UO2 REACTOR FUEL

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DISTRIBUTION OF CESIUM ISOTOPES

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DISTRIBTUION OF CESIUM ISOTOPES

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UO2 REACTOR FUEL

By

HARRY FARRAR, B.A.

A Thesis

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SCOPE AND CONFEDES:

The absolute abundances of several isotopes of cesium and uranium in an enriched uranium oxide fuel rod have been measured with a mass spectrometer, using the isotope dilution technique. Samples taken from the inside and edge of the rod have shown that the cesium abundances, particularly Cs^{134} , near the centre are much less than expected, whereas at the edge, the abundances are as expected. An explanation for this distribution and for the unexpected Cs^{134} behaviour has been put forward, along with a new value for the effective capture cross section of U^{235} . A suggestion as to the mechanism of the movement of uranium oxide has been made, to account for the cylindrical hole found in the centre of the non-stoichiometric fuel rods.

ACRINO JLADOROGINTS

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<u>iii</u>

TABLE OF CONTENTS

	Page
GEMERAL INTRODUCTION	l
HISTORICAL INTRODUCTION	6
(A) Advances in Reactor Design	6
(B) Development of UO2 23 Nuclear Fuel	7
EXPERIMENTAL	12
(A) Mass Spectroscopy	12
(a) Filament Assemblies	12
(b) Sample Loading	12
(c) Ionic Species	13
(d) Mass Discrimination	13
(B) Uranium Dioxide	14
(C) Chemical Procedure	15
(a) Treatment of Sample A	15
(b) Freatment of Sample B	16
(c) Treatment of Sample C	17
(d) Treatment of Sample D	17
(e) Treatment of Sample E	18
(f) Treatment of Sample F	20
EXPERIMENTAL RESULTS	21
(A) Introduction	21
(B) Absolute Abundances	22

Page

(a) Isotope Dilution	22
(b) Abundances in Atoms Per Gram of Uranium	22
(C) Uranium Cross Sections	22
(a) Calandria Temperature	22
(b) Uranium-235	25
(c) Uranium-238	26
(i) Threshold Energy	26
(ii) Thermal Neutron Cross Section	27
(iii) Fast Fission Factor	27
(D) Neutron Flux	27
(a) Uranium-235 Daplotion	28
(b) Uranium-236 Formation	29
(E) Calculated Abundances	29
(a) Cesium-133	29
(i) Cross Section of Cs ¹³³	29
(ii) Expected Abundance	31
(b) Cesium-134	31
(c) Cesium-135	32
(i) Cross Section of Xenon-135	52
(ii) Expected Abundance	32
(d) Cesium-137	32
DISCUSSION	34
(A) Movement of Uranium Oxide	34
(B) Re-evaluation of the Effective Cross Section of U235	35
(C) Movement of Cesium	30

	P2.20
BIBLIOGRAPHY	43
APPENDIX	45

LIST OF TABLES

Page

I	Mass Spectrometric Ratios of Isotopes of Cesium in Samples A and B	16
II	Mass Spectrometric Ratios of Isotopes of Cesium and Uranium in Samples C and D	19
III	lectope Dilution Data for Samples E and F	20
IV	Mass Spectrometric and Isotope Dilution Data for Samples E and F	23
v	Abundances of Cesium and Uranium Isotopes in Atoms/g U \dots	24
VI	Uraniun-235 Cross Sections For $r = 0.07$ at $40^{\circ}C$	26
VII	Uranium Depletion and Neutron Flux Data	30
VIII	Expected and Measured Abundances of Cesium in Atoms/g U	33
IX	Data Concerning the Ratio U^{236}/Cc^{133}	37
x	World Values of Neutron Cross Sections of U235	39

GENERAL INTRODUCTION

Although Canada is now rich in coal, oil, gas and water, which give her at present an adequate supply of power, she must look to the future, where the electric power demand will be many times the present level. Fortunately, Canada has very large supplies of uranium, which, if used as fuel for reactors, will generate power for many decades to come.

It has been estimated (1) that by 1980, Ontario would need to import from Pennsylvania 37 million tons of coal per year to fuel its generating plants. In the last five years, huge uranium ore deposits have been found in the Bancroft and Blind Niver areas of Ontario. Production of uranium at the present annual rate, if irradiated in reactors to a practicable 1% burn-up, would give an annual energy output ten times that required in 1980. Even if the world's supply of uranium should become exhausted, scientists are sure that man will soon be able to control the fusion process, where very light atoms combine together to produce an immense amount of energy. This is why so many scientists today are building and studying larger and more sophisticated reactors of every conceivable design.

In the present decade, where power is abundant, however, atomic fission reactors, as they now stand, are not economically competitive with the other power developments, which run at about six mills per kilowatt-hour. Hence, before any large scale power reactors, such as those in England and Sootland, are built, Canada must devise methods of reducing the capital investment and the running costs.

When uranium=235 undergoes fission, a great amount of heat energy is released by gamma rays, fission fragments, fast neutrons and other partigles. The neutrons, in turn, may cause further fission and the heat used to drive electric generators,

Of the many different types of reactors now in operation, there are two main classes, slow and fast reactors. The slow reactors may be further grouped into homogeneous and heterogeneous reactors, the latter being the more common. Homogeneous reactors, such as boiling-water reactors, have the uranium in salt form dissolved in a solvent moderator such as water or heavy water. Heterogeneous reactors, on which the Canadians have based their hopes, use the heterogeneous moderator principle, where uranium is in the form of rods intersperced with some moderator such as D_20 , H_20 or graphite. The function of the moderator is to slow down neutrons from fission to an energy too low for resonance absorption by U^{238} yet adequate to cause thermal fission of U^{235} . The liquid moderators can be cooled by circulation through heat exchangers and the graphite by a fast flowing gas such as air.

Some reactors use pure U^{235} , some use natural uranium, which contains less than 1% U^{235} , others use intermediate "enriched" mixtures. There are "breeder" reactors that convert U^{238} and Th^{232} , which are relatively useless as thermal fission fuel, to Pu^{239} and U^{233} respectively, which may replace U^{235} as the fissionable material. All these types of reactors have their particular advantages and disadvantages. Canada, being a country too small in population to tackle all these types, has chosen the heavy-water moderated reactors as being both the most economical and the easiest to expand to larger proportions. Heavy-water reactor techniques make it clear that one can depend on obtaining an energy yield directly from natural-uranium fuel, in the form of relatively inexpensive sintered oxide, as high as 8000 to 10,000 megawatt days per tonne of uranium (2). At such a yield, the uranium itself costs less than 1 mill per kilowatt-hour and if the cost of fabrication, insurance, etc. is included, the whole fuel inventory should not exceed 1.5 mills per kilowatt-hour. Uranium dioxide is now preferred as a reactor fuel both for enriched and for natural-uranium reactors, not only for the water cooled reactors such as Dresden, the Yankee and the Canadian N.P.D. heavy-water reactors, but also for the advanced gascooled reactors of the United Kingdom and Oak Ridge. Uranium dioxide also seems best for organic-moderated or organic-cooled reactors, and even for some designs of sodium-cooled and fast reactors. If it were possible to take superheated steam directly from the reactor to the turbine, UO₂ would again be preferable to the uranium metal.

There are many major advantages of using uranium dioxide for fuel. One advantage over uranium metal is that it resists corrosion in water, an important feature should the fuel rod sheath break. Uranium dioxide has a much higher melting point than the metal, which is an advantage since the reactor could thus be run at greater temperatures. The processing to oxide form is far easier and cheaper than to the metallic form, a factor that makes such reactors economically more feasible. It has been found (3) that the insoluble fission gases can be retained in the oxide at moderate burn-up levels without introducing undue lattice strains. Uranium dioxide allows a long burn-up without swelling; and there is no neutron

wastage, since oxygen has no great affinity for thermal neutrons. The main trouble lies in its very low heat conductivity, which causes a large heat gradient across the rod.

In the N.R.X. reactor at Chalk River there are 176 fuel rods, 1.36 inches in diameter and 10.25 feet long. These rods are suspended in a container known as the calandria, which contains the heavy-water moderator. Some of these uranium metal rods have been replaced by rods of UO2. One having the code name CR V-e Rod 7 has been analysed, in part, mass spectrometrically as reported in this thesis. The fuel part of the rod was nearly stoichiometric UO2, with a density 9.75 g/oc which is 89% of the theoretical density of UO2. This material has the appearance of a hard, grey metal. While in the reactor, this rod contributed toward the critical mass, and was exposed to a large flux of neutrons, on and off for more than half a year. When removed from the reactor and extracted from its protective aluminium sheath, the rod was found to have developed a central cylindrical hole down its length, with radial cracks as far as the edge. Rod 7 at the edge of the centre hole showed evidence of having been recrystallized and perhaps melted, whereas at the cuter surface, which had remained cool, no obvious change had occurred. Fortunately, the rod in general had not increased in diameter so that the aluminium sheath was not broken.

Work is in progress at Chalk River (4) to understand these peculiar effects. Measurements of the yield of rare gases from the rods are being conducted by heating and grinding experiments both at Chalk River and at the Bettis Plant of the Westinghouse Electric Corporation, who work on behalf of the U.S. Atomic Energy Commission (5). Recently it has been found

(6) that the ratios of the isotopes Cs^{133} , Cs^{135} and Cs^{137} in the rod were not the same as the accepted values for fission (7), and that the ratios vary themselves across the diameter of the rod. This discovery prompted the research that is described in this thesis.

The absolute abundances of Cs^{133} , Cs^{134} , Cs^{135} , Cs^{137} , U^{235} , U^{236} and U^{238} in two sections of the rod were measured by the isotope dilution technique using a 90° mass spectrometer. The sample pellets were cut from the rod at Chalk River, one coming from the recrystallized zone and three from the non-recrystallized zone. It has been found that there was more cesium in the outer pellets than in the centre pellet, per gram of uranium by a factor of four, in the case of Cs^{133} , Cs^{135} and Cs^{137} and a factor of twelve for Cs^{134} . Moreover in both places there was less cesium present than could be accounted for.

HISTORICAL INTRODUCTION

(A) Advances in Reactor Design

Atomic energy research in Canada started at the beginning of the century when Ernest Rutherford came to MoGill University. While in collaboration with F. Soddy before 1902, he investigated the nature of radioactivity that had been discovered in 1896 by Becquerel. Rutherford determined the fundamental laws that govern spontaneous disintegration of radioactive materials. Between this time and 1940 great strides in the development of atomic physics were made, new particles were discovered and postulated, and intricate theories developed. In 1940, G. C. Laurence of the National Research Council of Canada assembled a subcritical mass of uranium oxide imbedded in a form of carbon, seeking to determine the possibility of the release of energy.

During the war many of the British, French and other European scientists, then doing nuclear research, moved to the U.S.A., and others came to Canada, where a joint Canadian-U.K. project got underway in January 1943. It was around this time that the Americans, under Fermi, brought the first atomic pile, or reactor, into operation by piling layers of graphite on layers of uranium. It was Canada's role to try heavy-water as a moderator, a task that they accomplished at Chalk River. On September 5th, 1945, the ZEEP (Zero Energy Experimental Pile) went into operation at just 10 watts. This reactor is still used to study fuel rod arrangements. On July 22, 1947, the N.R.X. (National Research X-Netal) reactor started

operation. Built for operating at 10 megawatts, it has been modified and since 1952 has run at 40 megawatts; being for several years the reactor with the highest neutron flux in the world. Howadays, N.R.X. is playing a leading role in the determination of the fundamental properties of matter, producing isotopes of high specific activity and making possible experiments, such as the one here described, relating to the development of atomic power. In 1951, it was decided to build the 200 megawatt N.R.U. (National Research Universal) reactor, which is now in operation. Canada, in 1954, began to study the feasibility of a power reactor which resulted in the design of the small atomic power station, N.P.D., now well under construction, and the preliminary design study for a large power station.

Meanwhile, the British have specialized in, and are relying on, the gas-cooled graphite-reflector type reactor. Although they have several now producing power, the design has proven more expensive to build and operate than anticipated. The United States is experimenting with several varieties of reactor. The U.S.S.R. has developed, among others, the elightly enriched (3.5%) natural-water-moderated type of reactor.

(B) Development of UO2 as Nuclear Fuel

Much experience has been accumulated at Chalk River on the irradiation of uranium metal "canned" in aluminium, since it is this fuel that is used in the reactors N.R.X. and N.R.U. Unfortunately, it has been found that uranium metal suffers severe demage when irradiated in a neutron flux, or when thermally cycled (1). Uranium is a complex metal, existing in three crystallographic forms. At. N.R.X. and N.R.U. operating tempera-

tures, the stable α phase exists; an orthorhombic single crystal that, when irradiated to 0.1% burn-up, will grow 100 % in the [010] direction, shorten by 50% in the [100] direction and remain unchanged in the [001] direction. One can introduce preferred orientation by the technique of cold rolling so that, when irradiated, the uranium undergoes marked dimensional expansions in two directions and contractions in the third. Thus the aim has been to produce uranium metal with small, equiaxial grains of random orientation, or a duplex texture such that two orientations, one producing growth, and the other producing shortening, would cancel.

In N.R.X., experiments show that when a sheath develops a leak the uranium reacts chemically with the cool $(50^{\circ}C)$ water, slowly. For power reactors to be efficient the second law of thermodynamics dictates a large heat gradient; hence future power reactors might operate with water at as high a temperature as $300^{\circ}C$. At this temperature the corrosion rate of the uranium is about 100,000 times greater than the rate at $50^{\circ}C$. Chalk River experiments (1) showed that when a small hole was formed in the sheath, the uranium liberated hydrogen which dissolved in its special zirconium alloy sheath, causing it to embrittle and break, exposing more uranium. This realization convinced the Canadian group at Chalk River that uranium metal would be unsatisfactory as a fuel for water cooled power reactors.

It has been found (8), however, that uranium dioxide can be pressed and sintered to dense hard pellets which can resist water corrosion even at 350°C. Since uranium dioxide has a face-centered cubic crystal structure it was expected to be far more isotropic than the metal, under

irradiation. Hence, after 1956 great interest arose in the properties and fabrication methods of 80...

Each research has been done at Chalk Niver to compact the uranium dioxide powders and to change them to pure U_2 from the more common U_2U_3 , which may be considered $U_2(U_3)$. Other catablishments involved in similar research are the Elderado Mining and Hefining Ltd. and the Bettis Fight of Section house in Fitteburgh.

Economic reasons also contributed to the increased interest in UC_2 . Since the act fuel costs (9) are roughly 22% of the total cost of electricity from reactors, it is obvious that cheaper chanical processing costs would make such reactors more competitive with coal, gas and oil. To get maxima to its usual oxide form U_3C_0 , it is necessary only to erush and heat ere and to perform relatively simple and cheap chemical operations. To convert U_3O_6 to uranium metal, however, is not easy and is usually done either by electrolysis or by reacting the code with aluminium and calcium setals, which themselves must be reduced electrolytically. Lither method is very expendive.

hater (1), experimento were corried out irradiating fuel specinear with different aranium-to-exygen ratios. The comparison of two sample rods, one being $U_{2,02}^{0}$ and the other being $U_{2,15}^{0}$ after an irradiation of 5400 hMD/tenne showed that the former was relatively unchanged whereas the latter had developed a hole in the control with radial erachs extending to the edge. This discovery necessitated the development of the new technology of chemp production of stoichicmetric U_{2}^{0} . In 1956, Bolle (10) at Westinghouse reported that after ball milling of U_{2}^{0} followed by hydrostatic pressing at up to 50,000 p.s.i., and eintering in a hydrogen atmosphere at 1750°C, he obtained pellets with densities up to 97,2 of the theoretical density. A more recent development shows that when the standard Eldorado product UO_3 is suspended in the form of particles in a blast of air in a simple machine, they grind against each other until their size is reduced to a few microns. When hydrogen reduced and sintered, the UO_3 particles become UO_2 with a density of better than 10 g/cc.

Although UO₂ has the high melting point of 2750°C, its thermal conductivity is very low, and lowers further with increased temperature. Thus one can expect very high temperature gradients in fuel rods such as those for N.P.D. where the heat generation is over 100 watts per cc of fuel. It was feared that the sheath would be deformed by both the fission gases under pressure, and by the general fuel expansion, since the coefficient of thermal expansion of the fuel is twice that of the zirconium alloy cladding.

By January 1958, these fears appeared unfounded. Sintering of the fuel during irradiation can lead to drastic changes in appearance of the cone without producing any external deformation of the assembly. Runnalls (1) has tried to explain the cause of the central hole formation in non-stoichiometric oxide as being due to the following process. The non-stoichiometric oxide disproportionates at the high centre temperature, liberating gaseous UO₅ which decomposes to UO₂ + O₂ at the cooler outside. The O₂ is then thought to recycle. Hence it has been desirable to reduce or eliminate this small amount of excess oxygen which can transport an appreciable quantity of uranium oxide toward a cold surface at temperatures well below the melting point of the fuel. Reactor design is liable to be limited (11) by this poor thermal conductivity which demands that the fuel rod should be small in cross section. The trouble with this is that it reduces the number of U^{238} fissions and in some cases leads to a high resonance trapping of neutrons. Such factors limit the attainable burn-up without recycling.

EXPERIMENTAL

(A) Mass Spectroscopy

All the measurements of isotope ratios reported in this thesis have been obtained using the ten-inch radius 90 degree-sector solidsource mass spectrometer with magnetic scanning and a ten stage electron multiplier. This instrument has been previously described by D. Irish (12).

(a) Filament Assemblics

All the analyses were performed using a multiple filement source (13) consisting of two filements made of tungsten ribbon 0.001" thick and 0.030" wide. The principle is that high ion efficiencies may be obtained if the sample is evaporated from a sample filement on to an ion filement, where the ions are produced. This has the advantage over the single filement arrangement in that the emission rate of the sample is controlled by a relatively cool sample filement, and the ionization by a hot ion filement. The filements, renewed for each sample, were supported and insulated from the head assembly with standard 0.040" untinned Kovar-glass feedthroughs, and were heated, prior to use, in vacuo to approximately 2000⁰C for one hour to clean their surfaces.

(b) Sample Loading

A small drop of the concentrated solution to be analysed was added to the sample filament which had been removed from the head assembly. A current of 1.8 amperes was passed through the filament at this time to hasten the evaporation.

(c) Ionic Species

By using the proper combinations of ion and sample filament temperatures, one can obtain the spectrum of isotopes of one element such as cesium with minimum interference from nuclides of the same mass number such as those of barium. Cs⁺ and Rb⁺ ions appear with ion filament currents of 3.5 to 4 amperes even without current flowing through the sample filament; usually, however, more ideal conditions are obtained with about 1.4 amperes through the sample filament. Uranium appears as UO^+ and U^+ ions with the ion filament current between 4 and 5 amperes and the sample filament current between 2.5 and 3.5 amperes.

(d) Mass Discrimination

Mass discrimination is a commonly observed phenomenon in instruments employing electron multipliers as detectors (14), since this instrument tends to be non-linear with mass. Another discriminating effect can be caused by isotope fractionation at the source. Both these have been discussed by D. Irish (12). Values for the relative abundances of the neodymium isotopes (15) were the same, to within the 1% precision possible with the mass spectrometer with, and after the removal of, the electron multiplier as the ion collector. Thus it was concluded that the multiplier caused no mass discrimination.

When natural neodymium isotopes were measured, the ion currents differed systematically from the accepted relative abundances, indicating a non-linearity in the Applied Physics Corporation vibrating-reed electrometer, and/or the Leeds & Northrup Speedomax recorder. A potentioneter was used to give measured voltages required to duplicate the ion currents and it was found that these voltages gave abundances in agreement with the accepted values. The potentiometer was similarly used to compare ion currents differing by 100 fold or more. It was found that the corrections due to non-linearity of the electrometer and recorder were only significant for peaks of about 1.5 inches or less. Since all the peaks were considerably greater than 1.5 inches, no corrections were made.

(B) Uranium Dioxide

This study is related to the experimental fuel rod CR V-e $\frac{1}{27}$ at Chalk River. The rod was placed in the N.R.X. reactor on July 30th, 1957, being one of several CR V-e rods, nominally enriched to 7.95% of the total uranium, having a density (16) of about 9.75 g/cc. Rod 7 was removed from the reactor on February 11th, 1958, having been in for 196 days. During this period the reactor was run at full power and shut off intermittently, hence any calculations involving short half-lives, making use of the log data, would be prohibitive.

Two samples were received, after they had "cooled" considerably, in early November 1958. One, known as sample 25, consisted of a single pellet taken from the recrystallized zone of the rod at a radial position of 9 o'clock (relative to maximum flux at 12 o'clock)^{*}, weighing about 150 mg. The other, sample 26, consisted of three subler pollets, each about 30 mg, taken from the non-recrystallized zone at 12 o'clock. A sectional photograph of this rod is shown in Figure 1. From the uranium-235 depletion, it has been estimated by R. G. Hart (17) that the burn-up near sample 25 was 5000 \pm 230 M/D/tonne and near sample 26 was 7200 \pm 200 M/D/tonne.

*Notation to compare radial positions with direction of core centre.

(C) Chemical Procedure

All chemical operations were performed in a "dry box". Glassware, beakers, pipettes and graduated flasks were cleaned with redistilled 6-molar nitric acid, after which they were repeatedly rinsed in deionized distilled water to minimize contamination of the fission products with naturally occurring nuclides.

(a) <u>Treatment of Sample A</u>

One of the pellets of sample 26 was put into a beaker and a few drops of 6-molar redistilled nitric acid were added. The beaker was then placed under an infrared heat lamp both to aid in the dissolution, and to evaporate the solution. In about 10 minutes the uranium and the soluble fission products had dissolved. After a further 10 minutes the resultant uranyl nitrate solution had evaporated almost to dryness, removing the excess nitric acid. The uranyl nitrate and the fission products were then taken into solution with about 5 ml of deionized distilled water. A few drops of this yellow solution were taken in a fine tipped controlled pipette and were placed on the sample filament as described above. The source assembly was put together and the mass spectrometer evacuated to a pressure of 10⁻⁶ mm of mercury. The ratios of the isotopes 133, 134, 135 and 137 of cesium were then measured and recorded as shown in Table I. The solution remaining was transferred to a polyethylene bottle to be stored for further possible analysis. Tho beaker and pipette were repeatedly rinsed in distilled deionized water, which was then added to the bottle.

(b) Treatment of Sample B

The pellot from sample 25 was placed in a 10 ml beaker to which was added a few drops of 6-molar re-distilled nitric acid. When the nitric acid assumed a brownish tinge, indicating the oxidation and dissolution of some uranium, a portion was removed to another beaker where it was evaporated to near dryness. The remaining undissolved pellet, still about 100 mg in weight, was retained as such for later experimentation. Deionized distilled water was added, as before, to the solution, after which one drop was placed on a sample filament, which in turn was placed in the mass spectrometer for analysis. The ratios of the isotopes 133, 135 and 137 of cesium were measured and recorded as in Table I. To correct the ratios Cs^{135}/Cs^{137} and Cs^{135}/Cs^{137} for decay of Cs^{137} after its formation from fission, the correction factor, equation (2), appendix A, must be applied with t_1 extending from July 30, 1957 to February 11, 1958 (0.537 yr); and t_2 from February 11, 1958 to January 16, 1959 (0.9315 yr). The corrected ratios are recorded in the last two columns of Table I.

TABLE I

Mass Spectrometric Ratios of Isotopes of Cesium in Samples A and B

Sample	Cs135 Cs137	Cs133 Cs127	Cs134 Cs133 x 103	Cs135 Cs137 corrected for decay*	Csljj Cslj7 oorrected for decay
A (egbo)	0.349 ± 0.002	1.093 ± 0.007	4.82 ± 0.05	0.338 ± 0.002	1.059 ± 0.007
(inside)	0.4274 ± 0.0035	1.220 ± 0.008		0.4142 ± 0.0035	1.182 ± 0.008

"Corrected for 26.6 year half-life of Cs¹³⁷ (18).

(c) Treatment of Sample C

Another pellet of sample 26 was dissolved in about 0.2 cc of 6molar re-distilled nitric acid, heated and evaporated to iryness. About 5 cc of deionized distilled water was added and one drop of the resultant solution was placed on a sample filament. It was found that between 20 and 30 spectrograms were required in order that the average ratios obtained have standard deviations less than 1%. The ratios of the isotopes 133, 134, 135 and 137 of cesium were carefully measured along with the 235, 236 and 238 isotope ratios of uranium. Data are given in Table II.

(d) Treatment of Sample D

Since there was some fluffy green material adhering to the sample, probably uranyl nitrate plus uranium dioxide, that part of the pellet 25 not dissolved in sample B was vigorously shaken in its metal container. The specimen was then placed into a cleaned, dried beaker that had previously been weighed, and the weight was found to be 123.3 mg. The pellet was then completely dissolved in 6-molar re-distilled nitric acid and heated to dryness. A few drops of deionized distilled water were added and the uranyl nitrate again went into solution, which was then placed into a 10 ml graduated flask. The beaker and the controlled pipette were repeatedly rinsed into the flask, out of which one drop was taken for mass spectrometric analysis. The flask being then filled to the 10 ml mark contained virtually all the pellet weighed beforehand. The ratios of the 133, 134, 135 and 137 isotopes of cesium and the 235, 236 and 238 isotopes of uranium were then measured with the mass spectremeter. These results are recorded in Table II along with the ratios Cs^{133}/Cs^{137} and

 Cs^{133}/Cs^{137} corrected for decay of Cs^{137} according to equation (2), Appendix A, with $t_2 = 1.332$ yr and 1.3644 yr for samples C and D respectively. (e) Treatment of Sample E

Exactly half of sample D was taken into a 5 ml pipette which was discharged into another 10 ml graduated flask, and was duly rinsed with deionized distilled water. In order to obtain the absolute numbers of atoms per gram of uranium, the isotope dilution technique was employed.

Two standard isotope dilution solutions were prepared, one containing cesium, the other uranium. The cesium, supplied by British Drug Houses Ltd., was in the chemical form CsCl, and the uranium was 100,5 U^{238} supplied by Oak Ridge National Laboratories in the chemical form $U_z O_g$ known as "Q oxide".

Calculations were performed to estimate the total number of Cs^{133} and U^{238} atoms that were present in the aliquot of sample D. The number of U^{238} atoms was calculated from the knowledge that 123.3 mg of UO_2 was originally dissolved. An estimate of the number of Cs^{133} atoms in the aliquot was made using an approximate value of \emptyset , the thermal neutron flux, obtained from the Cs^{135}/Cs^{1370} ratio and the equation $\frac{Cal35}{Cs^{1376}} = \frac{\lambda}{\lambda + \sigma \emptyset} \frac{y^{135}}{y^{137}}$ derived in Appendix B, where λ and σ are the decay constant and effective absorption cross section of xenon-135 respectively. It is advantageous mass spectrometrically to add Cs^{133} in a quantity about 10 times that present already in the aliquot so that the new Cs^{137}/ds^{137} ratio would be about 10. A quantity of U^{238} equal to that estimated present was added, since a larger amount would have resulted in the ratio U^{238}/U^{235} being too large for accurate measurement.

TABLE II

llass	Spectrometric	Ratios	of	Isotopes	of	Cesium	and	Uranium	in	Samples	C	and	D
-------	---------------	--------	----	----------	----	--------	----------------------	---------	----	---------	---	-----	---

Semple	<u>Cs¹³⁵</u> Cs ¹³⁷	$\frac{Cs^{133}}{Cs^{137}}$	$\frac{Cs^{134}}{Cs^{133}}$ x 10 ³	$\frac{C_{B}^{135}}{C_{B}^{137}}$	Ca ¹³³ Ca ¹³⁷ c corrected*	υ ²³⁵ υ ²³⁸	<u>v</u> ²³⁶ v ²³⁵
C	0.3449	1.1179	4.28	0.331	1.072	0.0748	0.0310
(edge)	± 0.0025	± 0.0045	± 0.10	± 0.002	± 0.005	± 0.0005	± 0.0005
D	0,4444	1.186	1.378	0.4258	1.136	0.0777	0.02367
(inside)	± 0.0038	± 0.013	± 0.033	± 0.0085	± 0.013	± 0.0004	± 0.00036

*Corrected for 26.6 yr half-life of Calor (18).

Some cesium chloride was weighed and then dissolved in distilled deionized water. The solution was put in a 1000 ml graduated flask which was then supplemented with more water. A 500 lambda pipette was then used to transfer exactly 0.5 ml of this solution to the aliquot of sample D. A quantity of "Q oxide" was then weighed, dissolved in 6-molar redistilled nitric acid, dried, redissolved in deionized distilled water, and was added with the washings to the aliquot thus making sample E. Particulars are given in Table III.

(f) Treatment of Sample F

The third pellet of sample 26 was weighed and dissolved, but it was feared that possible contamination might have occurred so the sample was discarded. Instead, an aliquot of sample C was taken, containing an estimated 30 mg of uranium dioxide. To this was added a solution containing a weighed amount of "Q oxide". Another 0.5 ml of the CaCl solution used in the isotope dilution of sample E was transferred to the aliquot of sample C with the 500 lambda pipette. Data concerning the isotope dilution are given in Table III.

TABLE III

	A	LIQUOT	ISOTOPA	DILUTION	SOLUTION
Sample	Element	Estimated No. of Atoms Present	Chemical Form	Meight Used (mg)	No. of Atoms Addsd
E	v ²³⁸	2.424×10^{20}	⁰ 308	100.3	2.152 x 10 ²⁰
(inside)	Cs ¹³³	1.269×10^{17}	CsCl	73.2	1.3122×10^{18}
F	v ²³⁸	8 x 10 ¹⁹	ت ₃ 0 ₈	30.7	6.586 x 10 ¹⁹
(odge)	Cs ¹³³	6 x 10 ¹⁶	CaCl	73.2	1.5122 x 10 ¹⁸

Isotope Dilution Data for Samples E and P

EXPERIMENTAL RESULTS

(A) Introduction

It is of interest to compare the abundance of the cesium isotopes in the two parts of the rod with the amounts expected from U^{235} burn-up. With the help of the isotope dilution technique, the absolute numbers of atoms of the four cesium and three uranium isotopes in samples E and F have been found. For convenience of comparison the results were then expressed in terms of atoms per gran of uranium.

when a neutron is absorbed by U^{235} , either U^{236} is formed or fission occurs, the two events having a known ratio of probability. Thus the number of fissions that have occurred may be determined by measuring the loss of U^{235} or the gain in U^{236} during the irradiation. To predict the amounts of cesium isotopes accurately, requires a procise knowledge of the cross sections of U^{235} , Cs^{133} and Xe^{135} as well as an accurate value of the thermal neutron flux. In order to determine the depletion of U^{235} , a correction must be made for loss of U^{238} as a result of both fission and absorption. This can be done with a rough estimate of the flux and a knowledge of the cross section of U^{238} . The cross sections mentioned above are dependent on the neutron temperature, necessitating an estimate of the temperature of the heavy water modorator. The thermal neutron flux may be determined two ways, the merits of each will be discussed in a later section.

(B) Absolute Abundances

(a) Isotope Dilution

After they had been isotope diluted, samples E and F were analysed mass spectrometrically. The ratios of the 133, 135, 137 isotopes of cesium and the 235, 238 isotopes of uranium were measured. These ratios are shown in Table IV along with the pertinent ratios before isotope dilution, which were taken from Table II. The absolute number of atoms in each sample was then calculated and is given in the last column of Table IV.

(b) Abundances in Atoms per Gram of Uranium

For discussion of the results in Table IV it is convenient to express all abundances in terms of the number of atoms per gram of uranium present. This may be accomplished by calculating the weight of the U^{236} , U^{236} and U^{235} atoms for each sample in grams. The weight of uranium in samples E and F was calculated to be 5.306 x 10^{-2} g and 1.310 x 10^{-2} g respectively. The abundances of the cesium and uranium isotopes in atoms per gram of uranium have been summarized in Table V.

Since Cs^{134} decays with a half-life of 2.3 years, the abundance of Cs^{134} , at the time of removal from the reactor, Cs_0^{134} was calculated using the equation $Cs^{134} = Cs_0^{134} e^{-55^{\lambda_{134}}}$ where $55^{\lambda_{134}} = 0.301 y^{-1}$; and is given also in Table V.

(C) Uranium Cross Sections

(a) Calandria Temperature

When fission neutrons enter heavy water, they very quickly lose their energy by scattering, and assume a Maxwellian distribution of velocity corresponding to the same temperature as the heavy water molecules.

TABLE IV

Mass Spectrometric and Isotope Dilution Data For Samples E and F

Sample	Isotope	Ratio Before Isotope Dilution	No. of Atoms Added To Aliquot	Ratio After Isotope Dilution	Calculated No. of Atoms in Sample
E (inside)	cs ¹³³ cs ¹³⁵ cs ¹³⁷	1.186 ± 0.013 0.4444 ± 0.0038 1.0000	1.3122 x 10 ¹⁸ - -	.98.2 ± 1.5 0.4588 ± 0.0085 1.0000	1.604×10^{16} 6.008 x 10 ¹⁵ 1.352 x 10 ¹⁶
(Indiac)	ບ ²³⁵ ປ ²³⁸	0.0777 ± 0.0004 1.0000	- 2.152 x 10 ²⁰	0.02852 ± 0.00024 1.0000	9.695 x 10^{18} 1.247 x 10^{20}
F (edge)	Cs ¹³³ Cs ¹³⁵ Cs ¹³⁷	1.1179 ± 0.0045 0.3449 ± 0.0025 1.0000	1.3122 x 10 ¹⁸ - -	88.2 ± 1.3 0.347 ± 0.008 1.0000	1.685×10^{16} 5.198 x 10 ¹⁵ 1.507 x 10 ¹⁶
(2486)	ບ ²³⁵ ບ ²³⁸	0.0748 ± 0.0005 1.0000	- 6.588 x 10 ¹⁹	0.0239 ± 0.0002 1.0000	2.3096 x 10^{19} 3.088 x 10^{19}

TABLE V

Isotope	Abundance Sample E (Inside)	Abundance Sample F (Edge)		
v ²³⁸	2.346×10^{21}	2.352×10^{21}		
u ²³⁶	4.318 x 10 ¹⁸	5.453 x 10 ¹⁸		
u ²³⁵	$1,824 \times 10^{20}$	1.759 x 10 ²⁰		
Cs ¹³⁷	2.544×10^{17}	1.148×10^{18}		
Cs ^{137c*}	2.654 x 10^{17}	1.196 x 10 ¹⁸		
Cs ¹³⁵	1.130 x 10 ¹⁷	3.959 x 10 ¹⁷		
Cs ¹³⁴	4.158×10^{14}	5.492 x 10 ¹⁵		
Cs ^{134**}	6.271×10^{14}	7.715 x 10^{15}		
Cs ¹³³	3.018 x 10 ¹⁷	1.283 x 10 ¹⁸		

Abundances of Cesium and Uranium Isotopes in Atoms/g U

* Corrected for decay since formation $(t_{1/2} = 26.6 \text{ y}).$

** Corrected for decay since removal from the reactor $(t_{1/2} = 2.3 y)$. It has been found that neutron cross sections vary considerably with neutron velocity, and hence temperature; so that it is essential to know the temperature of the heavy water moderator. The moderator of the N.H.X. reactor is maintained at temperatures less than 50° C by circulating it to a light water heat exchanger. It has been estimated that the moderator had an average temperature of 40° C, over the irradiation period.

(b) Uranium-235

The effective cross section σ is defined by Vestcott (21) in terms of the density distribution function per unit velocity n(v) as follows:

$$\mathbf{\hat{\sigma}} = \frac{\int \mathbf{n}(\mathbf{v})\mathbf{\sigma}(\mathbf{v})d\mathbf{v}}{\mathbf{v}_{o}\int \mathbf{n}(\mathbf{v})d\mathbf{v}}$$
 where $\mathbf{v}_{o} = 2200 \text{ m/sec velocity}.$

Due to the velocity dependent definition of flux, it might be expected that σ , the effective cross section of a substance will vary as 1/v. Departures from this "1/v law" are usually in the form of resonances. It has been shown (22) that for a Haxwellian distribution of velocities σ -may be expressed as follows:

$$\sigma = \sigma(g + rs)$$

where g characterizes the departure of the substance from the 1/v law in terms of its effect on the Maxwellian spectrum, r characterizes the fraction of epithermal neutrons in the spectrum, s characterizes the departure of the substance from the 1/v law in the "resonance region" and σ is the cross section of the substance if all the neutrons were to have a single velocity v_{σ} .

It has been found (21) that the average epithermal index for a $M.R.\lambda$ fuel rod is r = 0.07, and since the epithermal neutrons emerge from the moderator, one would expect little variation of r across the

rod, With Westcott's tables (21), the values of the fission cross section $\hat{\sigma}_{\mathbf{r}}$, the capture cross section $\hat{\sigma}_{\mathbf{c}}$ and the total absorption cross section $\hat{\sigma}_{\mathbf{a}}$, have been calculated for $\mathbf{r} = 0.07$. The results are given in Table VI for $\mathbf{T} = 40^{\circ}$ C, the calandria temperature.

TABLE VI

Uranium-235 Cross Sections For r = 0.07 at $40^{\circ}C$

e _a	Sa	$\varepsilon_{ m P}$	s _F	$\hat{\sigma}_{a}$ barns*	$\hat{\mathbf{e}}_{\mathbb{F}}$ barns*	o barns
0.9698	0.0541	0.9702	-0.0368	675.2	563.9	111.3

*Using $\sigma_{\rm E}$ = 693.5 barns and $\sigma_{\rm F}$ = 582.8 barns, the world weighted average cross sections.

(a) Uranium-238

(i) Threshold Energy

Unlike U^{235} which has an even-odd nucleus, U^{238} has an even-even nucleus, therefore all its neutrons and protons are paired. U^{238} , which has potential barriers similar to U^{235} , releases less energy on capture of a neutron and therefore will require neutrons with at least 1 mev energy in order to cause fission. Both U^{238} and U^{235} have capture cross sections $\hat{\sigma}_{c}$ whereby the nucleus absorbs a neutron and forms enother isotope (U^{236} and U^{239} for U^{235} and U^{238} respectively). Below the 1 mev threshold energy of U^{238} essentially no fission will occur. At 6.75 ev, however, U^{238} has a large resonance absorption cross section. The moderator of a hotorogeneous reactor is arranged to slow the fission neutrons to energies below this and other smaller resonance energies, thus avoiding great neutron "losses".

(ii) Thermal Neutron Cross Section

The 2200 metre/sec neutron cross section σ_c of U^{238} is 2.74 barns. Since self-shielding leaves very few neutrons in the range of the resonance at 6.75 ev, the value of the effective neutron cross section $\hat{\sigma}_c$ is essentially the value above.

(iii) Fast Fission Factor

21.5

There is a chance that a high energy neutron released from fission will strike a U^{238} nucleus, before it leaves the rod to become thermalized. This probability is dependent on the dimensions of the rod. If the fast fission factor ϵ is defined as the ratio of the total neutrons from fission to the neutrons from U^{235} fission, then ϵ may be calculated with a knowledge of the mean free path of the neutron in the rod and the probability of its escape (23) without collision. For enriched $U0_2$, the fast fission factor was calculated to be $\epsilon = 1.014$.

(D) Noutron Flux

There are several methods of obtaining the flux of neutrons at any point in the rod, U^{235} , when it absorbs a neutron, either undergoes fission or becomes U^{236} . The ratio of the occurrence of the two processes is $\hat{\sigma}_{\rm F}^{1/6}$, which can be found from Table VI. The neutron flux may be determined either from the depletion of U^{235} or from the amount of U^{236} formed. The former method has the disadvantage that the percentage depletion of the U^{235} is small and hence is difficult to measure accurately.

(a) Uranium-235 Depletion

Assuming that the number of U^{238} atoms remained unaltored, it can be seen that

$$\frac{v^{235}}{v^{238}} = \frac{v_0^{235}}{v^{238}} e^{-\hat{g}_a} \phi t \qquad (i)$$

A further correction may be applied to this ratio for the fission of U^{238} . Since it has been shown that fissions of U^{238} account for only 1.45 of the total number of fissions, it can be seen that there is negligible depletion (0.017%) due to this effect. Therefore the value of U^{235}/U_{o}^{238} in column 5, Table VII, has been used to obtain a final value of ϕ using equation (i) as described before. The quantity U_{o}^{235} was then calculated and is shown, with ϕ , in Table VII, columns 7 and 6 respectively.

(b) Uranium-236 Formation

The value of the flux p' may also be obtained directly from the v^{236}/v^{235} ratio by solving the following two equations:

It is useful and often rigorous, in calculations involving the abundances of fission products, to use \overline{v}^{235} , the effective average number of v^{235} atoms present during the irradiation. \overline{v}^{235} has been calculated from equation (1), Appendix C, and is given in the last column of Table VII.

(E) Calculated Abundances

(a) Cesium-133

(i) Cross Section of Cs¹³³

At thermal neutron energies, the Cs^{133} cross section is small; probably obeying the 1/v law. There is, however, a resonance cross section of about 2500 barns (24) at 5.9 ev, in the "epithermal region". The Maxwellian distribution of energies, effective only in the thermal region, results in a cross section $\sigma = 29.0$ barns. Of the distribution of epithermal neutrons, a few will be close to this resonance energy. If

TABLE VII

Sample	$\frac{v^{235}_{o}}{v^{238}_{o}}$ (before)	$\frac{v^{235}}{v^{238}}$ (after)	ø* n/cm ² /sec x 10-13	$\frac{\sigma^{235}}{\sigma^{238}}$ (corrected)	ø n/cm ² /sec (from depletion) x 10 ⁻¹³	U ²³⁵ o atoms/g U xlC-20		<u></u> 235 atoms/g U x 10-20
E (inside)	0.08637	0.07772	0.923	0.07769	0,926	2.028	1.156	1.924
F (edge)	0.08637	0.0748	1.260	0.07476	1.261	2.034	1.511	1.894

Uranium Depletion and Neutron Flux Data

there were a large quantity of cesium in the rod, the neutrons around 5.9 ev would quickly be absorbed, changing the energy distribution. In the case of the UO₂ fuel rod, where the cesium present is only that due to fission, this effect is negligible. Bidinosti et al. (25) have measured the value of s in the equation $\sigma = \sigma(g + rs)$ where it was assumed that Cs^{133} is a 1/v detector with g = 1, and it was estimated that r, the epithermal index, was 0.026. From this, a value of the resonance integral Σ_{μ}^{μ} , the excess over the 1/v contribution was calculated using

$$s = \left(\frac{4T}{\pi T_o}\right)^{1/2} \frac{\sum_{\mu}^*}{o_o}$$

where $T_0 = 293^{\circ}$ C, giving $\Sigma_u = 320$ barns. Using the epithermal index r = 0.07, and a temperature of 40° C, the value of $\hat{\sigma}$ was found to be 55.4 barns for both samples in this work.

(ii) Expected Abundance

Equation (2), Appendix D, was used to calculate the expected abundance of Cs^{133} both for inside and outside the rod, with ${}_{54}\lambda^{133} =$ 1.508 x 10⁻⁶ sec⁻¹ and y¹³³ = 0.0659 (7). The results in atoms/g uranium are given in Table VIII where the expected abundances are tabulated beside the measured abundances which were taken from Table V.

(b) Cesium-134

Using equation (4), Appendix 9, the expected abundances of Cs^{134} were calculated for the time of removal from the reactor in terms of atoms/ g uranium. For these calculations values of $55s^{134} = 134$ barns (24) and $55\lambda^{134} = 0.301 \text{ y}^{-1}$ (26) were used, the results being given in Table VIII.

(c) Cesium-135

(i) Cross Section of Xe¹³⁵

In order to estimate the abundance of Cs^{135} , it is necessary to know the neutron cross section of its precursor Xe^{135} . The total neutron cross section of Xe^{135} has been measured for a variety of neutron temperatures independently by 3. Bernstein and by 2. C. Smith at Oak Ridge National Laboratories. Differences of about 10.5 exist in their results, there being no basis at present for favoring the data of either one or the other. Westcott has tabulated (21) the data, and with it has calculated the effective absorption cross sections. Fickel's (27) measurements of the effective cross section at two temperatures were found to be more consistent with the data of Bernstein. The effective cross section of Xe^{135} for a temperature of 40° G was obtained from a graph using Bernstein's results normalized to the two values obtained by Fickel, and was found to be 3.05 x 10^{6} barns.

(ii) Expected Abundance

With half-lives of λe^{135} and I^{135} as 9.20 h ($\lambda = 0.0753$ h⁻¹) and 6.70 h ($\lambda = 0.1034$ h⁻¹) respectively (26), and with $y^{135} = 0.0641$ as found by Petruska (7), the expected abundances of Cs^{135} for both samples E and F were calculated using equation (1), appendix B, and \overline{U}^{235} as recorded in Table VII. The results are given in Table VIII.

(d) Cesium-137

The amount of Ce^{137} formed, without regard to subsequent decay, was calculated from equation (3), appendix B, using $y^{137} = 0.0615$ (7) and \overline{U}^{235} as given in Table VII. The results are listed in Table VIII, along with the other data on cesium abundances.

TABLE VIII

Expected and Measured Abundances of Cesium in Atoms/g U

	Inside (Sample E)			Outside (Sample F)			
Isotope	lieasured Abundance	Expected Abundance	<u>Measured</u> Expected	Measured Abundance	Expected Abundance	Measured Expected	
Ca ¹³³	3.018 x 10 ¹⁷	1.410×10^{18}	0.214	1.283 x 10 ¹⁸	1.786 x 10 ¹⁸	0.718	
Co ¹³⁴	6.271 x 10 ¹⁴	7.338 x 10 ¹⁵	0.0855	7.715 x 10 ¹⁵	1.121 x 10 ¹⁶	0.688	
Cs ¹³⁵	1.130 x 10 ¹⁷	5.128 x 10 ¹⁷	0.220	3.959 x 10 ¹⁷	5.500 x 10 ¹⁷	0.720	
Cs ¹³⁷ corr	2.654 x 10 ¹⁷	1.327 x 10 ¹⁸	0,200	1.196×10^{18}	1.682 x 10 ¹⁸	0.711	

DISCUSSION

(A) Movement of Uranium Oxide

It has been mentioned in the introduction that, after irradiation, a cylindrical hole in the centre of the rod was found, and that the rod had cracked up without any destruction to the sheath. From early observations (10) of the phenomenon, it was thought that the central hole was caused by the melting of the uranium oxide. Although some rode achieve a centre temperature of 2760° C, sufficient for melting to occur, it is known from the power release per unit volume, and from the thermal conductivity of U0₂, that many rode, such as CR V-e which have central holes after irrediation, have stayed well below this temperature.

Experiments have been conducted by Auckern (8) at Westinghouse, to study the self diffusion of uranium in nearly stoichiometric UO_2 . This was done by vacuum depositing enriched UO_2 on to sintered natural UO_2 pellets, which were heated then analysed. Results indicate that the uranium diffused more than had been expected at $1615^{\circ}C$. It seems improbable, however, that the self diffusion of UO_2 can account for the movement of uranium oxide that must have taken place.

Any explanation for the formation of the holes in the centres of non-stoichiometric UO_2 rods must also account for the fact that stoichiometric UO_2 rods undergoing similar irradiation remain practically unchanged. Runnalls (1) has suggested that non-stoichiometric

oxide disproportionates at the hot interior, liberating gaseous UO_3 which then decomposes at a colder surface, liberating O_2 which then recycles. It is difficult to perceive, on thermodynamical grounds, why UO_3 should exist at the higher and decompose at the lower of two temperatures.

Although it is hard to understand the role of the non-stoichiometric oxide, it is well know to mass-spectroscopists that uranium oxides (probably $U_{3}O_{8}$) show considerable volatility even at low temperatures. For example, in the present work, the uranium oxide samples were evaporated from a tungsten filament at a temperature estimated to be 700° C, which is well below the temperature expected inside the rod. Therefore the hole in the centre could be accounted for by the volatilization and recrystallization of come form of uranium oxide from the centre onto cooler outer surfaces.

A possible explanation for the lack of movement in the stoichiometric UO_2 rod is also given by mass spectrometry. Although no studies have been made of emission from UO_2 , thorium dioxide and zirconium dioxide show no similar tendencies to volatilize until filament temperatures are in excess of about 2200°C. It is probable that stoichiometric UO_2 would behave in a similar manner. This explanation, however, must be subjected to more experimental confirmation.

(B) Re-evaluation of the Effective Capture Cross Sections of Uranium-235

Since both Cs^{133} and v^{236} are formed from neutron irradiation of v^{235} according to the relations:

 $Cs^{133} = \overline{U}^{235} \hat{\sigma}_{F} y^{133} \text{ ft}$ and $U^{236} = \overline{U}^{235} \hat{\sigma}_{C} \text{ ft}$, it is apparent that if the fission product Cs^{135} stays with the uranium

from which it was formed, that $U^{236}/C_{3}^{133} = \sigma_{c}^{\prime}/\sigma_{r}y^{133}$. This relationship has the asset that it depends on $\hat{\sigma}_{F}^{\prime}/\hat{\sigma}_{c}$ and y^{133} but not on the value ϕ t. The values of the U^{236}/C_{3}^{133} ratio at the centre and edge of the rod have been obtained from Table V, and are shown in Table IX along with $\hat{\sigma}_{c}^{\prime}/\sigma_{F}y^{133}$ found by using values from Table VI and the 6.59% fission yield given by Petruska et al. (7).

Evaluation of these ratios shows that there is no simple correlation between U^{236}/cs^{133} and $\hat{v}_{c}/\hat{v}_{c}y^{133}$ inside the rod, but at the edge, the difference is not so pronounced. This can be explained by the fractionation of Ce¹³³ with respect to v^{236} , by an error in the $\hat{\sigma}/\hat{\sigma}_{p}$ ratio or by an error in the fission yield. It has been found that the amounts of the isotopes of cecium at the edge are self-consistently low (see Table VIII) indicating no fractionation. For the basis of discussion it will be assumed that at the edge of the rod, the Cs^{137} and U^{236} have remained where formed. The possibility that y¹³³ differs significantly from the value quoted by Petruska, Thode and Tomlinson is very small, therefore it is worth re-examining the ratio $\hat{\sigma}_{a}/\hat{\sigma}_{p}$ which is related simply to α , the ratio of the 2200 metre/sec cross sections of U²³⁵ for capture and fission. It has been found that the two methods of finding ϕ , quoted in Table VII, which were dependent on the world weighted average cross sections σ_n and σ_r (24), are inconsistent in their results, indicating a probable error in of or or. Although the data found in this work will not give independent values of either σ_{F} or σ_{c} , a knowledge that σ_{a}/σ_{c} = U^{235} depleted/ U^{236} formed = 1 + $\hat{\sigma}_{\mu}/\hat{\sigma}_{\rho}$, will give a value of $\hat{\sigma}_{\mu}/\hat{\sigma}_{\mu}$ for both centre and edge; these are shown in Table IX. The values of σ_c/σ_y^{133} vere calculated using these new ratios and are shown in column 5, Table IX.

TABLE	13	

Data Concerning the Lotio U236/Ca153

	b ²³⁶ /ca ¹³³	o /6 3233	ô /ô, from	6 /8 3233	
Seaplo			1235 depletion		
E (incido)	14.51	2.995	6.269	4.00	
ਸ਼ (ਤਹੋਵਰ)	4.23	2.995	0.249	3.70	

"Cross sections oftained from Table VI.

. Waing values & /og from column 4.

Because the excitic leptotics required for obtaining the ratio $\hat{\sigma}/\hat{\sigma}$, cannot be obtained accurately from the scall changes in the t^{200}/t^{10} ratios before and after irradiction, the values of $\hat{\sigma}/\hat{\sigma}$, in orders 4, Table 13, are considered consistent. From Table 13, columns 2 and 3, it can be even that the two values of $\hat{\sigma}/\hat{\sigma}y^{100}$ found using the ratio of $\hat{\sigma}/\hat{\sigma}$, obtained in this work, are close to the ratio t^{200}/t^{000} found at the cutside of the rol. This is a further indication that the calculated ratio $\hat{\sigma}/\hat{\sigma}$, is the true value for the the true value of σ/σ , obtained in this work, are close to the ratio t^{200}/t^{000} found at the cutside of the rol. This is a further indication that the calculated ratio $\hat{\sigma}/\hat{\sigma}$, is the true value for the the true operator. That this rotic is such pressure than the 0.105 ratio of σ/σ , obtained that both has found (20) in the optimized region core resonances which have values of σ'/σ , > 1; notably, the resonance at 6 evolution (200 of $\sigma/\sigma = 0.25$ can be obtained from $\hat{\sigma}/\hat{\sigma}_{p}$ since $\hat{\sigma} = \sigma(z + rs)$. It will be noted that this value is not eigenfield from the average ratio of

 $\mathbf{\hat{v}}_{c}/\mathbf{\hat{v}}_{F} = 0.26$. In view of Bethe's observations noted above, the contribution to $\mathbf{\hat{v}}_{c}$ from epithermal neutrons was expected to be large, and therefore there may be some error in the calculation of $\mathbf{\sigma}_{c}$ using the data of Westcott. Hevertheless, the value of $\mathbf{\sigma}_{c}/\mathbf{\sigma}_{r} = \alpha$ has been shown in Table X along with other reported values. This value is apparently higher, but it is believed that this is the fault of the conversion of $\mathbf{\hat{\sigma}}_{c}$ to $\mathbf{\sigma}_{c}$. As noted above, the present data do not give either $\mathbf{\hat{s}}_{F}$ or $\mathbf{\hat{\sigma}}_{c}$ but only their ratio. It is of interest, however, to take an accepted value of $\mathbf{\sigma}_{c}$ (= $\mathbf{\sigma}_{F} + \mathbf{\sigma}_{c}$) and solve for the fission and capture cross sections individually. This has been done assuming that $\mathbf{\sigma}_{c}$ = 697 barns, the world weighted average. In Table X, values of $\alpha, \mathbf{\sigma}_{c}$ and $\mathbf{\sigma}_{c}$ (for 2200 metre/sec neutron velocity) measured in different countries are shown for comparison with the values of $\alpha, \mathbf{\sigma}_{c}$ and $\mathbf{\sigma}_{c}$ obtained in this work.

There is a wide variation in the reported values of σ_{Γ} ranging from 555 to 596 barns. The value 557 barns reported in this thesis is consistent with these results. The value of σ_{c} , however, is considerably higher than those reported by other workers, which is to be expected if σ_{c} was incorrectly evaluated from σ_{c} .

(C) novement of Cesium

Research is in progress in Chalk River by Edwards and Haukings, using counting techniques; and by Hart and Horgan, using mass spectrometric techniques, to try to ascertain the reason for the movement of the cesium. They have reported (4) analyses on several rods, including CR V-e rod 7. Counting experiments have given abundances of Cs¹⁵⁷ and

Quantity	This Work Assuming $\sigma_{a} = 697*$	Vorlâ Veighted Average	U.S.A.	U.K.	U.S.D.R.	India	France	Canada
$\alpha = \frac{\sigma_c}{\sigma_p}$	0.25 ± 0.01	0.183 ± 0.01	0.18 ± 0.01	0.21 ± 0.03	0.22 ± 0.04	in ordered		
σ barns	697 ± 6	697 ± 6	689 ± 7	725 ± 13	695 ± 20			
σ _F barns	557 ± 7	579 ± 4	584 ± 10	596 ± 14	570 ± 15	555 ± 15	58 ¹ + ± 20	578 ± 6
o barns	140 ± 6	118 ± 7	1 05 ± 12	129 ± 16	125 ± 25			

TABLE X

World Values of Neutron Cross Sections of ${\mathbb U}^{235}$

*697 barns is the world weighted average capture cross section for U^{235} (24).

 Ce^{134} at positions incide and at the edges of the rods. From the mass spectrometric work, only the abundances of the Cs^{137} have been reported. Since these analyses were performed on samples from various different regions in rod 7, the results are not strictly comparable, but in fact, do show the same trends.

From Table VIII it can be seen that at the edge of the rod, all the cesium isotopes measured are in quantities less than expected from calculations involving the flux and fission cross sections. It can also be seen that they are each low by nearly the same emount. Since the edge of the rod was nearly at room temperature, about the melting point of cesium, and the centre of the rod was above the boiling point, any cesium formed in the centre yet not trapped in the lattice would diffuse toward the cooler edge. At a certain point along the radius, where the saturation pressure of the cesium was reached, the vapour would condence onto grains of UO₂. At greater distances from the centre, cesium would virtually remain where it was formed; or would diffuse slowly, toward the edge perhaps depositing on the choath.

Thus one can expect, in a certain radial zone between the centre and edge, a large excess of cesium. If the expected abundances of the cesium isotopes are calculated using the value of $\hat{\sigma}_{\rm p}$ obtained from these experimental data (column 2, Table X) and using the flux obtained from U^{235} depletion (column 6, Table VII), it is found that at the edge the ratio of the observed number of atoms to the expected number of atoms, increases from about 0.7 to 0.9. It therefore appears that the cosium at the edge pallet has essentially remained where it was formed, or perhaps a small fraction has moved towards the sheath.

The inside pellet has shown that Cs^{133} , Cs^{135} and Cs^{137} have depleted by a factor of about 5, which is in agreement with the ideas expressed above; whereas the Cs^{134} has depleted by a factor of about 12. Since Cs^{134} has the same chemical properties as the other design isotopes, and since any isotope fractionation effect must be more pronounced in the heavier or lighter isotopes, it may be concluded that no obvious property of the Cs^{134} can account for its abnormal behaviour. An explanation for this effect can be found when it is noted that the main difference between the formation of the Cs^{134} and the formation of the other isotopes is that Cs^{134} is formed from Cs^{133} with the release of high energy gamma rays, whereas the others are formed by beta decay from xenon. Thus the effect may be attributed to the energy of receil given to the Cs^{134} , since, if this energy were of no importance, the Cs^{134} would have travelled in the same mode as its precursor.

In the general movement of UO_2 from the centre of the rod, the xenon formed by fission, that is trapped in the UO_2 lattice, is clouly exposed. As a gas, it can travel in the porce of the UO_2 . When the xenon decays to cesium, however, a high energy electron is released, giving the cesium a receil energy of a few electron volts. This energy, different for each occium isotope, is enough to knock the cesium atom a little way, where it perhaps embeds itself in the UO_2 surface. Thermal energy will be sufficient to release most of the cesium in this hot region, and as a vapour, it diffuses to a cooler zone where it condenses. When a Cs^{134} atom is formed from a Cs^{133} atom that is trapped in the lattice in the hot region of the red, however, it is given a large receil

energy. This could be sufficient to release the Cs^{134} , which also diffuses to the cooler zone, leaving behind Cs^{133} atoms which cannot escape. This accounts for the greater relative depletion of Cs^{134} at the centre of the rod.

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APPENDIX

(A) Correction of the Matios Cs¹³³/Cs¹³⁷ and Cs¹³⁵/Cs¹³⁷ for Cs¹³⁷ Decay Since Cs¹³⁷ has a half-life of 26.6 years (18) the measured ratio Cs¹³³/Cs¹³⁷ must be multiplied by a "correction factor" to give the Cs¹³³/ Cs¹³⁷⁰ ratio of fission, where Cs^{137c} is the number of Cs¹³⁷ atoms formed from fission without regard to subsequent decay. Since the precursors of Cs¹³⁷ in the 137 mass chain have very short half-lives, the whole yield of the 137 mass chain soon becomes Cs¹³⁷. A correction factor to allow for decay after the formation of the Cs¹³⁷ may be found by the solution of the following differential equations:

 $\frac{d}{dt} \operatorname{Cs}^{133} = \overline{v}^{235} \, \widehat{\sigma}_{F}^{\phi} \, y^{133} \qquad \frac{d}{dt} \operatorname{Cs}^{137} = \overline{v}^{235} \, \widehat{\sigma}_{F}^{\phi} \, y^{137} - \operatorname{Cs}^{137} \, {}_{55}^{\lambda^{137}}$ Here \overline{v}^{235} is the average total number of v^{235} atoms present, y^{133} and y^{137} are the percent cumulative yields of the 133 and 137 mass chains respeqtively, ϕ is the 2200 metre/sec neutron flux, $\widehat{\sigma}_{F}$ is the effective thermal neutron fission cross section of v^{235} and ${}_{55}\lambda^{137}$ is the decay constant of Cs^{137} . If t_1 is the length of time the uranium was in the reactor, then at the time of removal from the reactor,

$$\frac{Cs^{137}(t_1)}{Ca^{133}(t_1)} = \frac{1}{55^{\lambda^{137}t_1}} \left(1 - e^{-55\lambda^{137}t_1}\right) \frac{y^{137}}{y^{133}}$$
(1)

However, while the Cs^{133} remains the same after the rod is removed from the reactor, the Cs^{137} decays for a further time t_2 until analysis.

Since $y^{133}/y^{137} \approx Cs^{135}/Cs^{1370}$, therefore

$$\frac{c_{\rm g}^{133}}{c_{\rm g}^{137\rm c}} = \frac{1}{55^{\lambda^{137}\rm t_1}} \left(1 - e^{-55^{\lambda^{137}\rm t_1}}\right) e^{-55^{\lambda^{137}\rm t_2}} \frac{c_{\rm g}^{133}}{c_{\rm g}^{137}}$$
(2)

The same factor may be used to correct the Cs^{135}/Cs^{137} ratio assuming that t_2 is long compared to the half-lives of the precursors of Cs^{135} .

(B) To Calculate
$$Cs^{135}/y^{135}$$
 in Terms of $54s^{135}\phi$

The decay scheme may be represented by the following diagram:

Fission
$$\longrightarrow_{53} I^{135} \xrightarrow{\lambda}_{54} Xe^{135} \xrightarrow{\sigma\phi}_{55} S^{135} \xrightarrow{55} S^{135}$$

The ratio of the number of Cs^{135} atoms formed in thermal neutron fission of U^{235} to the total number of atoms in the 135 mass chain may be found as a function of ${}_{54}s^{135}\phi$ by the solution of the following differential equations:

$$\frac{d}{dt} c_{B}^{135} = X e^{135}_{54} \lambda^{135}$$

$$\frac{d}{dt} X e^{135} = I^{135}_{55} \lambda^{135} - X e^{135}_{54} \lambda^{135} - X e^{135}_{54} \sigma^{135} \phi + (0.05) v^{235} \hat{\sigma}_{F} \phi_{V}^{135}$$

$$\frac{d}{dt} I^{135} = (0.95) v^{235} \hat{\sigma}_{F} \phi_{V}^{135} - I^{135}_{55} \lambda^{135}$$

$$\frac{d}{dt} v^{235} = - v^{235} \hat{\sigma}_{F} \phi$$

where λ and σ are the decay constants and thermal neutron cross sections of the elements indicated by their superscripts and subscripts, and $\hat{\sigma}_{\mu}$ and $\hat{\sigma}_{\mu}$ represent the total effective neutron absorption and fission cross sections respectively. It has been assumed that there is a 5% primary fission yield of Xe^{135} as estimated from the Glendenin hypothesis of equal charge displacement (19), the other 95% become I^{155} either directly or almost immediately. It has also been assumed that at the time of analysis, all the iodine and xenon had long since decayed into the Cs^{135} , and that since the U^{235} depletion is small, $U^{235} = U_0^{235} e^{-\hat{\sigma}_2 \phi t}$ may be replaced by \overline{U}^{235} , the average number of U^{235} present during the irradiation as calculated in Appendix C.

The ratio of the number of Cs^{135} present to the total number of atoms of mass 135 produced, Y^{135} is

$$\frac{c_{s}^{135}}{r^{135}} = \frac{54^{\lambda^{135}}}{K} + \frac{54^{\sigma^{135}}}{t} \left[\left(\frac{1 - e^{-Kt}}{K^{2}} \right) + \frac{0.95(1 - e^{-53^{\lambda^{135}}t})}{53^{\lambda^{135}(K - 53^{\lambda^{135}})}} - \right]$$

$$\frac{0.95(1 - e^{-Kt})}{K(K - 53^{\lambda^{135}})}$$
(1)

where $Y^{135} = y^{135} \not o \ o_{F} t \ \overline{u}^{235}$ Since $\frac{54^{\lambda^{135}}}{K} \gg \frac{54^{\sigma^{135}} \not o}{t} \left[(\frac{1 - e^{-Kt}}{K^2}) + \frac{0.95(1 - e^{-53^{\lambda^{135}}t})}{53^{\lambda^{135}(K} - 53^{\lambda^{135}})} - \right]$

$$\frac{0.95(1 - e^{-Kt})}{K(K - 53^{\lambda^{135}})}$$

$$\frac{\text{Then}}{c_{9}^{135} \approx} \frac{r^{135}_{54} \lambda^{135}}{\kappa} = \frac{y^{135} \hat{\sigma}_{F} \not p_{t}}{54^{\lambda^{135}} + 54^{\sigma^{135}} \not p_{t}^{235}}$$
(2)

The number of Cs^{137} atoms formed without regard to subsequent decay, Cs^{1370} can be found by solution of the following differential equation:

$$\frac{d}{dt} c_{s}^{1370} = v^{235} \delta_{F} \phi y^{137}$$
whence $c_{s}^{1370} = \overline{v}^{235} \delta_{F} y^{137} \phi t$ (3)

when \overline{U}^{235} is defined in Appendix C. From equations (2) and (3)

$$\frac{c_{s}^{135}}{c_{s}^{1370}} = \frac{54^{\lambda^{135}}}{54^{\lambda^{135}} + 54^{\sigma^{135}}} \frac{y^{135}}{y^{137}}$$
(4)

with the value of $y^{135}/y^{137} = 1.043$ (20) and the decay constant of Xe¹³⁵ (9.20 hr) = 0.0753 hr⁻¹ the value of $54^{\sigma^{135}/p}$ may be calculated.

(C) The Amount of U²³⁵ Averaged Over the Time of Irradiation

The amount of uranium, \overline{U}^{235} , which, if it were to remain constant during the irradiation, would give an equivalent number of fission to that observed, can be found by solving

$$\overline{\overline{y}}^{235} = \frac{\int_{0}^{t} y^{235} dt}{\int_{0}^{t} dt}$$

where $U^{235} = U_0^{235} e^{-\hat{\sigma}_2 \beta t}$. Thus $\overline{U}^{235} = \frac{U^{235}}{\hat{\sigma}_2 \beta t} (1 - e^{-\hat{\sigma}_2 \beta t})$ (1)

(D) Expected Abundances of Cesium-133 and Cesium-134

The production of Cs¹³³ and Cs¹³⁴ may be traced from the following diagram:

Fission
$$33^{133}$$
 $\frac{133}{\frac{1}{2}} = 20 \text{ h} 54^{Xe^{133}} \frac{1}{\frac{1}{2}} = 5.3 \text{ d} 55^{Ca^{133}} \frac{1}{3} \frac{1}{3} \frac{1}{2} = 5.3 \text{ d} 55^{Ca^{133}} \frac{1}{3} \frac{1}{3$

The mashers of $G_2^{1,55}$ and $J_3^{1,54}$ expected from $U^{2,55}$ firston may be obtained by the solution of the following four differential constions:

$$\frac{d}{dt} U^{235} = -\sigma_{\mu} \not d U^{235}$$

$$\frac{d}{dt} \lambda e^{135} = U^{235} \sigma_{\mu} \not d y^{135} = \lambda \lambda e^{135}$$

$$\frac{d}{dt} c e^{135} = \lambda e^{135} 1 - c e^{135} \sigma \not d$$

$$\frac{d}{dt} c e^{154} = c e^{153} \sigma \not d - c e^{154} \Delta$$
where $\sigma_{a} = g_{2} \hat{\sigma}_{a}^{235}$, $\sigma_{F} = g_{2} \hat{\sigma}_{F}^{235}$, $\lambda = g_{4} \lambda^{135}$, $\sigma = g_{5} \hat{\sigma}^{133}$ and $\Delta = g_{5} \lambda^{134} + \beta g_{55} \sigma^{134}$.

The number of Co¹³⁵ present of the end of an irradiation of duration t is

$$Ce^{133} = \frac{P}{p(\sigma - \sigma_{a})} \left[e^{-\sigma_{a} p t} - e^{-\sigma_{a} p t} \right] = \frac{P}{(\sigma_{a} - \lambda)} \left[e^{-\lambda t} - e^{-\sigma_{a} p t} \right]$$
(1)
where $P = \frac{\lambda U_{a} y^{133} p \sigma_{p}}{\lambda - \sigma_{a} p}$.

at a time after the irreliation loag compared to the half-life of Xe^{155} (5.54), nearly all the Xe^{155} will have decayed to Ce^{155} . Then, aince $\lambda t = 25$ for this irrediation $e^{-\lambda t} \approx 10^{-11}$ is neglected; and since $\lambda \gg o\beta$, $\lambda = o\beta \approx \lambda$.

The abundance of Ga¹⁵⁵ at a time we ha after reacted from the reactor is

$$c_{3}^{133} = P\left(e^{-\sigma_{3}/t} - e^{-\sigma/t}\right) \left[\frac{1}{P(\sigma - \sigma_{1})} + \frac{1}{2}\right]$$
(2)

The rigorous equation for the number of Cs^{134} at the end of the irradiation, Cs^{134}_{0} , is

$$\frac{c_{0}^{134}}{\sigma F} = \frac{1}{(\sigma - \sigma_{F})(\Delta - \sigma_{a}\phi)} \left[e^{-\sigma_{a}\phi t} - e^{-\Delta t} \right] - \frac{\phi}{(\sigma \phi - \lambda)(\Delta - \sigma \phi)} \left[e^{-\sigma \phi t} - e^{-\Delta t} \right] + \left[\frac{\phi}{(\sigma \phi - \lambda)(\Delta - \sigma \phi)} - \frac{1}{(\sigma - \sigma_{a})(\Delta - \sigma \phi)} \right] \left\{ e^{-\sigma \phi t} - e^{-\Delta t} \right\}$$
(3)

Equation (3) as it stands is difficult to use since it involves multiple subtraction of approximately equivalent terms, the difference of which is the abundance of Cs^{134} . A rigorous expansion allows cancellation of many terms. Making the assumption that $e^{-\lambda t} = 0$, the following expansion of equation (5) has been obtained.

$$\frac{\mathbf{cs}_{0}^{134}}{\mathbf{\sigma}F} = \frac{\mathbf{t}^{2}}{2} \left[\left\{ 1 - \frac{\mathbf{t}\Delta}{3} + \frac{\mathbf{t}^{2}\Delta^{2}}{12} - \frac{\mathbf{t}^{3}\Delta^{3}}{60} \dots \right\} - \frac{\mathbf{t}\phi}{3} (\mathbf{\sigma} + \mathbf{\sigma}_{a}) \left\{ 1 - \frac{\mathbf{t}\Delta}{4} + \frac{\mathbf{t}^{2}\Delta^{2}}{20} \dots \right\} \right. \\ \left. + \frac{\mathbf{t}^{2}\phi^{2}}{12} (\mathbf{\sigma}^{2} + \mathbf{\sigma}\mathbf{\sigma}_{a} + \mathbf{\sigma}_{a}^{2}) \left\{ 1 - \frac{\mathbf{t}\Delta}{5} + \frac{\mathbf{t}^{2}\Delta^{2}}{30} \dots \right\} \dots \right] \\ \left. + \frac{\phi}{\mathbf{\sigma}\phi} \frac{\mathbf{t}}{-\lambda} \left[\mathbf{t} \left\{ 1 - \frac{\mathbf{t}}{2} (\Delta + \mathbf{\sigma}\phi) + \frac{\mathbf{t}^{2}}{6} (\Delta^{2} + \Delta\mathbf{\sigma}\phi + \mathbf{\sigma}^{2}\phi^{2}) - \frac{\mathbf{t}^{3}}{24} (\Delta^{3} + \dots) \right\} \right. \\ \left. + \frac{1}{(\Delta - \lambda)} \left\{ 1 - \mathbf{t}\Delta + \frac{\mathbf{t}^{2}\Delta^{2}}{2} - \frac{\mathbf{t}^{3}\Delta^{3}}{6} \dots \right\} \right]$$
(4)

FIGURE 1



Cross Sectional Photograph of Rod 7 Magnified Four Times