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Isotopic Correlation for Accounting and Control of Nuclear Materials In a Fuel Cycle

— A Review Paper —

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ABSTRACT

The isotopic correlation techniques (ICT) are emerging as an important supporting measure for the accounting and control of nuclear materials which pass through commercial fuel cycle for the production of nuclear energy. The present paper gives an overview on the development of the ICT and discusses their possible uses in various parts of a commercial fuel cycle. It is shown that ICT can be used advantageously both for the purposes of plant operation and safeguards. The paper ends with the conclusion that ICT may be particularly useful for the fuel cycle of the 1980's and that they can be utilized to the fullest advantage for safeguards when fuel cycle is considered as a whole.

Isotopenkorrelation für Buchführung und Kontrolle des nuklearen Materials in einem Brennstoffzyklus

ZUSAMMENFASSUNG

Die Isotopenkorrelationstechniken (IKT) erscheinen mehr und mehr als wichtige Zusatzmaßnahme für die Buchführung und Kontrolle des Kernmaterials, das einen kommerziellen Brennstoffzyklus zur Energieerzeugung durchläuft. In dem vorliegenden Bericht wird ein Gesamtüberblick über die Entwicklung der IKT sowie über die möglichen Anwendungen in verschiedenen Teilen des Brennstoffzyklus gegeben. Es wird gezeigt, daß diese Korrelationen sowohl für betriebliche Zwecke wie auch für Zwecke der Überwachung vorteilhaft verwendet werden können. Der Bericht endet mit der Bemerkung, daß die IKT besonders nützlich für den Brennstoffzyklus der 80-er Jahre sein wird und daß die Vorteile der IKT erst dann am günstigsten für die Überwachung verwendet werden können, wenn der Brennstoffzyklus in seiner Gesamtheit betrachtet wird.

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A Review Paper

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1. Introduction

Right from the time the nuclear material became known to be fissionable, the question of its accountancy and control has played an important role. Because of the possibility of its destructive use and also of its value, use of nuclear material is always accompanied by some accountancy and control measures.

Large scale commercial application of nuclear materials will be an integral part of the industrial landscape of many a country for many years to come. The amounts of nuclear materials involved in use and transport will increase manifold over those at the present time. The Pu production alone (from nuclear power plants) is expected to be about 65 - 70 t/a in 1980 in the western world. With increasing amounts and movements of nuclear materials, the problems associated with the accountancy and control of these materials increase also. Intensified efforts are required to keep these measures within reasonable limits.

The isotopic correlation techniques (ICT) are emerging as an important supporting measure for the accountancy and control of nuclear materials which pass through commercial fuel cycle facilities for the production of nuclear energy. In passing through the different steps in a fuel cycle, the nuclear material (U or Pu) may undergo a nuclear transmutation or changes in its isotopic composition. Three steps in such a fuel cycle are involved in this connection:

Steps in the fuel cycle	Types of changes between the initial and the final process streams
Uranium enrichment	Changes in the isotopic concentrations and ratios of U-234, U-235, U-236, U-238 in tails and product streams.
Reactors	Changes in the concentrations of U-234, U-235, U-238; production of U-236, Pu-239, Pu-240, Pu-241, Pu-242 etc.; production of fission products in irradiated fuel elements.
Reprocessing	Changes which occured in irradiated fuel elements in the reactors are measured for the first time at the accountability tank of a reprocessing plant. Some changes in isotopic composition of the input solution can be caused by recycling of process solutions into the input accountability tank.

2. Isotopic Correlations in Enrichment Plants

The changes in the isotopic composition (and the ratios of the different isotopes) in the product and the tails streams from an uranium enrichment plant for a given isotopic composition of the feed stream, are caused by the operating conditions in the cascades of such a plant. If these conditions were to be kept constant, the concentrations and the ratios of the isotopes in the tail and product streams, for the given feed stream are also expected to remain constant. Inversely, if the isotope ratios (e.g. U-235/U-234) in the tail/product streams for the same feed stream were found to be different to those found under normal process conditions, one could conclude in principle that the process conditions had changed. If therefore, a correlation between the isotopic ratios of the feed, product and tail streams on the one hand, and the process characteristics or parameters of the enrichment plant on the other, could be established on the basis of theoretical and experimental investigations, such correlations could be used to verify the operation characteristics of a cascade of an uranium enrichment plant with the help of measured isotopic ratios of the different

process streams. In case a necessity existed for controlling the operation characteristics of a plant as declared by the plant operator, such a possibility would provide a simple way of verifying or controlling a complex phenomena; since measurement of isotopic ratios in process streams from an enrichment plant is much simplier than controlling the intricate process characteristics of a cascade of the same enrichment plant through some other means.

This point is illustrated in Fig. 1 as developed by Blomkin and v. Halle /1/.They showed that in an ideal diffusion plant cascade for the enrichment of U-235, the ratio of U-235/U-234 concentrations for a given U-235 concentration in the tail stream or in the stream at any other particular point of the same cascade, depends on the U-235 concentration at the top of that cascade. Thus for example, in a cascade with a top U-235 concentration of 90 %, the ratio of U-235/U-234 at a point in the cascade with 4 % U-235 concentration is expected to be 170. In another ideal cascade with a top U-235 concentration of 4 %, the ratio of U-235/U-234 concentration for this stream is on the other hand, found to be 110. If therefore, a 4 % U-235 stream from an inaccessible diffusion plant cascade showed a U-235/U-234 concentration ratio of 170, one could theoretically infer that the 4 % stream was being produced in a cascade with a top concentration of 90 % U-235. In case the ratios could be measured accurately enough to reduce the probability of a false influence and furthermore, the ratios were an unique function of the cascade arrangement under actual operating condition, such a correlation could indeed provide a powerful mean in controlling the operational characteristics of an enrichment cascade with the help of such ratio measurements.

The example on the enrichment cascade is meant to illustrate the basic principle underlying the applications of ICT for accountancy and control of nuclear materials in a commercial nuclear fuel cycle. As shown above, at two of the facilities in such a cycle namely, enrichment and reactors, isotopic shifts in the feed materials as well as transmutation and production of new isotopes take place as an inherent part of the processes in these facilities. A number of straight forward correlations between the ratios of such isotopes or elements and some of the important process parameters (e.g. cascade arrangements in an enrichment plant, burn up or fission of feed elements in a reactor)

have been observed to exist. Correlations have also been shown to exist among ratios, differences and concentrations of isotopes involved in such processes. If such correlations are found to be real and unique, they could be used as redundant and complementary measures to the normal measures of accountancy and control for nuclear materials.

Although isotopic shifts may take place in fabrication facilities for fuel elements because of blending operations, they are not inherent of the process and therefore, cannot produce unique correlations with any process parameters. Since most of the correlations investigated sofar, has been in the area of reactor and reprocessing facilities in a nuclear fuel cycle, the rest of the discussions in this paper will be concentrated on the different types and use of ICT in this area.

3. Isotopic Correlations in Reactor-Reprocessing Facilities of a Nuclear Fuel Cycle

In a commercial fuel cycle of the present day, the fuel elements fabricated in fabrication plants may contain natural, enriched and recycled uranium. They may also contain recycled plutonium. Normally the amounts of nuclear materials and of the more relevant isotopes (U-235, U-238, Pu-239 etc.) are known in these fresh unirradiated fuel elements with fairly high accuracies (better than 0.5 % 1 σ-value) /46/. The irradiation of these nuclear materials in the reactor causes complex changes in its composition. They include a depletion of uranium and plutonium (if initially present) build up and transmutation of artificial isotopes like U-236, Pu-239, Pu-240, Pu-241, Pu-242 and build up of fission products (isotpes of light elements like Cs-127, Xe-132, Nd-148 etc.). The fact that simple functional relationship exists between the initial and final contents of some of the heavy isotopes, was known since about 15 years /2/. Both burnup experiments and calculational burnup codes have been used to study the transmutation of uranium and plutonium. The transmutation of isotopes is principally governed by simple first order differential equations. However, the coefficients of these equations depend on numerous core details and operating conditions. The complexity of core models and of approximations needed to solve the equations had obscured the indication of simple relationships in the past. It is however an interesting fact that these correlations if averaged over a whole batch of fuel elements or a whole core of a reactor, remain simple and can be established using the measurement results from a reprocessing plant.

What is more, they are mostly a unique feature for a particular core in question. Evidently, the nature is not encumbered by core models, cross-sections etc. and produces highly consistant relationships.

3.1 Historical Background

The existance of correlation between Pu/U ratio and depletion of U-235 and the possibility of their use for accountancy purposes were shown in 1969 by Christensen and Schneider /3,6/ of Battelle Pacific Northwest Laboratories, Richland, Washington and Moeken and Bokelund /4,5/ of EUROCHEMIC Reprocessing Plant, Mol, Belgium. The state of the art of the isotopic correlation techniques was summarized in a number of presentations at the Symposium on Safeguards Techniques at Karlsruhe in 1970 /7/. In 1972 the Atomic Energy Agency held a Working Group Meeting on the Use of Isotopic Composition Data for Safeguards with participation of experts from 8 countries and a number of international and national organizations /8/. A review of the isotopic correlation data was made and experts' opinions were sought on the possible use of isotopic correlation methods in the following five areas related to safeguards:

a) Verification of Pu input to reprocessing plants by correlation of isotopic data

b) Confirmation of information using correlation of isotopic data

c) Identification of nuclear materials using isotopic data

d) Dynamic determination of the process inventory

e) Other applications of isotopic correlations

The most important result of this working group was the corroboration of the fact that some correlations, notably the Pu/U-U-235 Depletion correlations, have been shown to have a good degree of consistancy and reliability in some selected systems, particularly the PWR system. Some other correlations have shown less consistancy and some reactor systems have not yet been examined in any depth. The group recognized the merits of the use of isotopic correlations.

However, for a full utilization of the technique, a continuous collection of data is required. A considerable amount of work on correlations was carried out in a number of countries during the period 1969-1975. Particular mention should be made of the work carried out by Battelle Northwest Laboratories, Richland, Washington, CEN, Mol, Belgium, IAEA, Vienna, EURATOM Joint Research Centers at Ispra, Italy and Karlsruhe, FRG, and Nuclear Research Center, Karlsruhe, FRG. They are very adequately summarized by Berg et al. /9/ and Christensen and Schneider /10/. Some of the isotopic correlations have been in use at Windscale, UK for operational purposes for a long time /30/. In the past few years work on heavy isotope correlations for BWR systems has been carried out by H. Umezawa et al. at the Japanese Atomic Energy Research Institute, Tokai Mura /49/.

The possible use of fission product correlations such as Xe-132/Xe-131, Kr-84/Kr-83 was pointed out by Koch et al. in 1970 /11/, working at the EURATOM Transuranium Institute, Karlsruhe. The data base for this type of correlations has been elaborated since then /12,13,14,15,16/. The main advantage of using this correlations may lie in the fact that integrated samples can be taken from the off gas stack from a reprocessing plant with very little intrusion to the plant operation.

Correlations based on selected fission product activities (e.g. Cs-137) or activity ratios (e.g. CS-134/Cs-137) have also been found to exist for burn-up or Pu/U ratios /17,18,37/ by Beets and Dragnev among others. The advantages for the use of such correlations may lie in the fact that they can be tested in intact irradiated fuel assemblies in the wet storage of a reactor or a reprocessing facility.

Detailed theoretical investigations have been carried out in the USA, at GfK, Karlsruhe, and at JRC-Ispra /19,20,21,22,23,24,25/. The main purpose of these investigations is to obtain a better physical understanding of the relevant parameters of reactor design and operation which influence such correlations. Other purposes are to provide adequate statistical representations of the available correlation data and to develop adequate physical models in order to cover the total spectrum of isotopic correlations existing in the spent fuel of the various types of reactors /25,26/.

Both the Atomic Energy Agency, Vienna and the European Safeguards Research and Development Association (ESARDA) /27/ have established data banks for informations on isotopic correlations. A part of the data bank of the ESARDA which is located at Ispra, has already started functioning.

3.2 Technical Basis for Correlations

It was stated earlier that the reason for the existance of isotopic correlations in the reactor-reprocessing area of a nuclear fuel cycle is the fact that,the production and depletion of isotopes in a nuclear reactor are basically governed by first order differential equations. The equations are of the type:

$$\frac{dN^i}{dt} = C^{i-1} - A^i - D^i \qquad \ldots\ldots \qquad (1)$$

where dN indicates the average change in the concentration of the isotope i in a given reactor volume after it has been irradiated in the reactor for a period of dt. The letters C, A and D represent the macroscopic reaction rates for the build up, absorption and radioactive decay of the isotope i. The index i-1 indicates an isotope which produces the isotope i through neutron capture and eventual β-decay. The production of the isotope i through decay of some higher isotopes can be neglected.

The simple burnup relation as given in eqn. (1) is complicated in actual core calculations as the macroscopic reaction rates are functions of many core parameters such as initial concentrations of heavy isotopes, neutron spectrums and leakage rates, moderator to fuel ratio, structural materials present etc. and they vary both spatially and in time. Such calculations are normally carried out for a relatively small core volume and small time intervals using the microscopic cross sections of the isotopes involved. For these small values, the correlations do vary from place to place. The relations between the plutonium buildup and U-235 depletion in a pellet at the top of a fuel element may for example be very much different to that at the middle of the same fuel element. For a macroscipic integration on a theoretical basis, so many factors need to be taken into consideration, so many simplifications and approximations have to be made, that exact functional relationships for the different types of correlations can no longer remain transparent and also cannot be traced back even though they may exist in reality. Because of these fundamental difficulties, isotopic correlations did not evolve as an exact science but developed on an empirical and phenomenological basis.

The first relations were established empirically /3/ on the basis of measurement results from the input accountability tank of a reprocessing plant. In general these measurements play a fundamental role in the establishment of isotopic correlations. Since one or more complete fuel elements from a reactor system are chemically dissolved as a batch in the accountability tank, the differential influences of most of the reactor parameters are integrated and averaged in the batch. The hidden relations are unmasked again and quite often, consistent correlations are obtained from these results.

Up to the present time two approaches have been followed to obtain stable and consistant isotopic correlations:

a) Collection of measured data obtained in reprocessing plants for fuel elements or special samples from different types of reactor systems and establishment of correlations from a set of these measured data for different combinations of isotopes, isotopic ratios and burnup parameters. The data base for the establishment of correlations has to be expanded regularly to improve the established correlations and to produce new ones.

b) Theoretical analysis of the influence of relevant reactor parameters for different reactor types on the depletion and build up of the isotopes of interest as well as, statistical evaluation and model development for the use of different sets of data.

Both these approaches are followed up currently at four places: Battelle Pacific Northwest Laboratories, Richland, Washington; EURATOM Research Centers, Ispra and Karlsruhe; Nuclear Research Center, Karlsruhe, FRG and CEN, Mol, Belgium. Investigations have also started at the Japan Atomic Energy Research Institute, Tokai Mura, Japan.

3.3 Types of Isotopic Correlations Established Sofar

The isotopic correlations established sofar, may be broadly devided into three categories depending on the types of isotopes and the source of the measured data which can be used for establishing correlations.

1) Correlations based on isotopes of heavy elements: Most of the necessary isotopic data are obtained from routine measurements at the reprocessing plant input. Fairly large data base has been created and a number of correlations are routinely used.

2) Correlations based on isotopes of gaseous fission products: At present necessary isotopic data can only be obtained by means of specific additional measurements at the reprocessing plant input (for example Kr, Xe). The situation could change if because of environmental restrictions fission gas evolved during reprocessing had to be stored. In that case isotopic measurement before storage would become necessary. Although basic steps involved for such correlations are understood and stable correlations have been obtained in a number of cases, historical data are scarce and not systematic. Further developments are required before such correlations can be used in a routine manner.

3) Correlations based on isotopes of radioactive fission products: Necessary isotopic data are obtained by means of gamma-spectrometric analysis prepared on fuel assemblies or on samples taken at the reprocessing plant input (for example Cs). These measurements are not performed on a routine basis. As in b) above, historical data are scarce and further development is necessary for reliable application.

3.3.1 Heavy Isotope Correlations

The correlations for isotopes of U and Pu are the most investigated ones. One of the first correlation between the Pu/U ratio and U-235 depletion D for the Yankee reactor was suggested by Schneider, Christensen and Granquist /6/.

$$\frac{\text{gm Pu/ton } U_f}{\text{D-235}} = 7038.6.r_o - 446.86.r_o^2 + 82757 \quad \ldots\ldots\ldots\ldots \quad (2)$$

where D is the relative depletion of U-235 = $1-r_f/r_o$; r is the U-235 enrichment in wt %; o denotes initial f denotes final. A typical example for the establishment of a heavy isotope correlation from a set of measurement data from different batches of irradiated fuels from Yankee core V, reprocessed in the NFS plant, is reproduced in Table I from /29/. In the framework of the Mol-III integral experiment /28/, isotopic correlations of the following types for the CANDU

type of fuel (natural uranium heavy water moderated) were found by Christensen, Schneider and Stewart.

Expected EUROCHEMIC Measured Values
for Similar Future Candu Fuels

$\frac{Pu/U}{U\text{-}235}$	$\frac{Pu/U}{\Delta U\text{-}236}$	$\frac{Pu/U}{Pu\text{-}240/Pu\text{-}239}$
7458±1.3 %	45581±2.4 %	10043±1.4 %

where the limits of uncertainties are at the 90 % confidence level.

In /10/ Christensen and Schneider have made a detailed analyses of the possibilities of heavy isotope correlations for four reactor systems namely, pressurized water (PWR), graphite moderated (GMR), boiling water (BWR) and heavy water (HWR). Altogether measured results from 353 input batches (166 PWR, 21 GMR, 154 BWR and 12 HWR) from these reactions and 38 sets of calculated data were analyzed to investigate the different possibilities of isotopic correlations. About 70 correlations involving concentrations, differences and ratios of the isotopes of uranium and plutonium were considered out of which seven were selected as useful correlations. Several criteria were applied in the selection of useful correlations:

a) Best linear indices
b) Application to more than one reactor system
c) Possibility of statistical identification of inconsistant data
d) Easily usable
e) Limited additonal information requirements over and above those which are routinely available.

Besides, the possible influence of reactor parameters like initial U-235 enrichment, cladding materials etc. were also taken into consideration. The seven correlations and their coefficients are reproduced in Tables II and III from /10/.

The most accurate isotopic measurements are normally those of U-235 and Pu-239 accounting for the use of these in the first four selections in Table II. These correlations are used to check U-235 and U-239 and are in turn used to check

Pu/U using correlations 5,6 and 7 of Table II. The data base cover the enrichment range of natural uranium to 4.9 wt. % of U-235 and the values apply to a wide range of exposures from zero to 30,000 MWd/MT.

For a given correlation, a mean correlation ratio vaue and a σ can be determined for a data set. The % σ is an indicator of how linear a correlation is. If the %σ is around 1-2 % the correlations is taken to be linear. Higher values of % σ may mean that the correlations are non-linear. In Fig. 2 (reproduced from /10/) values for the correlation ratios (100-Pu-239)/D-235 from PWR, BWR and HWR measured data, both dissolution batch data and post-irradiation data, are shown as a function of the initial U-235 enrichment values. The relation has been analyzed thoroughly in /10/ and will not be repeated here.

A number of other heavy isotope linear correlations has been established for a large number of reactor systems. Some of these correlations are illustrated in Figures 3-6.

Fig. 3: Pu/U ratio vs U-235 depletion for Latina reactor fuel (gas-graphite reactor) from Ref. /18/.

Fig. 4: U-236/U-238 ratio vs U-235/U-238 ratio for Chapel cross reactor fuel (gas-graphite reactor) from Ref. /30/.

Fig. 5: Pu/U ratio vs Pu-239, Pu-242/$(Pu-240)^2$ for the collective data. from Ref /31/.

Fig. 6: Pu/U ratio vs U-235 wt. % for the collective data. from Ref /31/.

3.3.2. Gaseous Fission Product Correlations

The correlations between U-235 depletion (D_5), total burnup (F_T) and the fission product ratio Kr-84/Kr-83 for a BWR system was shown to exist by Koch et al. in 1970 /11/. The correlation as reproduced in Fig. 7 from /11/ can be expressed by the following relation:

$$(U_o\text{-}235\text{-}U_f\text{-}235)/U_o\text{-}235 = 0.98\ (Kr\text{-}84/Kr\text{-}83)\text{-}1.68 \quad \ldots\ldots\ldots \quad (3)$$

Since 1970 irradiated fuels have been analyzed for both PWR (Obrigheim, Sena, Trino Vercellese) and BWR-Systems (Dodewaard, Garigliano, Gundremmingen, Kahl, Lingen). Fig. 8-10 illustrate the correlations obtained sofar for

gaseous fission products:

Fig. 8: Correlation Xe-132/Xe-131 ratio vs Pu-241 atoms/initial metal atoms (expressed as Pu-241 IMA) for different BWR fuels from Ref. /32/.

Fig. 9: Correlation Xe-132/131 vs fuel exposure FIMA % (F_T) (Fission per Initial Metal Atom) for PWR and BWR systems from Ref. /15/.

Fig.10: Correlation Xe-132/131 vs Pu-240 IMA from Ref. /15/.

Linear least square fit equations for some of the isotopic correlations for fission products as shown in Figs. 7-11 have been derived and are reproduced from /15/ in Table IV.

The accuracies of the equations arguments are expressed in averaged deviation (D %) and in single standard deviations (A %) according to the following equation:

$$D\ \% = 100\ \frac{\sum|(T-E/T)|}{n}\ ;\ A\ \% = 100\left(\frac{\sum(T-E)^2/T^2}{n-2}\right)^{1/2} \qquad \ldots\ldots \quad (4)$$

where

T represents the data calculated from the correlation equation
E represents the experimental data and
n is the number of data points.

The correlations were obtained by using the measurement data both from the post irradiation experiments on irradiated fuel pellets and from fuel elements. The correlations which have been derived from the post irradiation experiments are more sensitive to changes in the neutron energy spectrum than those obtained for fuel elements. As mentioned earlier, the correlations based on fission products have not been based on as broad and thorough a data base as in the case for heavy isotopic correlations. However, the inherent possibilities of the existence of these correlations have been amply demonstrated. The data base has to be broadened and stabilized.

The main advantage of using the fission gas correlations lies in the fact that they are unique to fission process i.e., they can enter the fuel cycle

at the point of reprocessing only through the fission process at the reactor of the previous stage (no recycling possibility as in the case of heavy isotopes). They are less intrusive as the samples for measurements can be obtained from the dissolver of gas stream of a reprocessing plant. At present the samples are not required for normal operation.

3.3.3 Correlations Based on Isotopes of Radioactive Fission Products

The only correlations which have been established sofar on a reasonable data base for the radioactive fission products are those for Cs-137 or Cs-134/Cs-137 ratios and Pu/U ratios or burnups. These correlations are illustrated in Figs. 11-13:

Fig. 11: Correlation Cs-134/137 vs Pu-240 IMA and FIMA % to BWR fuel from Ref. /15/.

Fig. 12: Burnup vs Cs-137 activity for Trino Vercellese reactor fuel from Ref. /17/.

Fig. 13: Cs-134/Cs-137 activity ratio vs Pu/U ratio for Trino Vercellese reactor fuel (destructive determination) from Ref. /18/.

Beets and Dragnev /33/, Beets et al /34,35/, Dragnev and Burgess /36/ Dragnev et al /37/ and Koch et al /15/ have reported on the advantages and possible use of such correlations. The main advantage, as indicated earlier, is the possibility of the use of non-destructive measurement techniques for complete irradiated fuel elements in the wet storages of a reactor station or a reprocessing plant. The possible use of such correlations will be discussed later.

3.4 Theoretical Investigations in Connection with Isotopic Correlations

3.4.1 Influence of Reactor Parameters

As mentioned earlier some detailed theoretical investigations on the influence of various reactor parameters on stable isotope- and fission product correlations for PWR- and BWR-systems have been carried out. As typical examples the results of some of these investigations which have been carried out at the EURATOM Research Center Ispra are reproduced in Figs. 15-19:

Fig. 14: Calculated Pu/U/U-235 Depletion vs initial fuel enrichments with different reactivity control methods and moderation ratios for different PWR fuels from Ref. /23/.

Fig. 15: Calculated Pu/U ratio vs U-235 depletion for different BWR fuels and different void contents of the moderator: comparison with experimental data from Ref. /23/.

Fig. 16: Calculated heavy atoms burnt (F_t %) vs Kr-84/Kr-83 for different fuel enrichments and moderator to fuel volume ratios from Ref. /25/.

Fig. 17: Calculated heavy atoms burnt (F_t %) vs Xe-132/Xe-131 for different fuel enrichments and moderated fuel volume ratios from Ref./25/.

Fig. 18: Calculated heavy atoms burnt (F_t %) vs Nd 148/145 for different fuel enrichments and moderated fuel volume ratios from Ref. /25/.

It is to be noted that the heavy isotope correlations and the correlation between F_t % and Nd-148/Nd-145 show a fairly good linearity whereas, the correlations for the Kr and Xe isotopic ratios are expected to be linear for a given range of these ratios. All the correlations chosen are expected to be specific for a given initial fuel enrichment and moderator fuel ratios. This means that they are characteristic of a given reactor system. Some of the correlations however, may overlap for different reactor systems in a given range of initial fuel enrichments as shown in Fig. 2 /Ref. 10/.

3.4.2 Statistical Evaluations of Measured Data Used for Correlation

In isotopic correlation studies for the area reactor-reprocessing in a nuclear fuel cycle, the first step is to establish linear relations between depletion- and build-up parameters for selected groups of isotopes based on a set of measured data. Since data on depletion and buildup of isotopes which may be of relevance to correlations, are subject to systematic and random errors related to operating conditions in reactors and reprocessing plants, to uncertainties in the input and calculations of burnup data or to measurement errors in the results of destructive and non-destructive analyses, there is often a large scatter in both the X- and Y-values which may be used as input

correlation diagram /26/. In establishing a linear functional relationship for a set of X, Y points in such diagrams, the scatter has to be considered for both the coordinates. One of the branches of theoretical considerations deal with statistical and modelling problems associated with the establishment of linear correlations from a wide range of data sets. As an example the work at the JRC, Ispra and RCN, Petten in this connection may be mentioned /26/.

In the framework of the ESARDA joint research program /26,27/ a computer program CORRELATIO has been developed which determines on the basis of the least square principle the parameters, and their standard deviations for the best fitting straight line from a given set of correlation data. In the case that both the variable are subject to error, the straight line is the orthogonal regression line, and the method is then called the minimum distance method.

The program is made to facilitate the statistical analysis of the isotope correlation data which are collected in the ESARDA data bank at the EURATOM JRC, Ispra.

3.5 Application of Isotpic Correlations

In the present paper it is proposed to emphasize the application of isotopic correlation techniques for the accounting and control of nuclear materials in a nuclear fuel cycle. In such a cycle nuclear material passes through different nuclear facilities in which the material is processed, used, stored or transported as well as changes in its isotopic composition or transmutations of some of the elements present take place. The material and information flows in a typical commercial nuclear fuel cycle are shown in Figs.19a and 19b.Some important characteristics in connection with the operation and measurement of nuclear materials in such a fuel cycle may be mentioned which are relevant to isotopic correlations.

a) Isotopic shifts and transmutations of U or Pu take place in enrichment plants and reactors respectively as an inherent characteristic of the process - and nowhere else in the cycle.

b) The flow of nuclear material from the enrichment to the reprocessing facility is unidirectional under normal conditions in a fuel cycle. The informations on isotopic characteristics of nuclear materials, after they have been generated at a given point, remain unchanged over long stretches or can be linked with each other at different points of the cycle.

c) For the area reactor-reprocessing in a fuel cycle all the data relevant to isotopic relations become available through direct measurements at the input of a reprocessing plant.

These characteristics can be used both by the operators of nuclear facilities and safeguards organisations to improve and streamline their respective procedures for the accountancy and control fo nuclear materials.

The possibility of using isotopic correlations for the optimization of safeguards efforts in a nuclear fuel cycle was investigated by Gupta et al /40/. It was shown there that some reduction in safeguards efforts could be expected if the relevant informations on isotopic correlations could be judicially put to use.

With the overall use in a nuclear fuel cycle in view, the more important applications of isotopic correlation techniques can be envisaged in four broad areas:

1. Verification and generation of reliable information on the Pu amount in the input tank of a reprocessing plant.

2. Checking of consistancy of different measured data which may be of relevance to a system of accountancy and control of nuclear materials.

3. Improvement of input and calculation data for burnup codes for different reactor systems.

4. Improvement of fuel management at different points of a nuclear fuel cycle.

It is expected that the application of ICT can be extended to other areas also in the course of time.

3.5.1 Verification and Generation of Reliable Information on the Pu Amount in the Input Tank of a Reprocessing Plant

It is to be noted that the maximum use of isotopic correlation techniques is expected to be in and around the input accountability tank of a reprocessing plant. The relevant steps for isotopic correlations around the dissolver of a typical reprocessing plant (which operates on a batchwise basis)are reproduced in Fig. 20 from /9/. In most of the cases the irradiated fuel elements are chopped before they enter the dissolver vessel in which the fuel is dissolved in nitric acid which may be recycled from a previous operation campaign. If insufficiently purified, the recycled acid may contain U and Pu with different Pu/U ratios than those to be dissolved. Corrections have to be made for recycled material (if present in significant quantities) if correlation techniques are to be used. A part of the U and Pu in the irradiated fuel gets lost as wastes (mostly in the range of 0.1-0.2 %) on account of incomplete dissolutions. They have to be taken into consideration for the determination of the total amount of Pu present in the irradiated fuel since the measurement of Pu takes place at the accountability tank after the losses have taken place.

Basically, two different measurement systems are in use in present reprocessing plants for the determination of the total amount of Pu going into the process. They are:

I) Volumetric method
II) Pu/U-ratio method

These two methods have been compared in great detail and the advantages of the Pu/U-method discussed in /22,39,42/. The Pu/U method has been analyzed in /43/. The volumetric method which is still used extensively in a number of reprocessing plants is highly plant specific and does not make use of any of the existing correlations in a cycle. The method requires a large amount of effort both for the generation of the required data and for its adequate verification. The Pu/U method is also in use in a number of reprocessing facilities (e.g. Windscale, UK). The importance of this method in connection with the ICT cannot be overemphasized and it may be worthwhile to discuss the different elements of this method to bring out its salient features and its relation to the isotopic correlation technique.

In the Pu/U-ratio method the absolute amount of Pu in kg in a batch of accountability solution is given by the following relation:

$$Pu = U_o \cdot \frac{U_f}{U_o} \cdot \left(\frac{Pu}{U_f}\right) \quad \ldots\ldots \text{ /kg/ } \ldots\ldots \qquad (5)$$

where U represents the total amount of Uranium in kg and the indices o and f denote the initial and post irradiation stages of the fuel elements dissolved in the accountability tank.

Three values are required in eqn. 5 to determine the absolute amount of Pu.

a) The initial amount of total Uranium in the fuel elements dissolved (measured accurately at the fabrication plant)

b) $\frac{U_f}{U_o}$ ratio. As is clearly seen, this ratio is a correction term for the loss of Uranium on account of burnup in the reactor in which the fuel elements under question were irradiated.

Depending on the burnup level of the fuel (up to about 30,000 MWd/t) this ratio has the range:

$$0.96 \leq \frac{U_f}{U_o} \leq 1.$$

The actual value of this correction term can be obtained in a number of ways.

A. For example it can be based on the burnup data of the reactor operator. The burnup values are required to be known only roughly, i.e. with low accuracy. A 10 % uncertainty in the burnup value contributes only 0.2 % uncertainty to the correction term /22/. A typical interpolation carpet diagram was prepared on the basis of /6/ and was presented in /39/. The diagram is reproduced in Fig. 21. For a given burnup value and a measured Pu/U-ratio, the value of the burnup correction term U_f/U_o can be obtained from the diagram.

In this method no additional measurements than those already required, are necessary. A special correlation between Uranium depletion and burnup in the reactor is used to obtain the value for this ratio.

B. The U_f/U_o ratio can also be obtained through the masspectrometric measurement of a fission product/U_f ratio for example of the type Nd-148/U_f. The measurement can be done from the same sample from which Pu/U ratio is to be determined. The ratio of Nd-148/U_f is related to the U_f/U_o through the following eqn. /39/.

$$U_o/U_f = 1 + \frac{Pu}{U_f} + \frac{Nd\text{-}148}{U_f} \cdot \frac{C\text{-}148}{Y\ 148} \qquad \ldots\ldots \quad (6)$$

where Y 148 is the average fission yield for Nd-148. Since the fission yield of Nd-148 is almost the same for all important heavy isotopes, it can be expressed fairly well by the relation:

$$Y\ 148 = 1.715 \pm 0.024\ \% \qquad \ldots\ldots \quad (6a)$$

According to /44/ the term C 148 (which is a correction term to account for the absorption losses of Nd-148 produced)can be expressed for a given burnup BU in MWd/kg by the following:

$$C\ 148 = 1 + 5.019 \cdot 10^{-4} \cdot (BU) + 2.275 \cdot 10^{-6} \cdot (BU)^2 \quad \ldots\ldots \quad (6b)$$

For a BU of 30 MWd/kg U a correction of only 1.7 % is required /39/. This term can therefore even be neglected.

The Nd-148/U_f ratio enables the determination of the absolute amount of Pu (by the Pu/U ratio method) on the basis of values,all of which are obtained by direct measurement. This method is routinely used at Windscale, UK.

The above discussion on the Pu/U method is meant to illustrate a number of important features of this method in connection with the use of isotopic correlation techniques for accountancy and control of nuclear materials in a fuel cycle.

i) The three terms on the right hand side of eqn. 5 correlate information generated at three different points of the fuel cycle, obtained by three different and independent methods. The values of the amounts of Plutoninium thus obtained, have therefore, a high credibility.

ii) The only absolute amount required for this method is the value of U_o, which is measured in an inactive environment with fairly high accuracy /46/. The rest of the values are ratios. The generation of values on isotopic ratios is normally less combersome and more transparent i.e. more easily verifiable than the corresponding absolute values required in the volumetric method.

iii) The linear correlations Pu/U-U-235 depletion, Pu/U-initial U-235 enrichment or Pu/U- some fission product ratios, when properly developed, may be used somewhat like a calibration curve. In principle the values of Pu/U-ratio could be taken for a given U-235 depletion, U-235 concentration or fission product ratios from such correlation curves and inserted into eqn. 5 to determine the absolute amount of Pu. No actual measurement of Pu/U ratio would then be required. As such they could then become an important tool for independent verification of data both for facility operators and safeguards organizations.

It is seen from the above discussions that four possible methods for the determination of the absolute amount of Pu in the accountability tank of a reprocessing plant could be considered in principle:

I. Volume-concentration, II Pu/U-ratio, III. heavy isotope correlations and IV. fission gas correlations. The different steps involved in these four methods are reproduced in Table V from /9/.

The precision of the results obtained by the methods I and II are higher ($\sim$ 1 %) than those obtained by III and IV. The analytical efforts are the highest for I decreasing then for II, III and IV in the same order. Verification efforts which are directly related to the analytical efforts and to the credibility of the data, are the highest for I decreasing then for the rest of the methods in the same order. For the methods II and III correction for recycle

Pu has to be made. For IV the correlation is not effected by recycle of heavy elements. In I the actual amount of Pu entering the process is determined. For the methods II ;III and IV corrections for wastes have to be made to determine the actual amount of Pu entering the process.

It is to be noted that in a commercial reprocessing plant either the method I or II must be available for routine plant operations. At the present state of the art,the methods III and IV are more useful for verification purposes.

The application of the methods II-IV in a reprocessing plant is possible under a number of boundary conditions. For example the identity of the fuel elements dissolved must be known. An integer number of fuel elements should be dissolved per batch. Fuel elements from the same reactor with the same initial U-235 enrichment should preferably be dissolved in a batch. Measurement methods for determining the Pu in recycle acids and in waste streams should be available.

3.5.2 Consistency Checking of Measured Data

Well established isotopic correlations can be used to check the consistency of measurement data at the input of a reprocessing plant. A typical example of consistency checking is given in Figs. 22 and 23 from /32/. A linear correlation between the ratios 240 IMA (Initial Metal Atoms) vs Xe 132/131 was established for Trino Vercellese reactor (PWR) based on historical data (Fig. 22) with an uncertainty band of $\pm$ 1.4 %. The measured values of the same correlation for Obrigheim and Sena Fuel (both PWR and measured from the same type of accountability tanks solutions by the same type of measurement methods) showed a large deviation from the correlation diagram already established. It was found that accountability tank solution contained recycled Plutonium from another campaign. After correction for the recycle Pu, the measured values fell within the error band of the original correlation. Simular effects were found for Obrigheim fuel for the correlation 240 IMA vs Pu-241/242 (Fig. 23). Consistency checking for different masspectrometric values of U-235 with the help of correlations as shown in Fig. 6 /Ref. 31/ is another example of this type of application of the ICT.

Such applications are mainly meant for the control of the information used for accountancy of nuclear materials.

3.5.3 Improvement of Input Data for Burnup Calculations in Reactors

The correlation data obtained from measurements at a chemical reprocessing plant can be useful for burnup calculations also. They are particularly applicable for testing the reactor operator's burnup models because they represent a volumetric average of the burnup in the reactor core /47/.

Comparison of calculations of reactor core isotopic inventory against measured inventory have shown several typical biases /47/. Mention may be made of the bias between the measured and calculated Pu/U/D-235 values, between the measured and the calculated values of U-235 final concentrations as well as biases between the measured and calculated values of U and Pu isotopes. Besides, isotopic correlations data can also reveal errors in input values to burnup models /47/. Some typical examples of biases between the measured and calculated values of U-235 concentrations are reproduced from /48/ in Table VI.

Table VI. Calculated and Measured Values of U-235 Concentrations in Irradiated Fuels /Ref. 48/

Campaign No.	Calculated Value U-235 /gms/	Measured Value U-235 /gms/	C-M % D
4	488704	503435	-3.0
5	1235452	1248670	-1.0
13	509320	529890	-3.99
15	140960	151650	-7.37
16	268760	277543	-3.2
17	244118	252143	-3.28

Bias -3.66 %

One of the causes of such a bias could be the choice of a wrong α-value (ratio of capture to fission cross sections) for U-235. In fact the ratio of

U-236 buildup to U-235 depletion for a specific reactor fuel is related to the α of the U-235. Isotopic correlations of the type shown in Fig. 4 could indicate the amount of correction to be applied to the α-value to eliminate the bias obtained by the comparison of the measured and the calculated data as shown in Table VI. It is however, important to note that such a bias need not necessarily be caused by a wrong α-value alone.

Similar uses of isotopic correlations in connection with burnup calculations for different reactor systems have been discussed in detail in /15,21,23,47/. The applications of ICT in this particular area relate more to the improvement and control of burnup calculations for reactors which may lead to better utilization of fuel. Thus they influence implicitly the accountancy and control of nuclear material.

3.5.4 Improvement in Fuel Management

The isotopic correlations between Cs 137 activities or Cs 134/137 activity ratios and burnup or Pu/U ratios may be of use for nondestructive determination of burnup and Pu content in irradiated fuel subassemblies /33,37/. The data obtained may be useful both for safeguards and plant operation purposes. With the present state of the art, the assay can only be relative to a reference fuel assembly, the isotopic composition and burnup level of which must be accurately determined by means of destructive analysis at the reprocessing plant.

At a reactor storage such correlations can be used to identify quickly fuel elements (with different cooling times, burnup levels etc.) /38/. In principle, they can also be used for verification of reprocessing plant data or burnup calculations /9,37/.

At a reprocessing plant these correlations could be used for optimal batching of the irradiated fuel elements (before dissolution) to obtain high isotopic homogeneity of final Pu produced or to ensure high plant throughput with respect to criticality problems.

The application of ICT in this particular area is mainly for ensuring an optimum management of fuel elements (besides its use as a mean of corroborating data from other sources). They influence the accountancy and control

procedures for nuclear materials only in an indirect manner.

4. Concluding Remarks

The present review is mainly meant to trace the development path of the isotopic correlation techniques and indicate in a broad manner the possible areas of its applications in the accounting and control of nuclear materials. Although considerable amount of effort has gone into the development of the different isotopic correlations, at present the ICT has to be considered to be in its initial phase of development. Further large scale activities at both international and national levels are required on a continuous basis to bring ICT at a level at which both the operators of nuclear facilities and safeguards organizations could use them as a reliable method for routine purposes.

The most important conclusion of this paper is that the ICT should be considered for use in a whole nuclear fuel cycle and not for use on a piecewise individual facility basis. Only then can the fundamental characteristics of this method be used to the fullest advantage.

The ICT have enormous potential for the large scale nuclear fuel cycles of the 1980's. Since they utilize inherent process characteristics of the cycle, the additional efforts in establishing the correlations are small, the steps involved in verifying the correlations are straightforward and transparent. Along with containment and surveillance measures the ICT may form the basis of a realistic system in the 80's for the verification of the accounting and control of nuclear materials in a fuel cycle.

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Raw Data

Input Batch #	Total Pu g/tu	Uranium Isotopics ^{235}U W/O	^{236}U W/O	Plutonium Isotopics ^{239}Pu W/O	^{240}Pu W/O	^{241}Pu W/O	^{242}Pu W/O
1	4968	3.176	0.2209	85.05	10.29	4.01	0.36
20	7104	2.796	0.2997	79.35	12.53	6.65	0.78
10	7930	2.675	0.3253	77.57	13.47	7.37	1.04
22	8863	2.471	0.3589	75.14	14.54	8.29	1.34
18	9794	2.254	0.3993	72.69	15.46	9.18	1.75
16	11119	2.046	0.4454	69.31	16.34	10.85	2.50

Pu/U - Isotopic Correlations

Input Batch #	Total Pu g/tu	$\frac{Pu/U}{^{235}D}$	% Dev.	$\frac{Pu/U}{\Delta^{236}U}$	% Dev.	$\frac{Pu/U + K}{^{240}Pu/^{239}Pu}$	% Dev.
1	4968	5371	-0.52	27463	-0.01	54078	-0.32
20	7104	5448	+0.91	27334	-0.48	55026	+1.43
10	7930	5417	+0.33	27516	+0.19	54820	+1.05
22	8863	5441	+0.78	27792	+1.19	54054	-0.36
18	9794	5306	-1.72	27258	-0.75	53565	-1.27
16	11119	5413	+0.26	27427	-0.14	53970	-0.52
		$\bar{X}$ = 5399		$\bar{X}$ = 27465		$\bar{X}$ = 54252	
		σ = 0.98%		σ = 0.67%		σ = 1.03%	

Where: ^{235}D = wt. % ^{235}U initial - wt. % ^{235}U final

$\Delta^{236}U$ = wt. % ^{236}U final - wt. % ^{236}U initial.

TABLE I. EXAMPLE OF PU/U AND ISOTOPIC CONSISTENCY
YANKEE ROWE CORE V DATA
(from ref. 29)

TABLE II. CORRELATIONS IDENTIFIED FOR USE

CORRELATION		CORRELATION RATIO
Y VARIABLE	X VARIABLE	
1. $100 - {}^{239}Pu$	${}^{235}D$	$(100 - {}^{239}Pu)/{}^{235}D$
2. ${}^{239}Pu \times (100 - {}^{239}Pu)$	$\Delta\left[{}^{235}U \times ({}^{239}Pu)^2\right]$	${}^{239}Pu \times (100 - {}^{239}Pu)/\Delta\left[{}^{235}U \times ({}^{239}Pu)^2\right]$
3. $\Delta({}^{239}Pu)^2$	$\Delta({}^{235}U \times {}^{239}Pu)$	$\Delta({}^{239}Pu)^2/\Delta({}^{235}U \times {}^{239}Pu)$
4. $({}^{239}Pu)^2 \times (100 - {}^{239}Pu)$	${}^{235}U\ (100 - {}^{239}Pu)$	$({}^{239}Pu)^2/{}^{235}U$
5. Pu/U	${}^{235}D$	$(Pu/U)/{}^{235}D$
6. Pu/U	$100 - {}^{239}Pu$	$(Pu/U)/(100 - {}^{239}Pu)$
7. Pu/U	$\Delta\left[({}^{235}U)^2/{}^{239}Pu\right]$	$(Pu/U)/\Delta\left[({}^{235}U)^2/{}^{239}Pu\right]$

THE Δ, D AND $100 - {}^{239}PU$ ARE USED TO INDICATE THE DIFFERENCES BETWEEN FINAL AND INITIAL VALUES (from ref. 10)

TABLE III. LINEAR INDICES AND MEAN RATIO VALUES FOR CORRELATIONS

CORRELATION NO. (TABLE 1)	LINEAR INDICES			MEAN RATIO VALUES*			
	NPD	DRESDEN 1	YANKEE ROWE	NPD	DRESDEN 1	YANKEE ROWE	
	%σ	% DIFF.	%σ	NAT.	1.474	3.404	4.935
1	1.05	0.72	2.93	64.5	42.5	19.5	13.2
2	0.53	0.95	1.21	35.5×10^{-2}	19.2×10^{-2}	8.50×10^{-2}	5.81×10^{-2}
3	1.01	1.03	1.05	94.0	53.8	24.0	16.5
4	3.97	1.44	1.30	18.7×10^{3}	6.36×10^{3}	2.69×10^{3}	1.85×10^{3}
5	1.25	2.45	0.91	7.35×10^{3}	6.23×10^{3}	5.78×10^{3}	4.99×10^{3}
6	1.35	1.63	2.83	114	147	297	379
7	2.38	0.17	0.87	79.8×10^{4}	32.3×10^{4}	13.1×10^{4}	7.88×10^{4}

* PU/U IS IN UNITS OF GRAMS PU/TONNE U. THE ABSOLUTE VALUES FOR CORRELATIONS 1,2,5 AND 7 ARE SHOWN. (from ref. 10)

NPD: HWR; DRESDEN 1: BWR, YANKEE: PWR

No.	LINEAR CORRELATION EQUATION	D%	A%	FIG.
1.	F_T = -4.58 + 3.07.Xe-132/1	1.6	9.9	9
2.	Pu-240 IMA = -1.3.E-4 + 0.017.Cs-134/7	2.0	4.4	10
3.	Pu-240 IMA = -3.73 E-3 + 2.48E-3.Xe-132/1	1.4	3.4	11
4.	Pu-240 IMA = -3.85 E-3 + 2.47E-3.Xe-132/1	3.5	6.1	11

TABLE IV. LINEAR EQUATION OF ISOTOPE CORRELATIONS FOR FISSION PRODUCTS. D % IS THE AVERAGED DEVIATION BETWEEN EXPERIMENTAL DATA AND THE EQUATIONS ARGUMENT. A % IS THE CORRESPONDING STANDARD DEVIATION.

OPERATIONS AND MEASUREMENTS → / METHODS ↓	VOLUME MEASUREMENT	DENSITY MEASUREMENT	ALIQUOTATION OF SAMPLE	ISOTOPE DILUTION ANALYSIS	MASS SPECTROMETER MEASUREMENT	ANALYSIS OF RECYCLED MATERIAL	INITIAL WEIGHT OF FRESH FUEL	ISOTOPE CORRELATION BY HISTORICAL DATA
I VOLUME--CONCENTRATION METHOD	*	*	*	*	*	*		
II PU/U RATIO METHOD				*	*	*	*	
III HEAVY ISOTOPES CORRELATION					*	*	*	*
IV FISSION GAS ISOTOPES CORRELATION					*		*	*

TABLE V. STEPS INVOLVED IN THE DETERMINATION OF PLUTONIUM IN INPUT ACCOUNTABILITY TANK FOR FOUR DIFFERENT METHODS (from ref. 9)

Table VI. Calculated and Measured Values of U-235 Concentrations in Irradiated Fuels /Ref. 48/

Campaign No.	Calculated Value U-235 /gms/	Measured Value U-235 /gms/	C-M % D
4	488704	503435	-3.0
5	1235452	1248670	-1.0
13	509320	529890	-3.99
15	140960	151650	-7.37
16	268760	277543	-3.2
17	244118	252143	-3.28

Bias -3.66 %
==========

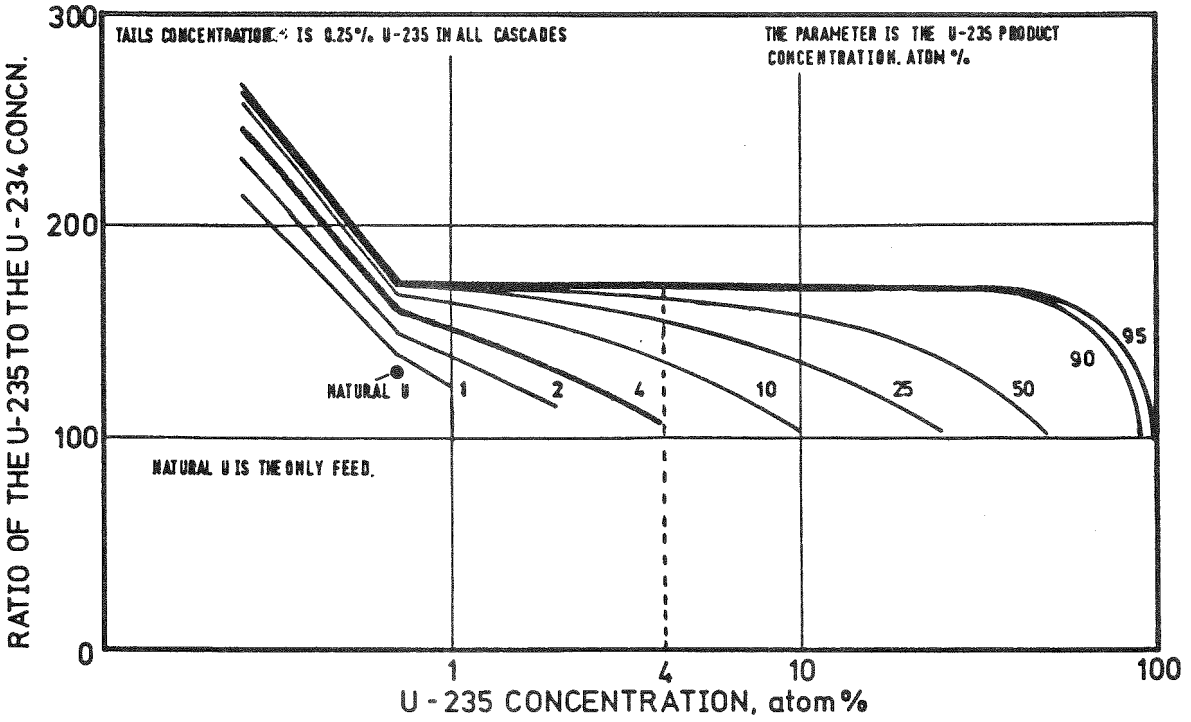
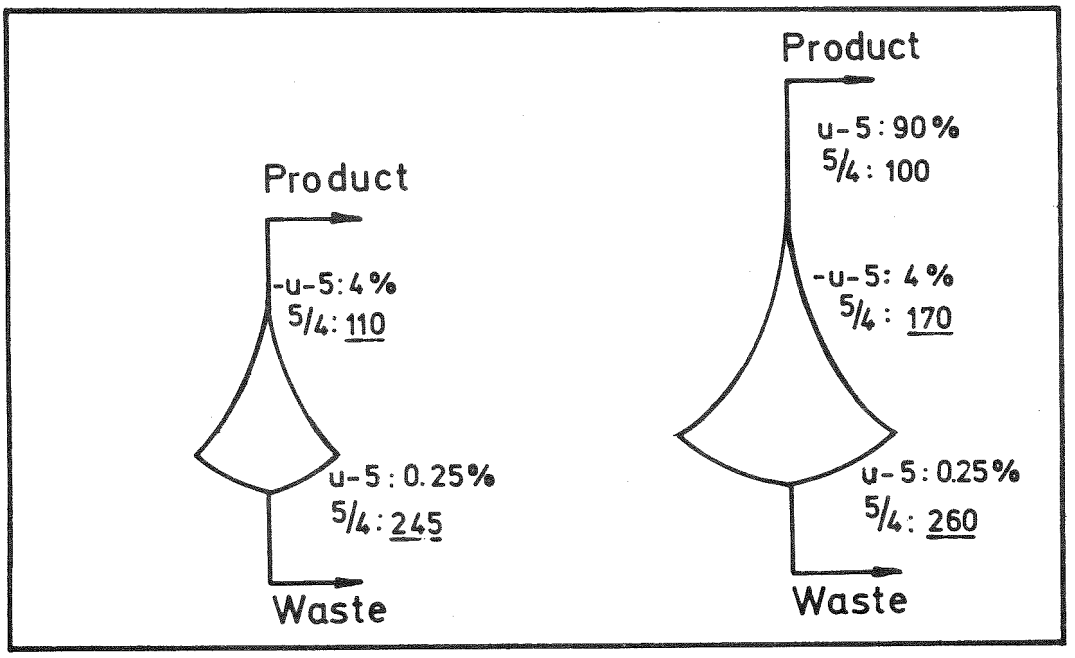


Figure 1 THE U-234 CONCENTRATION IN IDEAL CASCADES WITH DIFFERENT DESIGN PRODUCT CONCENTRATIONS (Ref 1)



CONCN. RATIOS U-235/U-234 FOR DIFFERENT IDEAL CASCADES (DIFFN. PLANT)

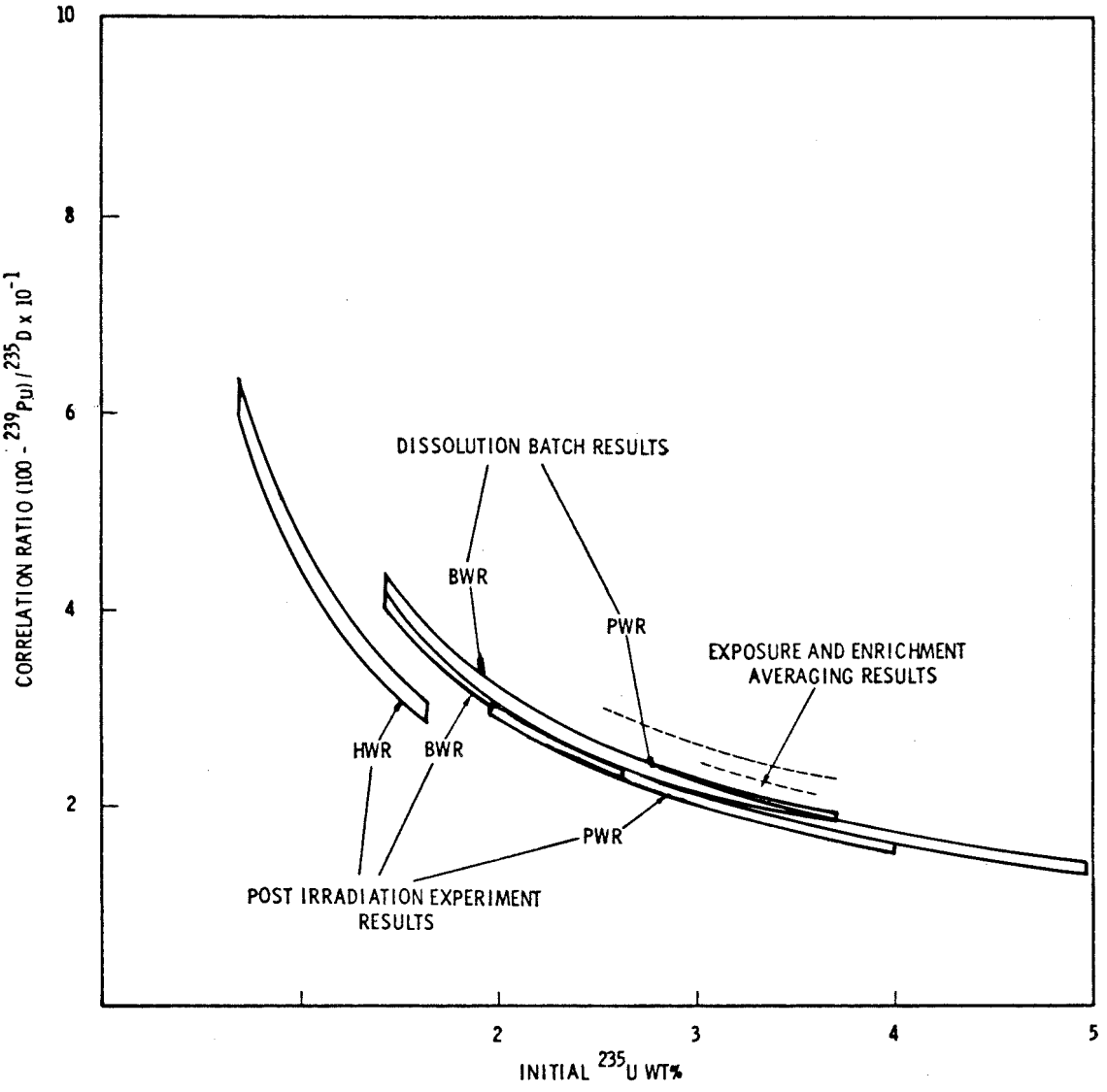


FIGURE 2. Composite Figure Showing Correlation Ratio Results Summary for ICT Data Base (Ref. 10)

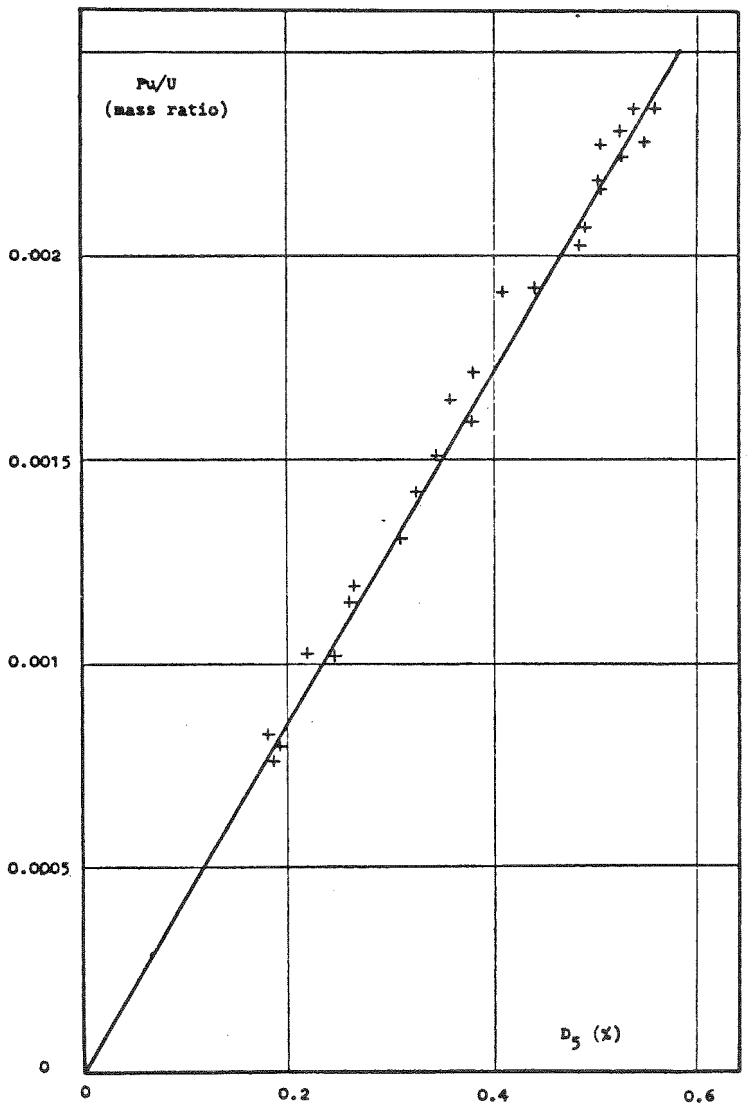


FIG. 3. Pu/U ratio versus ^{235}U depletion for Latina reactor fuel (from ref.18)

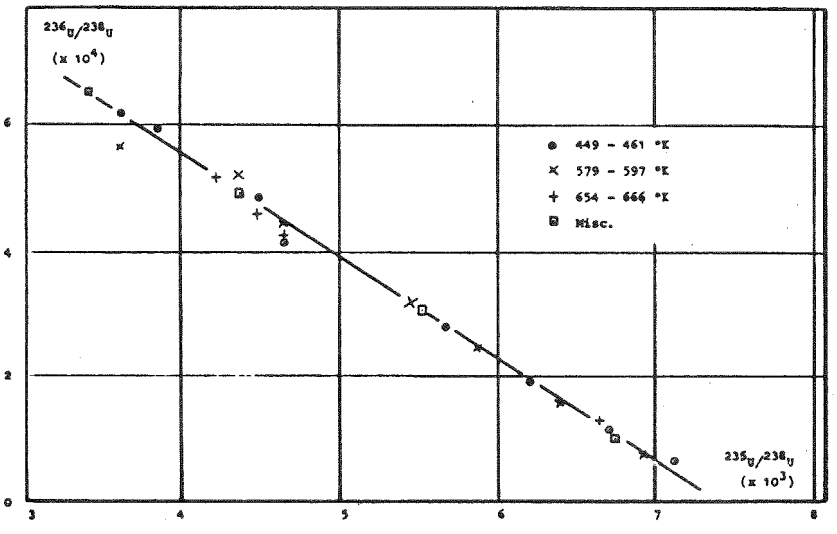


FIG. 4. $^{236}U/^{238}U$ ratio versus $^{235}U/^{238}U$ ratio for Chapelcross reactor fuel (from ref.30)

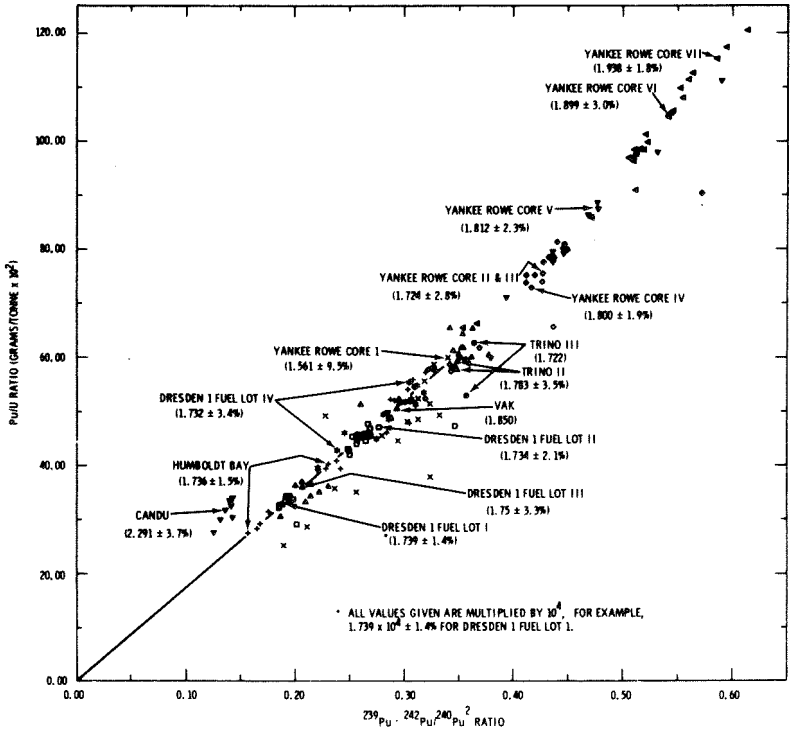


Fig. 5. Pu/U ratio vs $^{239}Pu.^{242}Pu/^{240}Pu^2$ for the collective data (from ref. 31)

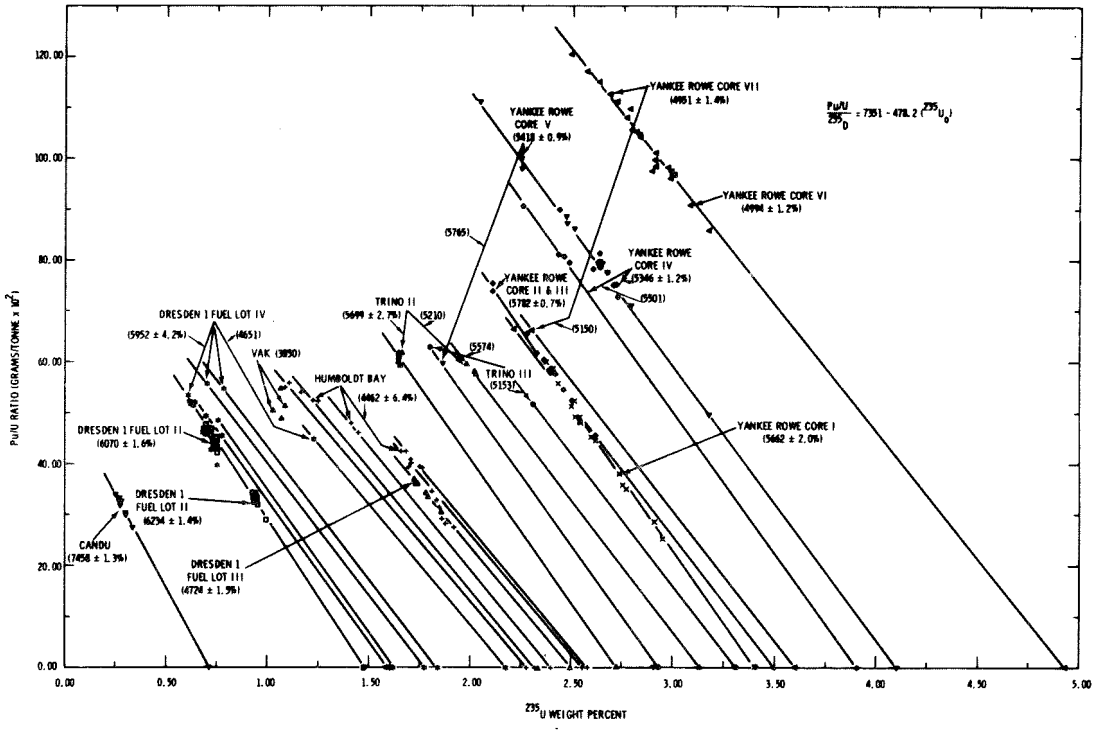


Fig. 6. Pu/U ratio vs ^{235}U weight percent for the collective data (from ref. 31)

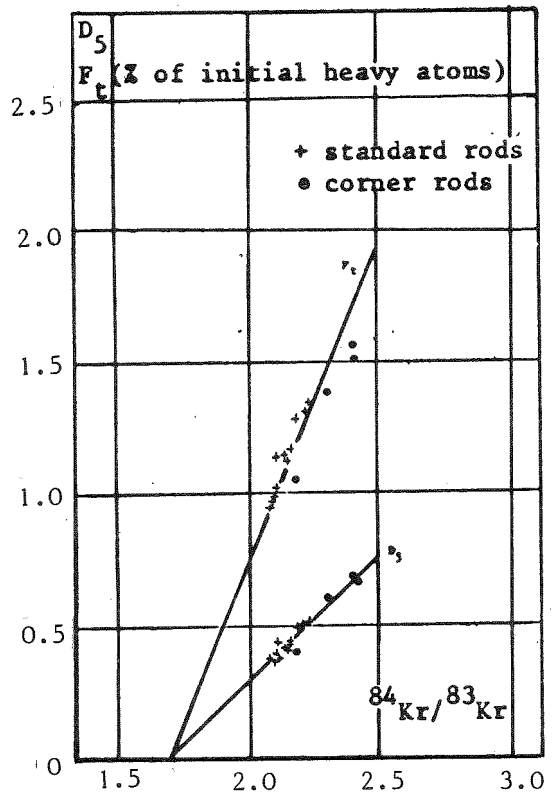


Fig. 7. Fractional ^{235}U depletion (D_5) and heavy atoms burnt (F_t), versus $^{84}Kr/^{83}Kr$ ratio, for Garigliano reactor fuel (from ref. 11)

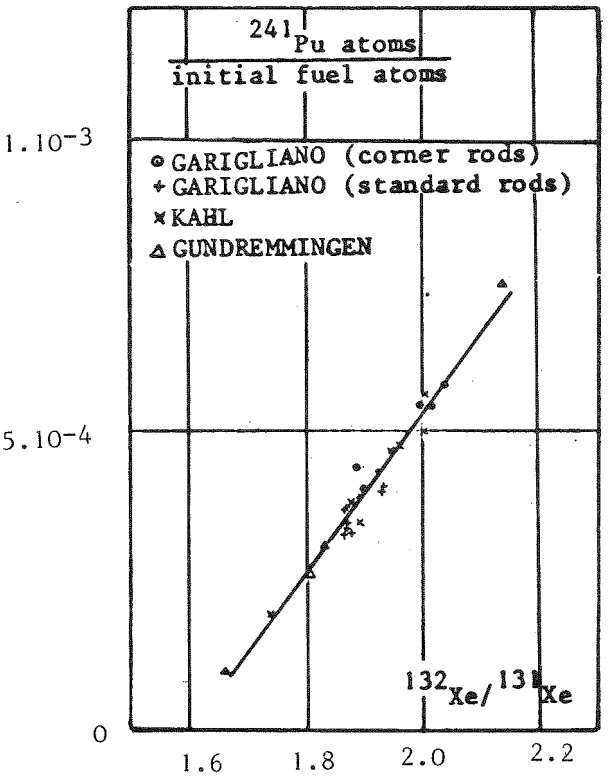


Fig. 8. Correlation Xe-132/Xe-131 ratio vs. Pu-241 atoms/initial metal atoms (expressed as Pu-241 IMA) for different BWR fuels. (from ref. 32)

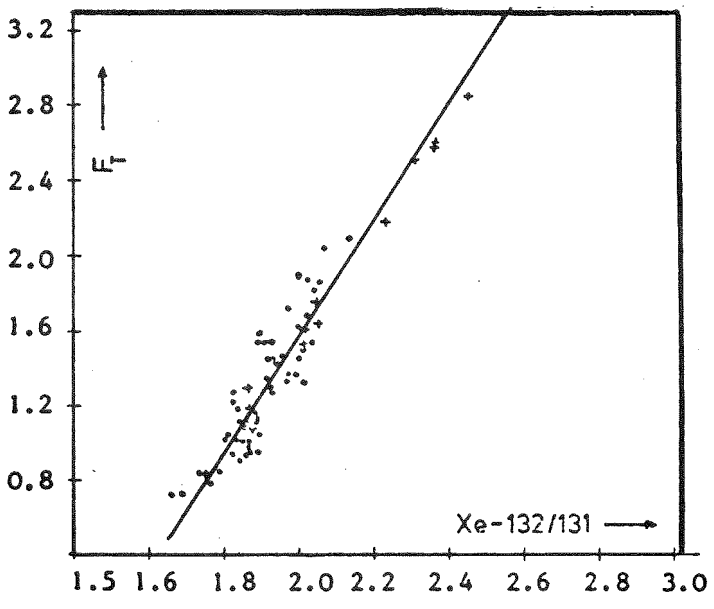


Fig. 9. Correlation Xe-132/131 vs fuel exposure FIMA % (F_T) (Fission per Initial Metal Atom) for PWR and BWR systems (from ref. 15)

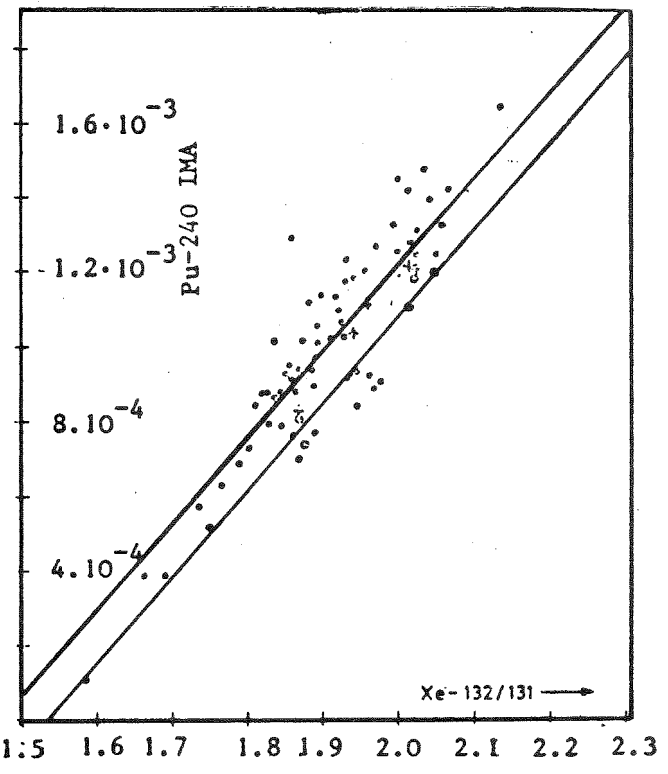


Fig. 10. Correlation Xe-132/131 vs Pu-240 IMA (from ref. 15)

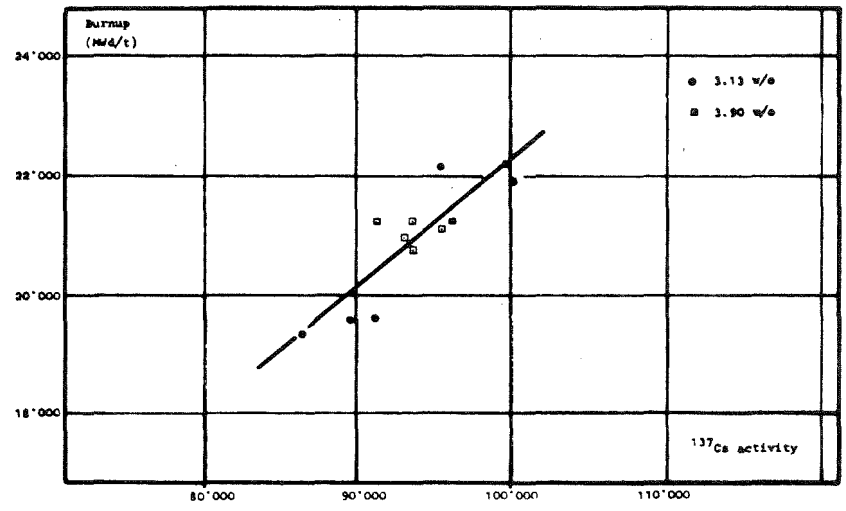


Fig. 12. Burnup vs Cs-137 activity for Trino Vercellese reactor fuel. (from ref. 17)

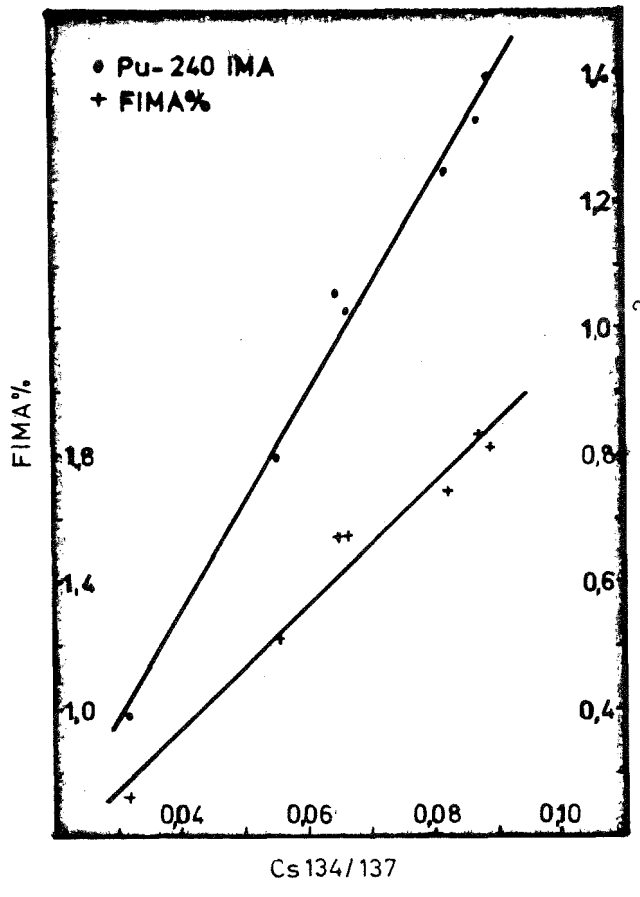


Fig. 11. Correlation Cs-134/137 vs Pu-240 IMA and FIMA % to BWR fuel (from ref. 15)

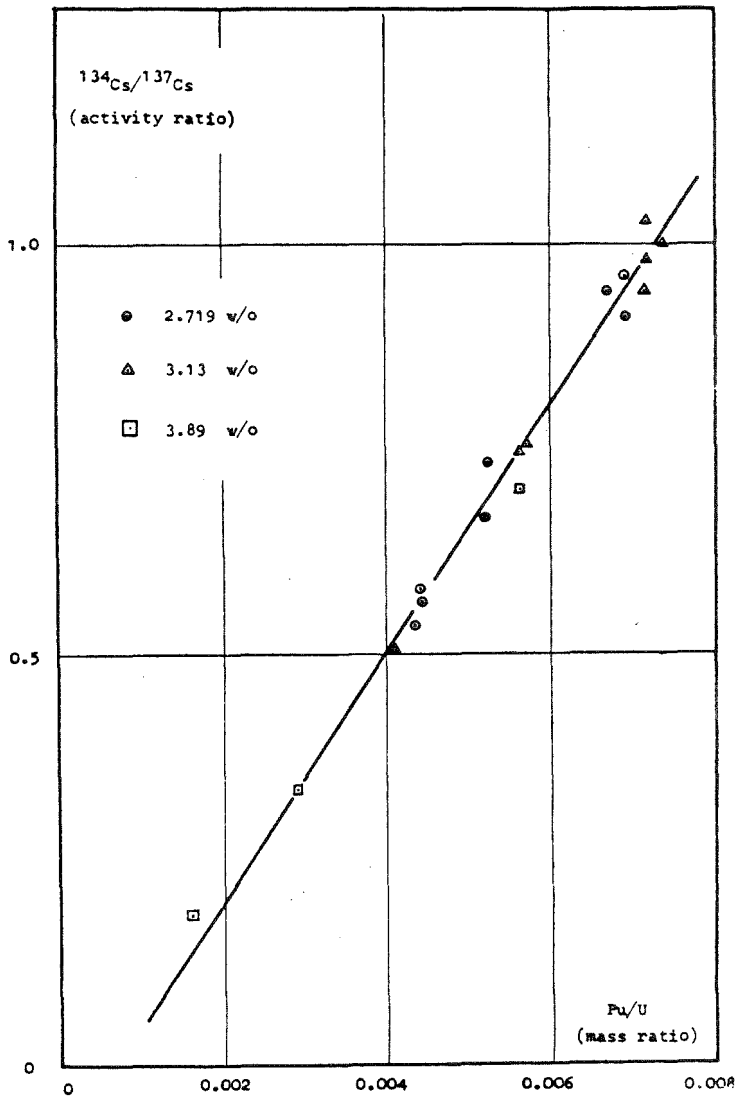


Fig. 13. Cs-134/Cs-137 activity ratio vs Pu/U ratio for Trino Vercellese reactor fuel (destructive determination). (from ref. 18)

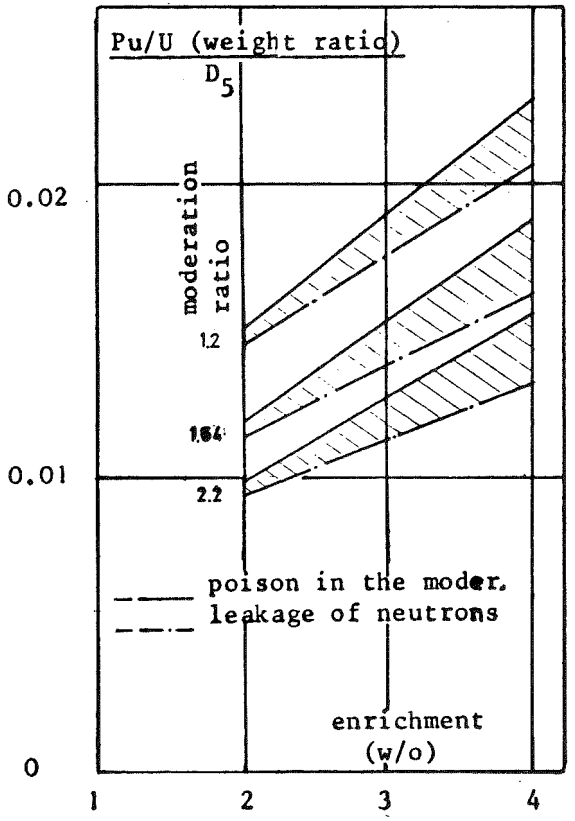


Fig. 14. Calculated Pu/U/U-235 depletion vs initial fuel enrichments with different reactivity control methods and moderation ratios for different PWR fuels. (from ref. 23)

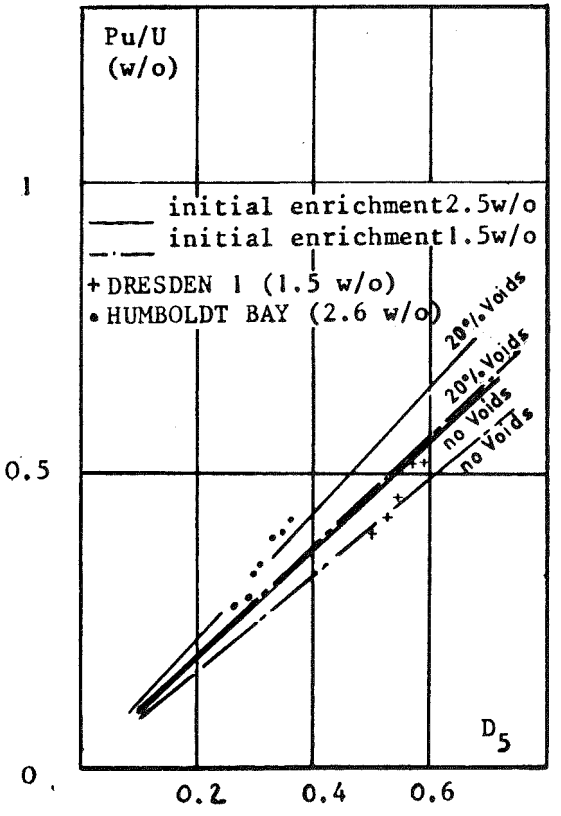


Fig. 15. Calculated Pu/U ratio vs U-235 depletion for different BWR fuels and different void contents of the moderator: comparison with experimental data (from ref. 23)

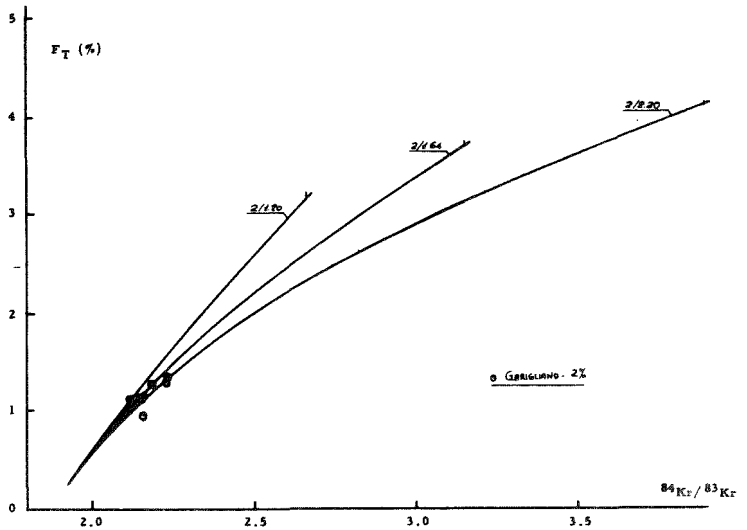


Fig. 16. Calculated heavy atoms burnt (F_t %) vs Kr-84/Kr-83 for different fuel enrichments and moderator to fuel volume ratios (from ref. 25)

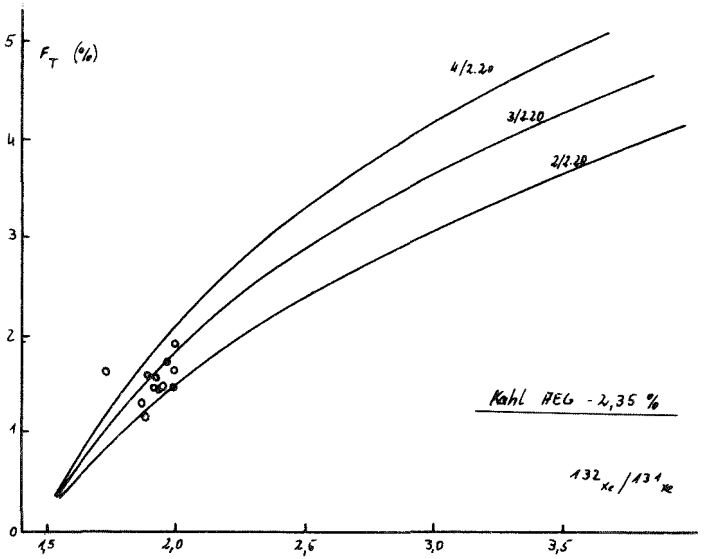


Fig. 17. Calculated heavy atoms burnt (F_t%) vs Xe-132/Xe-131 for different fuel enrichments and moderated fuel volume ratios (from ref. 25)

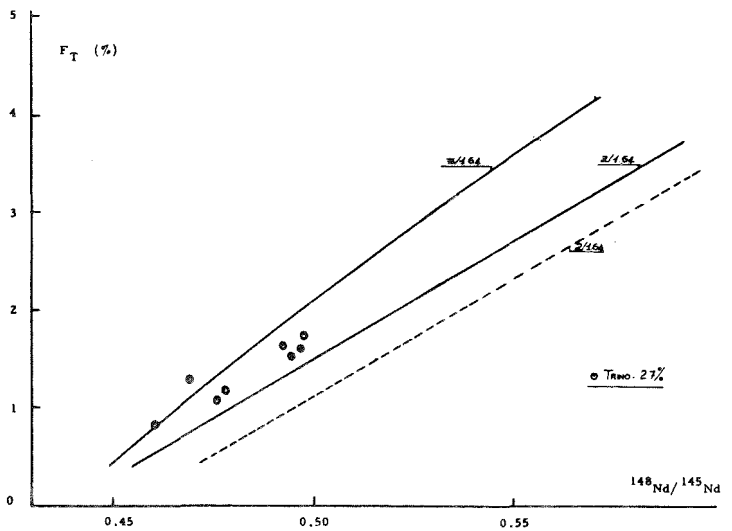
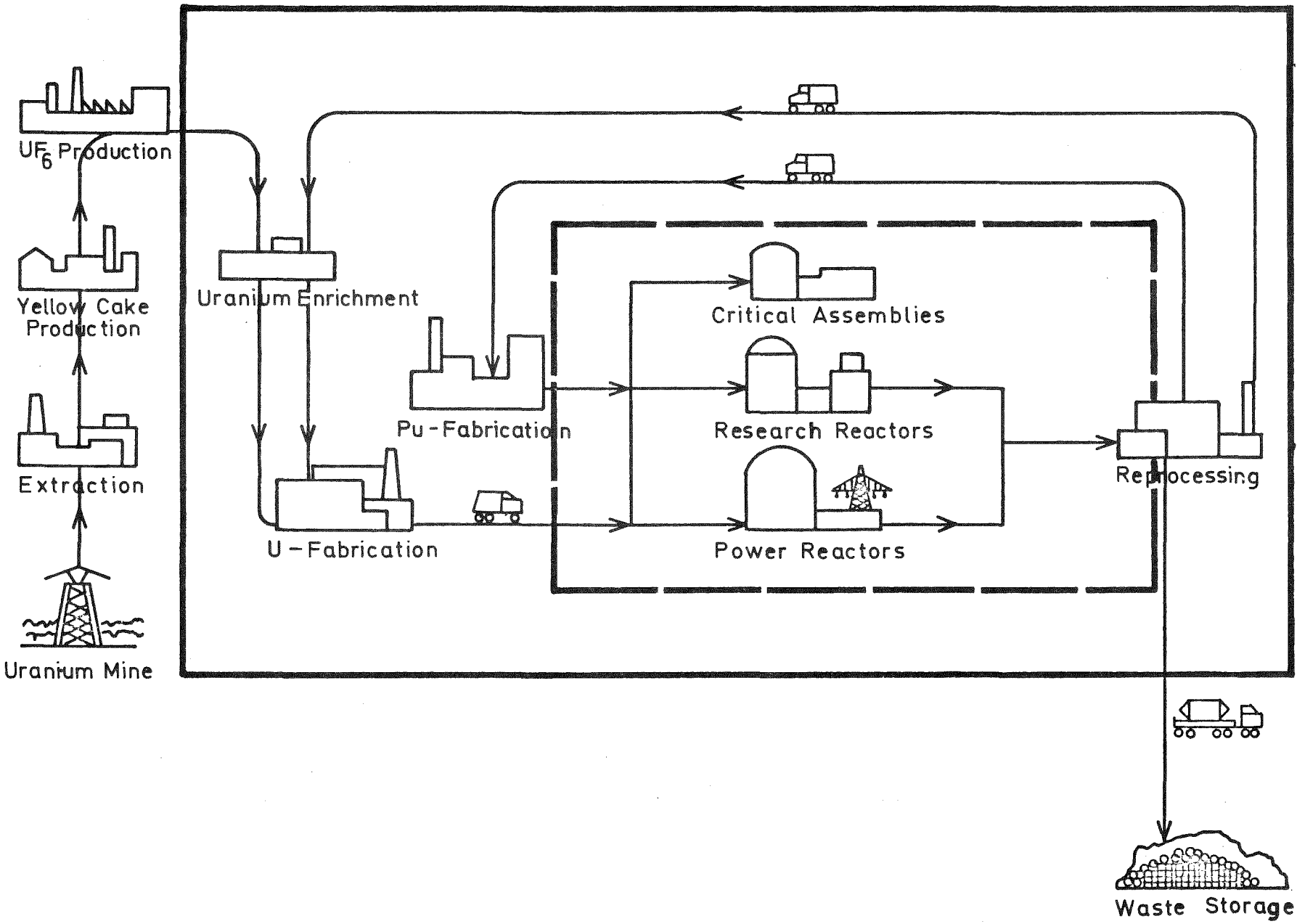


Fig. 18. Calculated heavy atoms burnt (F_t%) vs Nd 148/145 for different fuel enrichments and moderated fuel volume ratios (from ref. 25)

Fig. 19a Nuclear Material Flow in a Commercial Nuclear Fuel Cycle



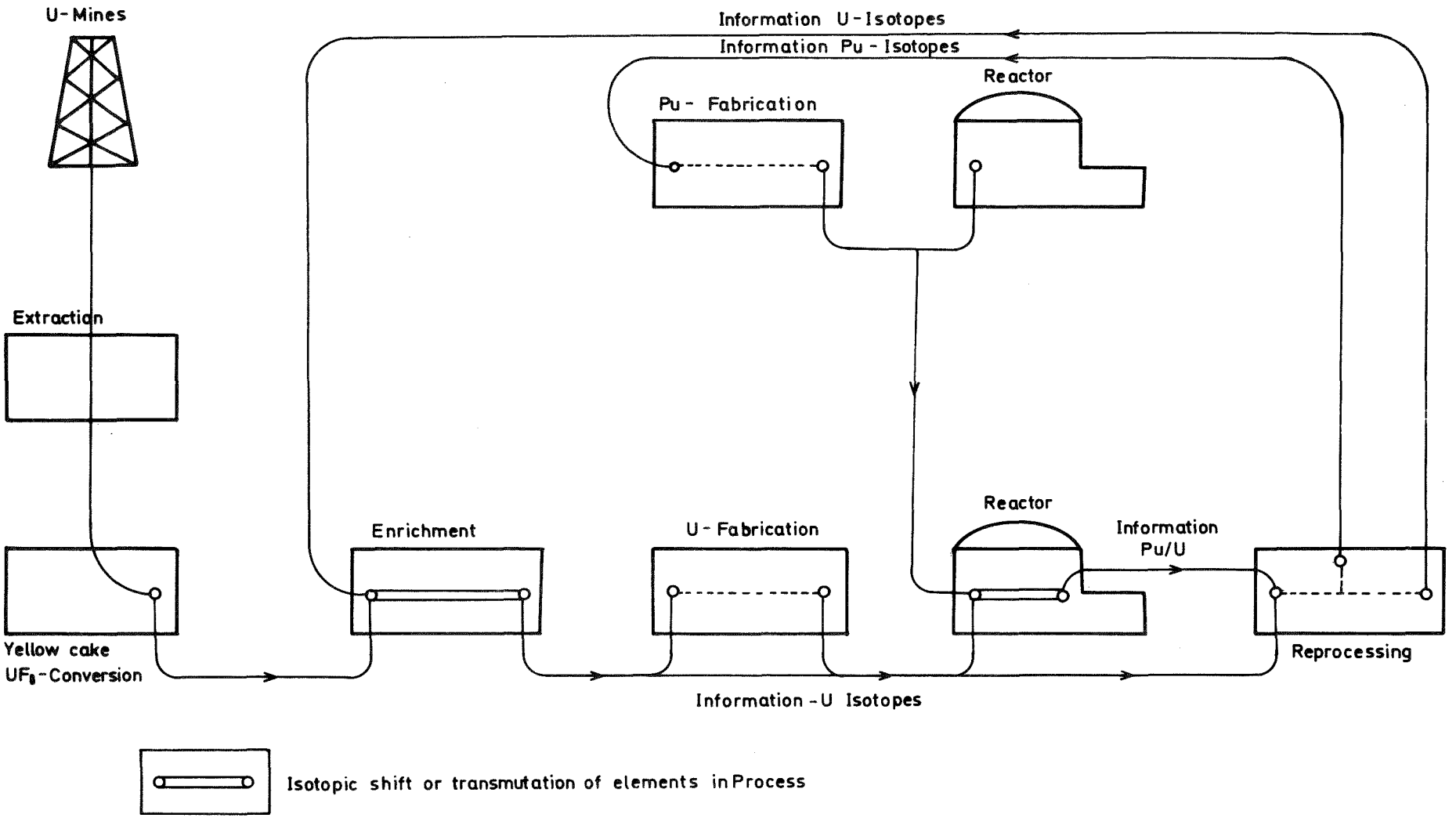


Fig.19b Flow of Information relevant to Isotopic Correlations in a Nuclear Fuel Cycle

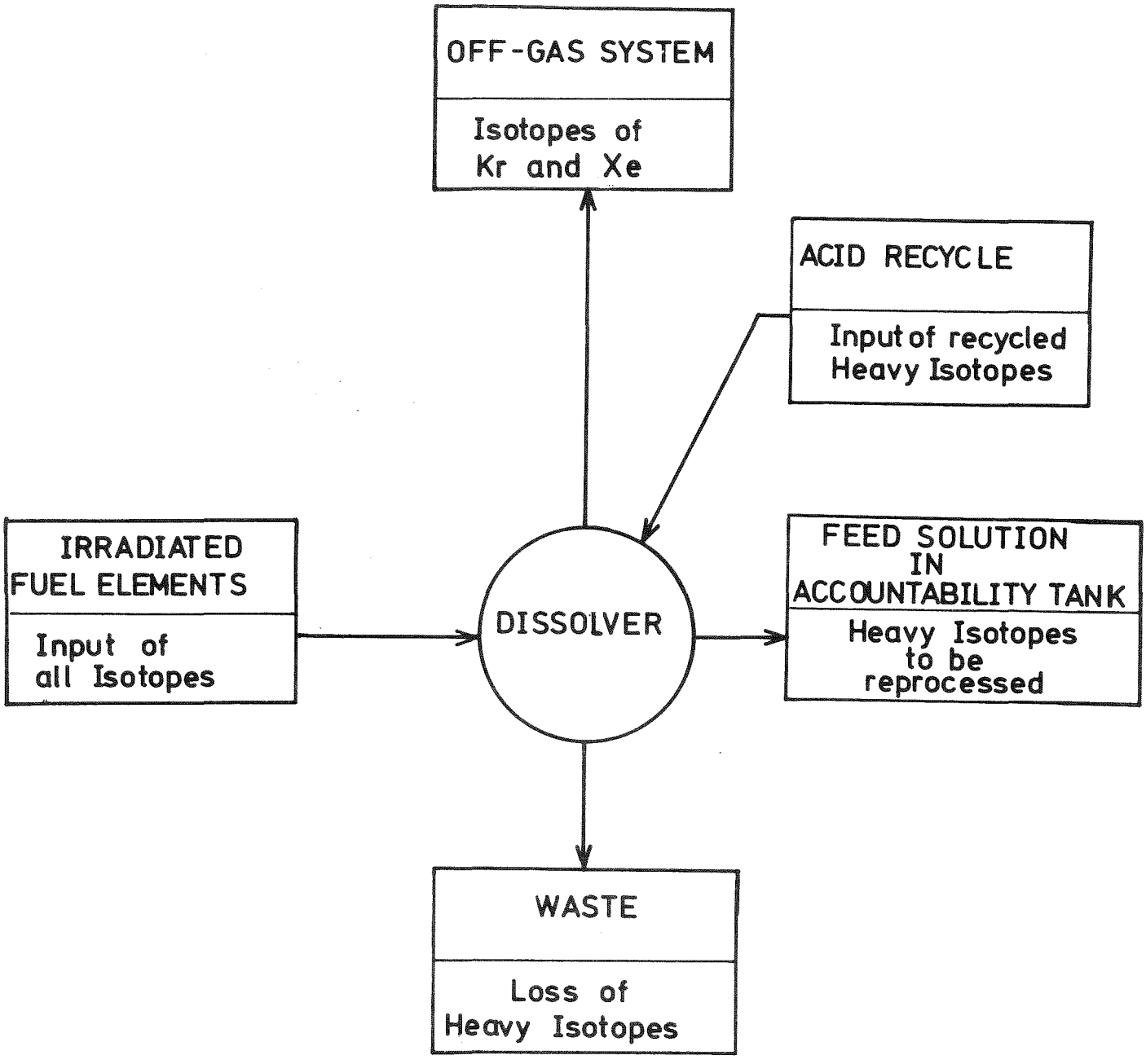


Fig. 20 Relevant Steps for Isotopic Correlations around the dissolver of a reprocessing Plant (Ref. 9)

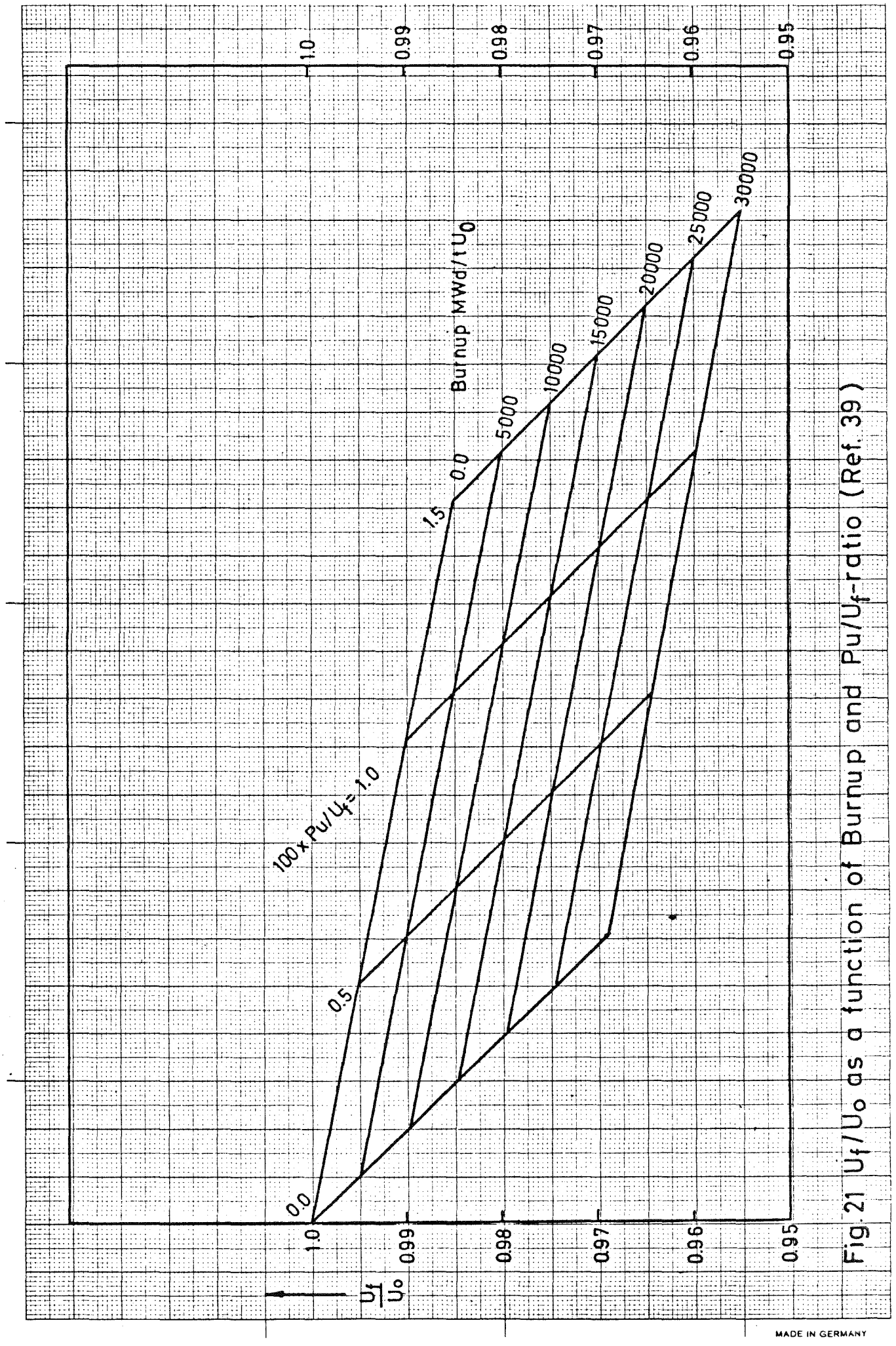


Fig. 21 U_f/U_o as a function of Burnup and Pu/U_f-ratio (Ref. 39)

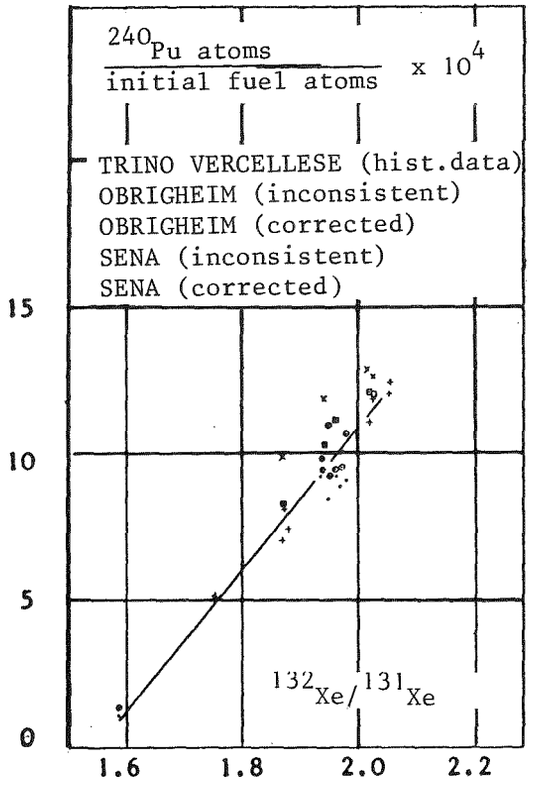


Fig. 22. Application of the correlation between ^{240}Pu buildup and $^{132}Xe/^{131}Xe$ ratio; correction of inconsistent data from Sena and Obrigheim reactor fuels (from ref. 32)

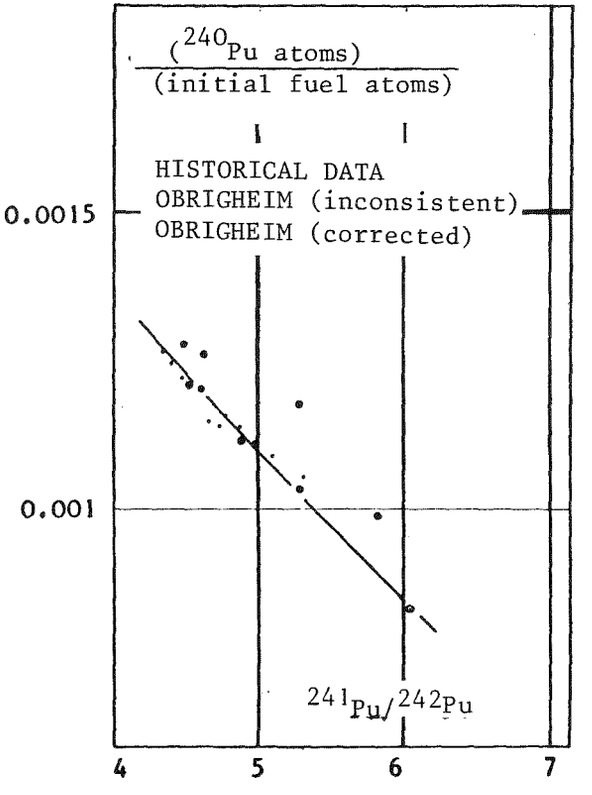


Fig. 23. Application of the correlation between ^{240}Pu buildup and $^{241}Pu/^{242}Pu$ ratio; correction of inconsistent data from Obrigheim reactor fuel (from ref. 32)