .5)}$$

$$+ e^{2\pi i(3\cdot .5 + 1\cdot .25 + 0\cdot(-.295)}$$

$$+ e^{2\pi i(3\cdot .545 + 1\cdot .5 + 0\cdot .25)}$$

$$+ e^{2\pi i(3\cdot .25 + 1\cdot .545 + 0\cdot .5)}$$

$$+ e^{2\pi i(3\cdot .5 + 1\cdot .25 + 0\cdot .545)}$$

$$+ e^{2\pi i(3\cdot .455 + 1\cdot 0 + 0\cdot .25)}$$

$$+ e^{2\pi i(3\cdot .25 + 1\cdot .455 + 0\cdot 0)}$$

$$+ e^{2\pi i(3\cdot 0 + 1\cdot .25 + 0\cdot .455)}]$$

Now we know $e^{i\varphi} = \cos\varphi + i\sin\varphi$
and $e^{-i\varphi} = \cos\varphi - i\sin\varphi$

Splitting all the exponential terms in terms of sine and cosine we have

$$/F/ = 2 [f_U(0 + i\ 3.804) + f_C(0 + i\ 2.65616)]$$

$$/F/^2 = 4 (3.804 f_U + 2.65616 f_C)^2$$

Similarly, we obtain the structure factor for (211), (220), (321), (332) and (510, 431)

$$/F/^2_{(211)} = 4 [(- 1.382 f_U + 2.70126 f_C)^2 + (1.902 f_U - 3.0365 f_C)^2]$$

$$/F/^2_{(220)} = 4 (5.236 f_U - 4 f_C)^2$$

$$/F/^2_{(321)} = 4 [(-1.382 f_U - 2.1432 f_C)^2 + (- 1,902 f_U - 2.30069 f_C)^2]$$

$$/F/^2_{(332)} = 4 \left[(-2\cdot.36\ f_U - 2.1432\ f_C)^2 + (3.07756\ f_U - 0.3552\ f_C)^2 \right]$$

$$/F/^2_{(510)} = 4\ (3.804\ f_U - 0.5032\ f_C)^2$$

$$/F/^2_{(431)} = 4 \left[(-\ 4.736\ f_U + 2.65616\ f_C)^2 + (0.36322\ f_U)^2 \right]$$

More details are given in Table III.

3.1.4 Structure Factor of UC_2 and $(U,Pu)C_2$

The dicarbide phase is a CaC_2 type body centred tetragonal phase. The atom positions in the unit cell given in short as /4/

2'U'atoms in (000)
4'C'atoms in 00.38, $00.\overline{38}$

For (112), the selected analytical line we calculate the structure amplitudes

$$F_{(112)} = 2 \left[f_U \cdot e^{2\pi\ i(1\cdot 0 + 1\cdot 0 + 2\cdot 0)} + f_C \{ e^{2\ \pi i(1\cdot 0 + 1\cdot 0 + 2\cdot .38)} + e^{2\ \pi i(1\cdot 0 + 1\cdot 0 + 2\cdot .\overline{38})} \right]$$

or

$$F_{(112)} = 2\{ f_U + 2\cos(2\ \pi \cdot 2 \cdot .38)\ f_C \}$$

$$= 2\ (f_U + 0.1256\ f_C)$$

$$/F/^2_{(112)} = 4\ (f_U + 0.1256\ f_C)^2$$

Similarly we get for (101), (002), (110), (114) and (312) the following structure factor

$$/F/^2_{(101)} = 4\ (f_U - 1.4579\ f_C)^2$$

$$/F/^2_{(002)} = 4\ (f_U + 0.1256\ f_C)^2$$

$$/F/^2_{(110)} = 4\ (f_U + 2\ f_C)^2$$

$$/F/^2_{(114)} = 4\ (f_U - 1.9842\ f_C)^2$$

$$/F/^2_{(312)} = 4\ (f_U + 0.1256\ f_C)^2$$

Further details are given in Table IV.

3.1.5 Structure factor of UO_2 and $(U,Pu)O_2$

The oxide phase has a CaF_2 type F.C.C. structure with atom position of uranium and oxygen in short form given as /4/

4 'U' atoms at 000 + face centering translation

8 'O' atoms at $\frac{1}{4}\frac{1}{4}\frac{1}{4}$, $\frac{3}{4}\frac{3}{4}\frac{3}{4}$ + face centering translation

For (220) which is the selected analytical line for the oxide phase we well get for the structure amplitude.

$$F_{(220)} = 4\left[f_U \cdot e^{2\pi\ i(0\cdot 2+0\cdot 2+0\cdot 0)} + f_O \cdot e^{2\pi i(\frac{1}{4}\cdot 2 + \frac{1}{4}\cdot 2 + \frac{1}{4}\cdot 0)} + f_O\ e^{2\pi i(\frac{3}{4}\cdot 2 + \frac{3}{4}\cdot 2 + \frac{3}{4}\cdot 0)}\right] = 4\ (f_U + 2\ f_O)$$

$$/F_{(220)}/^2 = 16\ (f_U + 2\ f_O)^2$$

Similarly we get the structure factor for (111), (200), (311) and (222)

$$/F/^2_{(111)} = 16\ f_U^{\ 2}$$

$$/F/^2_{(200)} = 16\ (f_U - 2\ f_O)^2$$

$$/F/^2_{(311)} = 16\ f_U^{\ 2}$$

$$/F/^2_{(222)} = 16\ (f_U - 2\ f_O)^2$$

Further details are given in Table V.

3.2 Temperature Factor /5/

The atoms in the lattice are in a continual state of thermal vibration and the amplitude of this vibration increases with temperature. This results in making the spectra weaker than they might otherwise have been, particularly in the higher orders, besides increasing the amount of incoherent scattering and increasing the background. A correction factor developed by Debye and Waller /9 / for a simple cubic lattice but which can also be used for a closed packed lattice is a follows -

$$T' = \frac{6\ h^2}{m\ k\ \Theta} \left(\frac{1}{4} + \frac{\emptyset\ (\alpha\ )}{\alpha} \right) \frac{\sin^2 \theta}{\lambda^2}$$

where m = mass of the vibrating atom

h = Planck's constant = 6.62 x 10^{-27} erg sec.

k = Boltzmann constant = 1.38 x 10^{-16} erg · A

T = absolute temperature in Kelvin

Θ = Debye temperature of the substande or the characteristic temperature

α = Θ/T

$\emptyset\ (\alpha) = 1/\alpha \int_0^{\alpha} \frac{zdz}{e^{z-1}}$, the well known Debye function.

For UC for example, the Debye temperature is 269 °K. Also the $\sin^2\theta$ values for (111) and (511, 333) are 0.0723 and 0.6510 respectively. The room temperature is assumed to be 295 °K. Putting all these values in the expression for temperature factor we have seen that the temperature factor is relatively small and hence not taken into consideration while calculating K values.

3.3 Lorentz Polarisation Factor /1/

This is the combination of certain trigonometrical factors which influence the intensity of the reflected beam. The Lorentz polarisation factor corresponding to Bragg angle θ is

$$LP = \frac{1 + \cos^2 2\theta}{\sin^2\theta \cdot \cos\theta}$$

The overall effect of these geometrical factors is to decrease the intensity of reflections at intermediate angles compared to those in forward or backward directions. The Lorentz polarisation factor as calculated for all the planes chosen for the different phases are given in the Tables II - V.

3.4 Multiplicity Factor /1/

This may be defined as the number of different planes in a form having the same spacing. The value of 'p' depends on the crystal system. Its value is higher when indices are unequal and depending upon the space group, it reaches a maximum value of 48 for the general form (hkl) in the cubic system.

Values of multiplicity factor as a function of (hkl) and crystal system (only cubic and tetragonal as all the phases under consideration belong to these two crystal system only) are given in Table VI.

3.5 Absorption Factor /2/

In caculating the integrated intensity allowance must be given for the photoelectric absorption of radiation within the crystal.

In this particular investigation the spectra emerges from the crystal on the same side as the incident beam. The specimen is in the form of a flat plate of effectively infinite thickness and makes equal angles with the incident and diffracted beams.

In such a symmetrical case of Bragg reflection the absorption factor'A'is independent of the angle and is effectively equal to $A = 1/ 2\mu$.

Where 'μ' = linear absorption coefficient of the sample. The specimens under consideration are chemical compounds. Their mass absorption coefficient is found from the following relation.

$$\left(\frac{\mu}{\rho}\right)_{UC} = \left(\frac{\mu_1}{\rho_1}\right)_U \left(\frac{A_U}{A_U+A_C}\right) + \left(\frac{\mu_2}{\rho_2}\right)_C \left(\frac{A_C}{A_U+A_C}\right)$$

$$\left(\frac{\mu}{\rho}\right)_{U_2C_3} = \left(\frac{\mu_1}{\rho_1}\right)_U \left(\frac{2\,A_U}{2A_U+3A_C}\right) + \left(\frac{\mu_2}{\rho_2}\right)_C \left(\frac{3\,A_C}{2A_U+3A_C}\right)$$

$$\left(\frac{\mu}{\rho}\right)_{UC_2} = \left(\frac{\mu_1}{\rho_1}\right)_U \left(\frac{A_U}{A_U+2A_C}\right) + \left(\frac{\mu_2}{\rho_2}\right)_C \left(\frac{2\,A_C}{A_U+2A_C}\right)$$

$$\left(\frac{\mu}{\rho}\right)_{UO_2} = \left(\frac{\mu_1}{\rho_1}\right)_U \left(\frac{A_U}{A_U+2A_O}\right) + \left(\frac{\mu_3}{\rho_3}\right)_O \left(\frac{2\,A_C}{A_U+2A_O}\right)$$

$$(\frac{\mu}{\mathcal{S}})_{U,Pu\ C} = (\frac{\mu_1}{\mathcal{S}_1})_U(\frac{A_U \cdot C_U}{A_U \cdot C_U + A_{Pu} \cdot C_{Pu} + C}) + (\frac{\mu_4}{\mathcal{S}_4})_{Pu}(\frac{A_{Pu} \cdot C_{Pu}}{A_U \cdot C_U + A_{Pu} \cdot C_{Pu} + C})$$

$$+ (\frac{\mu_2}{\mathcal{S}_2})_C(\frac{A_C}{A_U \cdot C_U + A_{Pu} \cdot C_{Pu} + C})$$

Similarly for the other mixed phases where C_U and C_{Pu} are 'U' and 'Pu' concentration,

$$(\frac{\mu_1}{\mathcal{S}_1})_U, (\frac{\mu_2}{\mathcal{S}_2})_C, (\frac{\mu_3}{\mathcal{S}_3})_O, (\frac{\mu_4}{\mathcal{S}_4})_{Pu}$$

are the mass absorption coefficient of uranium, carbon, oxygen and plutonium respectively. A_U, A_C, A_O, A_{Pu} are the atomic weights of uranium, carbon, oxygen and plutonium respectively. Putting the values of the mass absorption coefficient of U, Pu, C and O as obtained for Cu K α radiation from Ref./7/and their respective atomic weights we get the mass absorption coefficients for each compound.

Once the mass absorption $(\frac{\mu}{\mathcal{S}})$ for a particular compound is calculated the absorption factor $1/2\ \mu$ can be easily found knowing the theoretical density $\mathcal{S}$ of the compound under consideration.

The value of the absorption factor thus obtained for all the phases are given in Table II - V.

3.6 Unit Cell Volume

For a cubic crystal the unit cell volume is given by the cube of the lattice parameter

$$V = a^3$$

Where V = unit cell volume

a = lattice parameter value.

For UC, U_2C_3 and UO_2 the values of unit cell volume are as follows /3,4/

$$V_{UC} = (4.9605)^3 \cdot A^3$$

$$V_{U_2C_3} = (8.088)^3 \cdot A^3$$

$$V_{UO_2} = (5.4691)^3 \cdot A^3$$

where A = ~ 10^{-8} cm.

In case of mixed U-Pu monocarbide, sesquicarbide or oxide the lattice parameter is calculated in the following way by applying vegard's law, for example

$$a_{U,PuC} = C_{UC} \cdot a_{UC} + C_{PuC} \cdot a_{PuC}$$

and

$$V_{U,PuC} = (C_{UC} \cdot a_{UC} + C_{PuC} \cdot a_{PuC})^3$$

For the tetragonal dicarbide phase the unit cell volume is $V = a^2 \cdot c$ where 'a' and 'c' are the unit cell lattice parameter in horizontal and vertical directions respectively C_{UC} and C_{PuC} are the concentration of 'U' and 'Pu' monocarbide respectively.

3.7 K-Values as a Function of Plutonium Concentration

The 'K' values as they are defined by equation (2) were found for the U-Pu composition of 10/30, 75/25, 80/20, 85/15 for the mixed monocarbide, dicarbide, sesquicarbide and the mixed oxide.

The 'K' values are not so much affected by the variation of the plutonium content from zero to thirty percent. All the variations in 'K' values were less than one pereent. So as the influence of the plutonium content on the 'K' value was very small a linear function was choosen to describe their dependence. The 'K' values falling slightly as the plutonium content went up.

Equations of 'K' for the different lines of the different phases are given as a function of plutonium concentration in Table VII.

4. Experimentally Determining the Intensity Values for the Selected Lines

All the pellets used for these determinations were hot mounted in a resin. Fresh surface was exposed by cutting the mounted pellets into halves by the diamond tipped cut off wheels. The exposed fresh surface was then ground, polished, cleaned and finally loaded on the X-ray diffractometer.

For each plane first the diffracting angle corresponding to the maximum intensity was approximately determined. If this value is 'x'. Then the specimen was scanned from $(x - 1.5)^{o}$ to $(x + 1.5)^{o}$. In all the case a good peak with a uniform background was obtained.

The intensity corresponding to a particular diffracting angle 2θ was automatically punched on a paper tape. After the scanning, with a computer program which reads the datas from the paper tape, plots intensity versus '2θ', cuts off the background intensity by a least mean square method and finally calculates the area under the peak by normal trapezoidal method, the integrated intensity values of the different lines were obtained.

For the UO_2 and UC phase we see that there is a fairly good agreement between the K-ratio and intensity-ratio. The difference in their values being less then 10 %. For the UC_2 and U_2C_3 the K-ratio and intensity-ratio matching was quite good but not as close as compared to UO_2 and UC phase. The reason being the UC_2 sample had in it UC, U_2C_3 and UO_2 phases(as was seen from the diffraction pattern). Same with U_2C_3 pellet which had also UC, UC_2 and UO_2 in it. Now many lines of the different phases have the nearly same diffraction angle(2θ values). For example (110) of UC_2 have 2θ value 36.06^{o} which nearly overlaps with (200) of UC having 2θ value 36.19^{o}. Similarly (220) of U_2C_3 and (111) of UC have very close 2θ values of 31.25 and 31.2^{o} respectively. Also (112) of UC_2 and (220) of UO_2 have close 2θ values of 47.5^{o} and 47.2^{o}. Similarly $\left(\begin{smallmatrix}510\\431\end{smallmatrix}\right)$ of U_2C_3 and (222) of UO_2 have 2θ values of $58.1o^{o}$ and 58.4^{o} respectively. These overlapping of the phases present may be responsible for the slightly higher variation between 'K' and intensity ratio of the dicarbide and sesquicarbide phases. Table VIII, IX, X and XI gives the detail of the closeness of 'K' and 'A'-ratio (i.e. the intensity ratio).

4.1 Checking the Closeness of 'K'-Ratio with the Corresponding Intensity Ratio for $(U_{.85}Pu_{.15})C$ Lines

An unknown sample of (U,Pu)C containing good amount of $(U,Pu)_2C_3$ and low quantity of $(U,Pu)C_2$ and $(U,Pu)O_2$ was scanned to obtain the patterns for (111), (200), (220) and (311) of (U,Pu)C. The intensities of all these lines were corrected as they all overlapped with some lines of $(U,Pu)_2C_3$ or $(U,Pu)C_2$. Table X gives the detail of the lines overlapping. The corrected intensity values are obtained from the following relations

$$I^{(111)}_{(U,Pu)C}\ \text{(corrected)} = I^{(111)}_{(U,PuC)}\ \text{(measured)} - \left(\frac{K_{220}}{K_{310}}\right)_{(U,Pu)_2C_3} \cdot I^{(310)}_{(U,Pu)_2C_3}\ \text{(measured)}$$

$$I^{(200)}_{(U,PuC)}\ \text{(corrected)} = I^{(200)}_{(U,PuC)}\ \text{(measured)} - \left(\frac{K_{110}}{K_{112}}\right)_{(U,Pu)C_2} \cdot I^{(112)}_{(U,Pu)C_2}\ \text{(measured)}$$

$$I^{(220)}_{(U,Pu)C}\ \text{(corrected)} = I^{(220)}_{(U,Pu)C}\ \text{(measured)} - \left(\frac{K_{200}+K_{103}}{K_{112}}\right)_{(U,Pu)C_2} \cdot I^{(112)}_{(U,Pu)C_2}\ \text{(measured)}$$

$$I^{(311)}_{(U,Pu)C}\ \text{(corrected)} = I^{(311)}_{(U,Pu)C}\ \text{(measured)} - \left(\frac{K_{004}}{K_{112}}\right)_{(U,Pu)C_2} \cdot I^{(112)}_{(U,Pu)C_2}\ \text{(measured)}$$

The 'K'-ratio matched quite well with the intensity ratio but not as well as compared to the matching in case of the pure uranium monocarbide.

The 'K'-values of (111), (200),(220) and (311) for $(U_{.85}Pu_{.15})C$ are given in Table 12.

5. Analysis of the Mixed Carbide Pellets

Before starting analysing unknown carbide samples we must select one analytical line from each phase. The basis of choosing are as follows -

a) The line should be of relatively higher intensity.

b) Overlapping or closely adjacent lines from different phases should be avoided as far as practicable.

c) The diffracting angle to be selected must be less than 110° which is the upper limit of scanning with the existing goniometer set up.

In monocarbide all the lines are overlapping with some lines of the dicarbide,sesquicarbide or the oxide. So only on the basis of relatively stronger intensity (111) was selected as an analytical line.

For the dicarbide, sesquicarbide and oxide (112), (310) and (220) are chosen respectively as analytical line as they satisfy all the chosen criteria given above. Though (112) of dicarbide and (220) of oxide have very close 2θ values of 47.5° and 47.05° but it was possible to resolve them nicely. Now let X_{MC}, X_{MC_2} , $X_{M_2C_3}$, X_{MO_2} be the volume precent of the mixed monocarbide, dicarbide, sesquicarbide and oxide respectively. Then

$$X_{MC} + X_{MC_2} + X_{M_2C_3} + X_{MO_2} = 1 \quad \ldots \qquad (17)$$

Let the intensity value, i.e. the area of the peak under the diffraction pattern be I_{MC}, I_{MC_2}, $I_{M_2C_3}$ and I_{MO_2} respectively. Then for the (310) M_2C_3 which is a line free from overlapping from other lines,

$$I^{(310)}_{M_2C_3} = X_{M_2C_3} \cdot K^{(310)}_{M_2C_3} \quad \ldots \qquad (18)$$

Similarly for (112) MC_2 and (220) MO_2 which are also free from overlapping

$$I^{(112)}_{MC_2} = X_{MC_2} , K^{(112)}_{MC_2} \qquad (19)$$

$$I^{(220)}_{MO_2} = X_{MO_2} \cdot K^{(220)}_{MO_2} \qquad (20$$

For (111) MC which is overlapping with (220) M_2C_3

$$I_{MC^{111} + M_2C_3{}^{220}} = I^{(111)}_{MC} + I^{(220)}_{M_2C_3} \qquad (21)$$

Again comparing the intensity ratio of M_2C_3 between (310) and (220)

$$\frac{I^{(310)}_{M_2C_3}}{I^{(220)}_{M_2C_3}} = \frac{K^{(310)}_{M_2C_3}}{K^{(220)}_{M_2C_3}} \qquad (22)$$

$$I^{(220)}_{M_2C_3} = \frac{I^{(310)}_{M_2C_3} \cdot K^{(220)}_{M_2C_3}}{K^{(310)}_{M_2C_3}}$$

From equation (21) and (22) intensity contribution from (111) of MC alone will be

$$I^{(111)}_{MC} = I^{(111)}_{MC} + M_2C_3^{(220)} - \frac{I^{(310)}_{M_2C_3} \cdot K^{(220)}_{M_2C_3}}{K^{(310)}_{M_2C_3}} \qquad (23)$$

$$\frac{X_{M_2C_3}}{X_{MC}} = \frac{K^{(111)}_{MC} \cdot I^{(310)}_{M_2C_3}}{K^{(310)}_{M_2C_3} \cdot I^{(111)}_{MC}} = \frac{K^{(111)}_{MC} \cdot I^{(310)}_{M_2C_3}}{K^{(310)}_{M_2C_3} \left(I^{(111)}_{MC} + M_2C_3^{(220)} - \dfrac{K^{(220)}_{M_2C_3}}{K^{(310)}_{M_2C_3}} \cdot I^{(310)}_{M_2C_3} \right)} \qquad (24)$$

Similarly

$$\frac{X_{MC_2}}{X_{MC}} = \frac{K^{(111)}_{MC} \cdot I^{(112)}_{MC_2}}{K^{(112)}_{MC_2} \left(I_{MC}^{(111)} + M_2C_3^{(220)} - \frac{K^{(220)}_{M_2C_3}}{K^{(310)}_{M_2C_3}} \cdot I^{(310)}_{M_2C_3} \right)} \qquad (25)$$

$$\frac{X_{MO_2}}{X_{MC}} = \frac{K^{(111)}_{MC} \cdot I^{(220)}_{MO_2}}{K^{(220)}_{MO_2} \left(I_{MC}^{(111)} + M_2C_3^{(220)} - \frac{K^{(220)}_{M_2C_3}}{K^{(310)}_{M_2C_3}} \cdot I^{(310)}_{M_2C_3} \right)} \qquad (26)$$

Solving equations (17), (24), (25) and (26) we can find out the volume percent of the mixed carbides and oxide that is

$$X_{MC},\ X_{MC_2},\ X_{M_2C_3} \text{ and } X_{MO_2}$$

Table XIII and XIV gives the detail of the analysis of Mol-11/K2 samples. The weight percent in Table XIV for the respective phases were calculated from the volume percent using the following ratio.

$$C_{MC} = \frac{X_{MC} \cdot MC}{G} ; \quad C_{M_2C_3} = \frac{X_{M_2C_3} \cdot M_2C_3}{G}$$

$$C_{MC_2} = \frac{X_{MC_2} \cdot MC_2}{G} ; \quad G = X_{MC} \cdot MC + X_{M_2C_3} \cdot M_2C_3 + X_{MC_2} \cdot MC_2$$

Where C_{MC}, $C_{M_2C_3}$, C_{MC_2} are the weight percent of the respective carbides are their theoretical densities.

The carbon equivalent as obtained from the analysis figure agreed quite well with the figures obtained from chemical analysis as given by the suppliers.

References:

/1/ B.D. Cullity:
Elements of X-ray Diffraction, Addison-Wesley Publishing Comp. Inc. Reading (1959)

/2/ International Tables of X-ray Crystallography, Kynoch Press Birmingham (1952)

/3/ E.K. Storms:
The Refractory Carbides, Academic Press, New York (1967)

/4/ W.B. Pearson:
Handbook of Lattice Spacing and Structure of Metals and Alloys, London Perganon Press 1958

/5/ A. Taylor:
X-ray Metallography, John Wiley and Sons. INC, New York, London, 1961, Pg. 928

/6/ R. Conti, C.J. Toussaint, G.Vos:
Anal. Chem. Acta 41 (1968) 83

/7/ R. Theisen, D. Vollath:
Tabellen der Massenschwächungskoeffizienten von Röntgenstrahlen, Verlag Stahleisen 1967

/8/ T. Atoda, I. Higashi, J. Takahaski, Y. Sasa, M. Kobayaski:
Science Papers I.P.C.R. 55 (1961) 217

/9/ I. Waller:
Z. Physik 51, 213 (1923)

Table 1   Detail of the Crystal Structure of Dioxide, Mono, Di and Sesquicarbide of 'U', 'Pu' and (U-Pu)

	Structure Type	Space Group	No. of Atoms in Unit Cell	Atomic Positions	Detail of the Exact Atom Position
Di Oxide	Cubic, CaF_2 (C-1)	F m 3 m (Internat. Table 225)	4 'U' 8 'O'	$(0\ 0\ 0,\ 0\ \frac{1}{2}\ \frac{1}{2},\ \frac{1}{2}\ 0\ \frac{1}{2},\ \frac{1}{2}\ \frac{1}{2}\ 0)\ +$ 4 'U' in (a) 0 0 0 8 'O' in (c) $\frac{1}{4}\ \frac{1}{4}\ \frac{1}{4},\ \frac{3}{4}\ \frac{3}{4}\ \frac{3}{4}$	
Mono Carbide	Cubic, NaCL (B-1)	F m 3 m (Internat. Table 225)	4 'U' 4 'C'	$(0\ 0\ 0,\ \frac{1}{2}\ \frac{1}{2}\ 0,\ \frac{1}{2}\ 0\ \frac{1}{2},\ 0\ \frac{1}{2}\ \frac{1}{2})\ +$ 4 'U' in(a) (0 0 0) 4 'C' in(b) $(\ \frac{1}{2}\ \frac{1}{2}\ \frac{1}{2}\ )$	
Di Carbide	Tetrag. CaC_2 (C-11a)	I_4/m m m (Internat. Table 139)	2 'U' 4 'C'	$(0\ 0\ 0,\frac{1}{2}\ \frac{1}{2}\ \frac{1}{2})\ +$ 2 'U' in(a)(0 0 0) 4 'C' in(e)(0 0 Z), $(0\ 0\ \bar{Z})$, where Z = 0.38	'U' in (0 0 0) 'C' in (00.38), $(00.\overline{38})$
Sesqui Carbide	Cubic $D5_c$	I $\bar{4}$ 3 d (Internat. Table 220)	16 'U' 24 'C'	$(\ 0\ 0\ 0,\ \frac{1}{2}\ \frac{1}{2}\ \frac{1}{2}\ )\ +$ 16 'U' in (c) (xxx), $(\frac{1}{4}+x,\frac{1}{4}+x,\frac{1}{4}+x)$ $(\frac{1}{2}+x,\ \frac{1}{2}-x,\ \bar{x})\circlearrowright\ (\frac{3}{4}+x,\ \frac{1}{4}-x,\ \frac{3}{4}-x)\circlearrowright$ 24 'C' in (d) $(y\ 0\ \frac{1}{4})\circlearrowright\ ,\ (\bar{y}\ \frac{1}{2}\ \frac{1}{4})\circlearrowright$ $(\frac{1}{4}+y,\ \frac{1}{2},\frac{1}{4})\circlearrowright\ (\frac{3}{4}-y\cdot 0\cdot\frac{1}{4}\ )\circlearrowright$ where x = 0.05 for 'U' y = 0,295 for 'C'	'U' - (I) (0.05, 0.05, 0.05) (II) $(0.55,0.45,0.\overline{05})$ (III) $(0.\overline{05}, 0.55, 0.45)$ (IV) $(0.45,0.\overline{05},0.55)$ (V) (0.30, 0.30, 0.30) (VI) (0.80,0.20,0.70) (VII) (0.70, 0.80, 0.20) (VIII) (0.20,0.70,0.80) 'C' - (I) (0.295,0,0.25) (II) (0.25,0.295,0) (III) (0,0.25,0.295) (IV) $(0.2\overline{9}5,0.50,0.25)$ (V) $(0.25, 0.2\overline{9}5,0.5)$ (VI) $(0.5, 0.25,0.2\overline{9}5)$ (VII) (0.545,0.5, 0.25) (VIII) (0.25,0.545,0.5) (IX) (0.5,0.25,0.545) (X) (0.455,0,0.25) (XI) (0.25,0.455,0) (XII) (0,0.25,0.455)

Table 2 Pure Uranium-Monocarbide and Uranium Plutonium Mixed Monocarbide

Phase	Line	$\sin^2\theta$	$\frac{\sin\theta}{\lambda}$	f_{U-Pu}	f_c	Equation for 'F'	$/F/^2 \cdot 10^5$	L.P.	P	$\frac{1}{2}\mu \cdot 10^{-5}$	$\frac{1}{\nu^2} \cdot 10^{-5}$	'K'$\cdot 10^{-1}$
UC	111	0.0723	0.174	79.89	3.98	F = 4 (fu-fc)	0.9220	24.867	8			1.158
ϕ= 13.63	200	0.0964	0.201	77.33	3.57	F = 5 (fu+fc)	1.0472	18.023	6			0.715
g/cc	220	0.1929	0,285	69.98	2.64	F = 4 (fu+fc)	0.8438	7.947	12	9.4078	6.7119	0.508
a=4.9605Å	311	0.2652	0.334	66.01	2.27	F = 4 (fu-fc)	0.6500	5.368	24			0.529
	422	0.5787	0.494	55.05	1.70	F = 4 (fu+fc)	0.5153	2.728	24			0.213
	511,333	0.6510	0.524	53.34	1.65	F = 4 (fu-fc)	0.4275	2.837	32			0.245
$U_{.85}Pu_{.15}C$	111	0.0723	0.174	80.17	3.98		0.9287	24.867	8			1.149
	200	0.0964	0.201	77.60	3.57		1.0542	18.023	6			0.709
ϕ=13.6105	220	0.1927	0.285	70.23	2.64	same as the	0.8496	7.957	12	9.2870	6.6967	0.504
g/cc	311	0.2650	0.334	66.25	2.27	lines for pure	0.6549	5.374	24			0.525
	422	0.5782	0.493	55.32	1.70	UC	0.5203	2.728	24			0.212
a=4.9624Å	511, 333	0.605	0.523	53.60	1.65		0.4318	2.836	32			0.244
$U_{.80}Pu_{.20}C$	111	0.0723	0.174	80.26	3.98		0.9309	24.867	8			1.146
	200	0.0963	0.201	77.69	3.57		1.0565	18.044	6		0.	0.708
ϕ=13.604	220	0,1927	0.285	70.31	2.64	same as the	0.8516	7.957	12	9.2474	6.6917	0.503
g/cc	311	0.2649	0.334	66.33	2.27	lines for pure	0.6566	5.376	24			0.524
	422	0.5780	0.493	55.39	1.70	UC	0.5215	2.728	24			0.211
a=4.9630Å	511, 333	0.6503	0.523	53.67	1.65		0.4330	2.835	32			0.243
$U_{.75}Pu_{.25}C$	111	0.0722	0.174	80.35	3.98		0.9331	24.905	8			1.144
	200	0.0963	0.201	77.78	3.57		1.0588	18.044	6			0.706
ϕ=13.5975	220	0.1926	0.285	70.40	2.64	same as the	0.8536	7.962	12	9.2084	6.6866	0.502
g/cc	311	0.2648	0.334	66.41	2.27	lines for pure	0.6582	5.379	24			0.523
	422	0.5779	0.493	55.46	1.70	UC	0.5228	2.728	24			0.211
a=4.9636Å	511, 333	0.6502	0.523	53.74	1.65		0.4341	2.835	32			0.242
$U_{.70}Pu_{.30}C$	111	0.0722	0.174	80.45	3.98		0.9354	24.905	8			1.142
ϕ= 13.59	200	0.0963	0.201	77.87	3.57		1.0612	18.044	6			0.704
g/cc	220	0.1926	0.285	70.48	2.64	same as the	0.8555	7.962	12	9.1697	6.6816	0.501
	311	0.2648	0.334	66.49	2.27	lines for pure	0.6599	5.379	24			0.522
a=4.9642Å	422	0.5778	0.493	55,53	1.70	UC	0.5242	2.728	24			0.210
	511, 333	0.6500	0.523	53.81	1.65		0.4352	2.834	32			0.242

Table 3 Pure Uranium Sesquicarbide and Uranium Plutonium Mixed Sesquicarbide

Phase	Line	$\sin^2\theta$	$\frac{\sin\theta}{\lambda}$	f_{U-Pu}	f_c	Equation for 'F'	$/F/^2 \cdot 10^5$	L.P.	P	$\frac{1}{2\mu}\cdot 10^{-4}$	$\frac{1}{\nu}2\cdot 10^{-6}$	'K' value $.10^{-1}$
U_2C_3 ϕ=12.88 g/cc a_o=8.088Å	211	0.0544	0.151	82.08	4.34	$F=-1.382f_u+2.70126f_c+i(1.902f_u-3.0365f_c)$	1.2309	33.917	24	1.0192	3.5723	0.365
	220	0.0726	0.175	79.80	3.98	$F=5.236f_u-4fc$	6.4627	24.754	12			0.699
	310	0.0907	0.195	77.90	3.66	$F=i(3.804fu+2.65616fc)$	3.7466	19.310	24			0.632
	321	0.1270	0.231	74.73	3.21	$F=-(1.382fu+2.1432fc)-i(1.902fu+2.30069fc)$	1.3800	13.117	48			0.316
	332	0.1995	0.290	69.55	2.60	$F=-(2.236fu+2.1432fc)+i(3.07756fu-0.3552fc)$	2.8547	7.626	24			0.190
	510,431	0.2358	0.315	67.52	2.40	$F=i(3.804fu-0.5032fc)$	2.6140		24			
						$F=-4.736fu+2.65616fc-i.0.36322fu$	3.9530	6.200	48			0.570
$(U_{.85}Pu_{.15})_2C_3$ ϕ=12.859 g/cc a_o=8.095Å	211	0.0543	0.151	82.36	4.34	Same as the lines for pure U_2C_3	1.2403	33.965	24	1.0070	3.5549	0.362
	220	0.0724	0.175	80.17	3.98		6.5232	24.818	12			0.695
	310	0,0905	0.195	78.17	3.66		3.7716	19.348	24			0.627
	321	0.1268	0.231	74.90	3.21		1.3854	13.146	48			0.313
	332	0.1992	0.290	69.80	2.60		2.8754	7.640	24			0.189
	510,431	0.2354	0.315	67.76	2.40		2.6331		24			
							3.9528	6.218	48			0.563
$(U_{.85}Pu_{.15})_2C_3$ ϕ=12.84 g/cc a_o=8.097Å	211	0.0543	0.151	82.46	4.34	Same as the lines for pure U_2C_3	1.2445	33.991	24	1.0030	3.5491	0.361
	220	0.0724	0.175	80.26	3.98		6.5388	24.833	12			0.694
	310	0.0905	0.195	78.26	3.66		3.7800	19.360	24			0.625
	321	0.1267	0.231	74.99	3.21		1.3885	13.155	48			0.312
	332	0.1991	0.290	69.89	2.60		2.8823	7.645	24			0.188
	510,431	0.2353	0.315	67.85	2.40		2.6395		24			
							3.9821	6.222	48			0.563
$(U_{.85}Pu_{.15})_2C_3$ ϕ=12.83 g/cc a_o=8.099Å	211	0.0543	0.151	82.55	4.34	Same as the lines for pure U_2C_3	1.2465	34.005	24	0.9990	3.5433	0.360
	220	0.0724	0.174	80.35	3.98		6.5543	24.845	12			0.692
	310	0.0904	0.195	78.35	3.66		3.7885	19.370	24			0.623
	321	0.1266	0.231	75.07	3.21		1.3915	13.162	48			0.311
	332	0.1990	0,289	70.06	2.61		2.8963	7.650	24			0.188
	510,431	0.2352	0.314	67.99	2.41		2.6505		24			
							3.9919	6.226	48			0.563
$(U_{.70}Pu_{.30})_{22}C_3$ ϕ=12.82 g/cc a_o=8.101A	211	0.0542	0.151	82.64	4.34	Same as the lines for pure U_2C_3	1.2496	34.026	24	0.9951	3.5376	0.359
	220	0.0723	0.174	80.45	3.98		6.5699	24.860	12			0.690
	310	0.0904	0.195	78.44	3.66		3.7969	19.382	24			0.622
	321	0.1989	0.289	70.14	3.21		1.3946	7,655	48			0.310
	332	0.1266	0.231	75.16	2.61		2.9031	13.170	24			0.188
	510,431	0.2350	0.314	68.07	2.41		2.6566		24			
							4.0178	6.230	48			0.563

Table 4 Pure Uranium Dicarbide and Uranium Plutonium Mixed Dicarbide

Phase	Line	$\sin^2\theta$	$\frac{\sin\theta}{\lambda}$	f_{U-Pu}	fc	Equation for 'F'	$/F/^2 \cdot 10^5$	L.P.	P	$\frac{1}{2\mu}\cdot 10^{-4}$	$\frac{1}{\nu^2}\cdot 10^{-4}$	$'K'\cdot 10^{-1}$
Pure UC_2. a=3.5190Å c=5.9790Å ϕ=11.68 g/cc	101	0.0645	0.165	80.75	4.12	F = fu − 1.4589 fc	0.2234	28.190	8	1.1499	1.8242	1.056
	002	0.0664	0.167	80.56	4.09	F = fu + 0.1256 fc	0.2575	27.317	2			0.295
	110	0.0958	0.201	77.33	3.57	F = fu + 2 fc	0.2854	18.161	4			0.435
	112	0.1622	0.261	72.00	2.87	F = fu + 0.1256 fc	0.2094	9.810	8			0.345
	114	0.3614	0.390	61.82	1.99	F = fu − 1.9842 fc	0.1339	3.729	8			0.084
	312	0.5455	0.479	56.01	1.74	F = fu + 0.1256 fc	0.1265	2.742	16			0.116
$(U_{.85}Pu_{.15})C_2$ a=3.5356Å c=5.9962Å ϕ=11.49 g/cc	101	0.0640	0.164	81.12	4.14	Same as the lines for pure UC_2	0.2255	28.450	8	1.1526	1.7798	1.052
	002	0.0660	0.167	80.84	4.09		0.2581	27.490	2			0.291
	110	0.0949	0.200	77.69	3.58		0.2880	18.342	4			0.434
	112	0.1609	0.260	72.34	2.88		0.2114	9.904	8			0.344
	114	0.3589	0.389	62.12	2.00		0.1353	3.756	8			0.083
	312	0.5406	0.477	56.35	1.75		0.1280	2.747	16			0.115
$(U_{.80}Pu_{.20})C_2$ a=3.5412Å c=6.002Å ϕ=11.42 g/cc	101	0.0638	0.164	81.22	4.14	Same as the lines for pure UC_2	0.2261	28.538	8	1.1536	1.7652	1.051
	002	0.0659	0.167	80.93	4.09		0.2587	27.544	2			0.290
	110	0.0946	0.200	77.78	3.58		0.2886	18.408	4			0.434
	112	0.1605	0.260	72.43	2.88		0.2119	9.935	8			0.343
	114	0.3581	0.388	62.27	2.00		0.1359	3.766	8			0.083
	312	0.5390	0.476	56.49	1.75		0.1286	2.749	16			0.115
$(U_{.75}Pu_{.25})C_2$ a=3.5467Å c=6.0077Å ϕ=11.36 g/cc	101	0.0636	0.164	81.31	4.14	Same as the lines for pure UC_2	0.2266	28.626	8	1.1547	1.7509	1.050
	002	0.0657	0.166	81.12	4.11		0.2599	27.613	2			0.290
	110	0.0943	0.199	77.96	3.60		0.2901	18.474	4			0.433
	112	0.1601	0.260	72.51	2.88		0.2124	9.966	8			0.343
	114	0.3573	0.388	62.35	2.00		0.1363	3.775	8			0.083
	312	0.5374	0.475	56.62	1.75		0.1292	2.751	16			0.115
$(U_{.70}Pu_{.35})C_2$ a=3.5523Å c=6.0135Å ϕ=11.29 g/cc	101	0.0634	0.163	81.50	4.16	Same as the lines for pure UC_2	0.2276	28.715	8	1.1558	1.7366	1.049
	002	0.0656	0.166	81.21	4.11		0.2605	27.664	2			0.289
	110	0.0940	0.199	78.05	3.60		0.2907	18.539	4			0.433
	112	0.1597	0.259	72.69	2.89		0.2134	9.998	8			0.343
	114	0.3565	0.388	62.42	2.00		0.1367	3.784	8			0.083
	312	0.5358	0.475	56.69	1.75		0.1296	2.753	16			0.114

Table 5 Pure Uranium Dioxide and Mixed Uranium Plutonium Dioxide

Phase	Line	$\sin^2\theta$	$\frac{\sin\theta}{\lambda}$	f_{U-Pu}	fo	Equation for 'F'	$/F/^2.10^5$	L.P.	P	$\frac{1}{2\mu}\cdot 10^{-4}$	$\frac{1}{\nu}2\cdot 10^{-5}$	$'K'\cdot 10^{-1}$
UO_2	111	0.0595	0.158	81.42	6.36	F = 4 (fu + 0.fo)	1.0606	30.781	8			1.230
	200	0.0793	0.182	79.13	5.94	F = 4 (fu - 2 fo)	0.7236	22.446	6			0.459
ϕ=10.96g/cc	220	0.1587	0.258	72.25	4.70	F = 4 (fu + 2 fo)	1.0667	10.071	12	1.2595	3.7368	0.607
a=5.4691Å	311	0.2182	0.303	68.47	4.06	F = 4 (fur + 0.fo)	0.7501	6.830	24			0.579
	222	0.2380	0.316	67.44	3.90	F = 4 (fu - 2 fo)	0.569	6.135	8			0.131
$(U_{.85}Pu_{.15})O_2$	111	0.0597	0.158	81.70	6.35		1.0679	30.669	8			1.222
	200	0.0797	0.183	79.32	5,92		0.7285	22.33	6			0.455
ϕ=11.03g/cc	220	0.1593	0.259	72.43	4.68	Same as for pure UO_2	1.0705	10.023	12	1.2331	3.7821	0.600
a=5.458A	311	0.2190	0.304	68.64	4.04		0.7538	6.799	24			0.574
	222	0.2390	0.317	67.70	3.89		0.5727	6.103	8			0.130
$(U_{.80}Pu_{.20})O_2$	111	0.0598	0.159	81.69	6.33		1.0678	30.602	8			1.216
	200	0.0798	0.183	79.40	5.92		0.7303	22.299	6			0.454
ϕ=11.06g/cc	220	0.1595	0.259	72.51	4.68	Same as for pure UO_2	1.0727	10.008	12	1.2246	3.7973	0.599
	311	0.2194	0.304	68.72	4.04		0.7556	6.785	14			0.572
a=5.4545Å	222	0.2393	0.318	67.61	3.88		0.5732	6.094	8			0.130
$(U_{.75}Pu_{.25})O_2$	111	0.0599	0.159	81.79	6.33		1.0703	30.555	8			1.213
	200	0.0799	0.183	79.49	5.92		0.7322	22.268	6			0.454
ϕ=11.08g/cc	220	0.1597	0.259	72.60	4.68	Same as for pure UO_2	1.0750	9.991	12	1.2161	3.8126	0.598
	311	0.2196	0.304	68.81	4.04		0.7575	6.774	24			0.571
a=5.4508Å	222	0.2396	0.318	67.69	3.88		0.5748	6.083	8			0.130
$(U_{.70}Pu_{.30})O_2$	111	0.0599	0.159	81.88	6.33		1.0727	30.510	8			1.210
	200	0.0799	0.184	79.49	5.90		0.7329	22.233	6			0.452
ϕ=11.11g/cc	220	0.1599	0.260	72.60	4.67	Same as for pure UO_2	1.0743	9.975	12	1.2077	3.8280	0.595
	311	0.2199	0.304	68.89	4.04		0.7593	6.762	24			0.570
a=5.4472Å	222	0.2399	0.318	67.77	3.88		0.5763	6.074	8			0.129

Table 6 Multiplicity Factors for 'Cubic' and 'Tetragonal' Crystal System

Cubic:	hkl	hhl	okl	okk	hhh	ool	
MO_2, MC, M_2C_3	48^+	24	24^+	12	8	6	
Tetragonal:	hkl	hhl	okl	hko	hho	oko	ool
MC_2	16^+	8	8	8^+	4	4	2

$^+$These are the usual multiplicity factors. In some crystals, planes having these indices comprise two forms with the same spacing but different structure factor, and the multiplicity factor for each form is half the value given above.

Table 8 'K' and 'A' Ratio Comparison of 'UC '

Line	2 θ	K-values $.10^{-1}$	Intens. Area	Comparison of 'K' and 'A' ratio		Percent Diff.
				K-ratio	A-ratio	
(111)	$31{\cdot}2^o$	1·158	$4{:}2{\cdot}10^5$	K_{111}/K_{220} = 2.27	A_{111}/A_{220} = 2.10	7.40
				K_{111}/K_{311} = 2.19	A_{111}/A_{311} = 2,01	8,20
(220)	$52{\cdot}10^o$	0.508	$1.9969{\cdot}10^5$	K_{111}/K_{422} = 5.436	A_{111}/A_{422} = 5.217	4.02
				K_{220}/K_{311} = 0.960	A_{220}/A_{311} = 0.958	2.08
(311)	61·99	0.529	$2.0844{\cdot}10^5$	K_{220}/K_{422} = 2.385	A_{220}/A_{422} = 2.480	3.83
				K_{311}/K_{422} = 2.48	A_{311}/A_{422} = 2.59	4.40
(422)	99·05	0.213	$0.80498{\cdot}10^5$			

Table 7 Equations giving the 'K' values, of different lines of different phases, as a function of the Plutonium concentration 'C', between 0 % to 30 % Pu

Phase	Line	Equation 'K' = f (c)
Mono Carbide	(111)	$K^{111} = 1.158\cdot 10^{-1}-5.33\cdot 10^{-3}\ C_{Pu}$
	(200)	$K^{200} = 0.715\cdot 10^{-1}-3.66\cdot 10^{-3}\ C_{Pu}$
	(220)	$K^{220} = 0.508\cdot 10^{-1}-2.33\cdot 10^{-3}\ C_{Pu}$
	(311)	$K^{311} = 0.528\cdot 10^{-1}-2.266\cdot 10^{-3}\ C_{Pu}$
	(422)	$K^{422} = 0.213\cdot 10^{-1}-1.00\cdot 10^{-3}\ C_{Pu}$
	$\binom{511}{333}$	$K^{\substack{511\\333}} = 0.245\cdot 10^{-1}-1.00\cdot 10^{-3}\ C_{Pu}$
Di Carbide	(101)	$K^{101} = 1.056\cdot 10^{-1} - 2.33\cdot 10^{-3}\ C_{Pu}$
	(002)	$K^{002} = 0.295\cdot 10^{-1} - 2.00\cdot 10^{-3}\ C_{Pu}$
	(110)	$K^{110} = 0.435\cdot 10^{-1} - 6.66\cdot 10^{-4}\ C_{Pu}$
	(112)	$K^{112} = 0.345.10^{-1} - 6.66\cdot 10^{-4}\ C_{Pu}$
	(114)	$K^{114} = 0.0838\cdot 10^{-1} - 2.66\cdot 10^{-4}\ C_{Pu}$
	(312)	$K^{312} = 0.116\cdot 10^{-1} - 6.66\cdot 10^{-4}\ C_{Pu}$
Sesqui Carbide	(211)	$K^{211} = 0.365\cdot 10^{-1} - 2.00\cdot 10^{-3}\ C_{Pu}$
	(220)	$K^{220} = 0.699\cdot 10^{-1} - 3.00\cdot 10^{-3}\ C_{Pu}$
	(310)	$K^{310} = 0.632\cdot 10^{-1} - 3.33\cdot 10^{-3}\ C_{Pu}$
	(321)	$K^{321} = 0.316\cdot 10^{-1} - 2.00\cdot 10^{-3}\ C_{Pu}$
	(322)	$K^{332} = 0.190\cdot 10^{-1} - 6.66\cdot 10^{-4}\ C_{Pu}$
	(510,431)	$K^{510,431} = 0.570\cdot 10^{-1} - 2.33\cdot 10^{-3}\ C_{Pu}$
Oxide	(111)	$K^{111} = 1.23\cdot 10^{-1} - 6.66\cdot 10^{-3}\ C_{Pu}$
	(200)	$K^{200} = 0.459\cdot 10^{-1} - 2.33\cdot 10^{-3}\ C_{Pu}$
	(220)	$K^{220} = 0.607\cdot 10^{-1} - 4.00\cdot 10^{-3}\ C_{Pu}$
	(311)	$K^{311} = 0.579\cdot 10^{-1} - 3.00\cdot 10^{-3}\ C_{Pu}$
	(222)	$K^{222} = 0.131\cdot 10^{-1} - 6.66\cdot 10^{-4}\ C_{Pu}$

Table 9 'K' and 'A' Ratio Comparison of U_2C_3

Line	2 θ	K-value $\cdot 10^{-1}$	Intensity Area	Comparison of 'K' and 'A' ratios		Percent Diff.
				K-ratio	A-ratio	
(220)	31.25	0.699	$1.514 \cdot 10^5$	K^{220}/K^{310} = 1.106	A^{220}/A^{310} = 1.147	3.70
				K^{220}/K^{321} = 2.212	A^{220}/A^{321} = 2.082	5.90
(310	35.05	0.632	$1.320 \cdot 10^5$	K^{220}/K^{510}_{431} = 1.22	A^{220}/A^{510}_{431} = 1.10	9.83
				K^{310}/K^{321} = 2.000	A^{310}/A^{321} = 1.816	9,20
(321)	41.75	0.316	$0.727 \cdot 10^5$	K^{310}/K^{510}_{431} = 1.10	A^{310}/A^{510}_{431} = 0.964	12.27
				K^{510}_{431}/K^{321} = 1.804	A^{510}_{431}/A^{321} = 1.887	4.6
$\left(^{510}_{431}\right)$	58.10	0.570	$1.3718 \cdot 10^5$			

Table 10 'K' and 'A' Ratio Comparison of UC_2

Line	2 θ	K-value $\cdot 10^{-1}$	Intensity Area	Comparison of 'K' and'A' ratios		Percent Diff.
				K-ratio	A-ratio	
(110)	36.06	0.435	$3.56 \cdot 10^5$	K^{110}/K^{112} = 1.26	A^{110}/A^{112} = 1.13	10.31
				K^{110}/K^{114} = 5.19	A^{110}/A^{114} = 5.02	3.27
(112)	47.50	0.345	$3.15 \cdot 10^5$	K^{110}/K^{312} = 3.75	A^{110}/A^{312} = 3.95	5.33
				K^{112}/K^{114} = 4.12	A^{112}/A^{114} = 4.44	7.76
(114)	73.90	0.0838	$0.7088 \cdot 10^5$	K^{112}/K^{312} = 3.09	A^{112}/A^{312} = 3.44	10.17
				K^{312}/K^{114} = 1.38	A^{312}/A^{114} = 1.27	7.9
(312)	95.22	0.116	$0.90104 \cdot 10^5$			

Table 11 'K' and 'A' Ratio Comparison of UO_2

Line	2 θ	K-value $\cdot 10^{-1}$	Intensity Area	Comparison of 'K' and 'A'-ratio		Percent Diff.
				K-ratio	A-ratio	
(111)	28.30	1.230	$9.200 \cdot 10^5$	$K^{111}/K^{200} = 2.6799$	$A^{111}/A^{200} = 2.4084$	10.13
				$K^{111}/K^{220} = 2.0259$	$A^{111}/A^{220} = 1.9205$	5.20
(200)	32.80	0.459	$3.820 \cdot 10^5$	$K^{111}/K^{311} = 2.1241$	$A^{111}/A^{311} = 1.9700$	7.25
				$K^{220}/K^{200} = 1.3228$	$A^{220}/A^{200} = 1.2540$	5.20
(220)	47.2	0.607	$4.7903 \cdot 10^5$	$K^{220}/K^{311} = 1.0485$	$A^{220}/A^{311} = 1.0258$	2.16
				$K^{311}/K^{200} = 1.2617$	$A^{311}/A^{200} = 1.2225$	3.10
(311)	55.8	0.579	$4.670 \cdot 10^5$			

Table 12 'K' and 'A' Ratio Comparison of Mixed (U-Pu) Monocarbide

Line	2θ Scanned	Phase Present in this Range (Within Monocarbide Peaks)	Total Area under Peak $\cdot 10^5$	Corrected Area for Monocarbide $\cdot 10^5$	K-values $\cdot 10^{-1}$	Comparison of 'K' and 'A' ratios		Percent Diff.
						$\frac{K^{111}}{K^{200}} = 1.620$	$\frac{A^{111}}{A^{200}} = 1.49$	8.02
(111)	29.5°-32.35	$(U,Pu)C^{111}$ + $(U,Pu)_2C_3^{220}$	3.5132	3.36456	1.149	$\frac{K^{111}}{K^{200}} = 2.270$	$\frac{A^{111}}{A^{220}} = 1.973$	13.08
						$\frac{K^{111}}{K^{311}} = 2.190$	$\frac{A^{111}}{A^{311}} = 1.822$	16.8
(200)	34.5°-37.5	$(U,Pu)C^{200}$ + $(U,Pu)C_2^{110}$	2.2786	2.26417	0.709	$\frac{K^{200}}{K^{220}} = 1.405$	$\frac{A^{200}}{A^{220}} = 1.327$	5.5
						$\frac{K^{200}}{K^{311}} = 1.32$	$\frac{A^{200}}{A^{311}} = 1.226$	6.8
(220)	50.5°-53.5	$(U,Pu)C^{220}$ + $(U,Pu)C_2^{200}$ $(U,Pu)C_2^{103}$	1.719	1.7054	0.504	$\frac{K^{311}}{K^{220}} = 1.04$	$\frac{A^{311}}{A^{220}} = 1.08$	3.84
(311)	60.5°-63.5	$(U,Pu)C^{311}$ + $(U,Pu)C_2^{004}$	1.8477	1.8466	0.525			

Table 13 'K' values of some lines of $(U_{.85}Pu_{.15})C_2$ which will be needed for getting the exact intensity values of (U,Pu)C lines.

Line	$\sin^2\theta$	$\frac{\sin\theta}{\lambda}$	f_{U-Pu}	f_C	Equation for (F)	$(F^2)\cdot 10^5$	L.P.	P	$\frac{1}{2\mu}\cdot 10^{-4}$	$\frac{1}{v^2}\cdot 10^{-4}$	$K\cdot 10^{-1}$
(200)	0.1916	0.284	70.31	2.65	$F = f_U + 2\ f_C$	0.2287	8.011	4	1.1526	1.7798	0.1503
(103)	0.1973	0.288	69.97	2.63	$F = f_U + 1.2748\ f_C$	0.2150	7.732	8			0.2727
(004)	0.2655	0.335	66.17	2.67	$F = f_U - 1.9842\ f_C$	0.1521	5.360	2			0.0334

Table 14:

Angle of Scanning 2 θ	2 θ and (hkl) of the phases present				'K' values of the lines of the respective phases $\cdot 10^{-1}$				Peaks of the diffraction pattern	
	(U,Pu)C	$(U,Pu)_2C_3$	$(U,Pu)C_2$	$(U,Pu)O_2$	(U,Pu)C	$(U,Pu)_2C_3$	$(U,Pu)C_2$	$(U,Pu)O_2$	Separate	Overlapped
29.5°-32.5°	(31.2°) (111)	(31.25°) (220)	29.86 (002)	-	1.14897	0.695	-	-		(111)MC + $(220)M_2C_3$
34.0°-37.5°	(36.19°) (200)	(35.05°) (310)	36.06 (110)	-	-	0.627	-	-	(310) M_2C_3	-
46° - 49°	-	-	47.5° (112)	47.05° (220)	-	-	0.344	0.6005	$(220)MO_2$ $(112)MC_2$	-

Table 15:

Sample Number	Phases	Intensity of the Phases	Volume Percent	Weight Percent	Carbon Equivalent	
					As given	As found by xray
Mol-11 /K2	$MC+M_2C_3$	$3.51 \cdot 10^5$ ($MC^{111} + M_2C_3^{220}$)	92	92.5	5.00	5.04
	M_2C_3	$1.34 \cdot 10^4$ ($M_2C_3^{310}$)	7	6.5		
	MC_2	$1.1 \cdot 10^3$ (MC_2^{112})	1	1		
X-2	$MC+M_2C_3$	$1.84 \cdot 10^5$ ($MC^{111} + M_2C_3^{310}$)	87.8	88.7		
	M_2C_3	$0.923 \cdot 10^4$ ($M_2C_3^{310}$)	8.6	8.2		
	MC_2	$0.214 \cdot 10^4$ (MC_2^{112})	3.6	3.1		