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USE OF ISOTOPIC CORRELATIONS IN VERIFICATION SAFEGUARDS APPLICATION

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THE USE OF ISOTOPIC CORRELATIONS IN VERIFICATION (SAFEGUARDS APPLICATION)

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by

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THE USE OF ISOTOPIC CORRELATIONS IN VERIFICATION

I. INTRODUCTION

Previous work[1-11] has shown the potential value of heavy element isotopic correlations to international safeguards. Potential applications are centered on that part of the fuel cycle which extends from the output of fuel fabrication through the input to reprocessing. This paper addresses two specific applications. These are:

1) Verification of the plutonium to uranium ratio of spent low enriched uranium fuels.
2) Confirmation of information which is stated about the spent fuels prior to dissolution at reprocessing, including tests for substitution.

Other applications include the use of isotopic correlations in evaluating the quality of the analytical measurements that are made on reprocessing plant dissolver solutions. The correlations can also serve as a diagnostic tool for deducing additional information about fuel history. These other applications are discussed in previous publications[1-11] and other papers[13] presented at this Panel.

The purpose of this report is to describe the technical bases and conditions for the valid application of heavy element isotopic correlations in international safeguards. Further and equally important aims are to describe the (1) information requirements, (2) inspection and verification activities, and (3) evaluation of results.

As an aid to the reader, the fundamental points are covered in the first 16 pages. This is followed by a detailed discussion of technical applications which includes specific examples, information needs, suggested procedures, and methods of evaluation.

II. TECHNICAL BASES AND CONDITIONS OF APPLICATION

In discussing the safeguards verification of materials accounting data, several terms require introduction. These include:

- Technical Reliability - The precision and accuracy or the exactness of a given method to measure the quantities or compositions of materials or to calculate such properties from other measured data and physical constants.
- Validity of Verification - The ability of a given verification procedure to truly affirm the integrity and quality of materials accounting data.
- Reliance - The extent to which a safeguards authority relies upon the results of a given verification procedure or activity in making tests of significance.

A. Verification of Plutonium Input to Reprocessing

The major safeguards impact of isotopic correlations stems from their potential use in verifying the Pu/U ratio of spent low enriched uranium fuels. In this application, isotopic correlations are used in conjunction with the Pu/U ratio method[7] in verifying the total plutonium input to the reprocessing plant. Here the primary verification of total plutonium is by the Pu/U ratio method. Plutonium input to the reprocessing plant is computed by applying individual or the weighted Pu/U ratio(s) to the shipper's value (fabrication value corrected for burnup) for total uranium. Conventionally, the Pu/U ratio is obtained by the direct measurement of the Pu/U ratio of input dissolver solutions. The isotopic correlations are used to verify or substantiate individual or the weighted

Pu/U ratio of an input campaign. Alternatively, isotopic correlations may be used to estimate the Pu/U ratio from historical relationships and the measured uranium and plutonium isotopic composition of the spent fuel. The isotopic correlations of interest in this application are those relating the plutonium concentration (Pu/U ratio) of a particular spent fuel to the uranium or plutonium isotopic composition, e.g.,

$$Pu/U = k(^{235}D)$$

where $D = \text{wt. \% } ^{235}U \text{ initial} - \text{wt.\% } ^{235}U \text{ final}$

or $$Pu/U = m(^{240}Pu / ^{239}Pu) + b$$

where k, m, and b are constants.

Valid application of the Pu/U ratio method in verification requires 1) that the uranium content of fabricated fuel be accurately known and verified, 2) that the burnup of total uranium can be fairly accurately determined and verified, and 3) that the Pu/U ratio itself be verified. Validity of the method also requires that effective containment and surveillance procedures be applied after fuel fabrication primarily for detecting any additions of uranium. In addition, the reprocessing plant head-end process must allow use of the ratio method.

The technical reliability (as opposed to validity of verification) for the use of the Pu/U ratio method is well established for low enriched uranium fuels; i.e., the capability to measure the total U in fabricated UO_2 fuels, the capability to measure or calculate total U burnup, and the capability to measure the

Pu/U ratio of dissolver solution are well established. A similar reliability is expected for the Pu/U ratio method for the case of plutonium thermal recycle fuels which contain mostly uranium (> 96% UO_2).

The ratio method under the conditions previously described may be used as a primary method of verification, independent of support from the isotopic correlations. The main benefit, however, which can arise from application of isotopic correlations lies in their use to substantiate, reinforce, and validate the Pu/U ratio. Depending on the sampling plan used to independently measure Pu/U ratios, the correlations can be used to reinforce direct independent measurements or they can be a necessary part of the verification process. For fractional sampling plans they can be necessary to the verification in protecting against gross atypical errors (or falsifications) occurring on a few of the input batches. In this case, the correlations minimize this maximum risk by verifying the collective ratio data of a series of input batches.

In a fractional sampling plan approach, the Agency would directly measure the Pu/U ratio and the uranium and plutonium isotopics on a few input batches from a reactor lot. From these measurements it would form correlations between Pu/U and the isotopes, e.g.,

$$Pu/U = K(^{235}D)$$

It would then compare those relationships with similar ones formed from plant operators data on all the batches (assuming $Pu/U = K(^{235}D)$ to hold for all batches). If no differences are noted between the Agency and the operator, the problem of gross atypical errors (or falsification) reduces to errors in isotopic composition. That is (under conditions where the correlations hold) the correct relationships between Pu/U and isotopic composition could not have been reported without both the Pu/U ratio and the isotopic compositions being in error. To protect against such errors the Agency monitors the isotopic compositions across the plant, e.g., the weighted isotopic composition of input equals the weighted isotopic composition of product when computed over reasonable periods of time. The Agency could also use composite samples to provide collective verification of the Pu/U ratio and isotopic compositions.

The technical reliability of using isotopic correlations to verify the Pu/U ratio is dependent on many factors. These include the characteristics of the spent fuel, the scope, exactness, and accuracy of historical information, the reprocessing plant head-end process, and the batching sequence used for dissolving and measuring the spent fuel. By contrast, the extent of application in verification depends on the degree of reliance placed on the correlations for verification of the Pu/U ratio as opposed to the reliance placed on direct measurements by the Agency. Various degrees of reliance have been suggested.(4,8,12) These range from a minimum level of using the correlation as an "addition-to" to reinforce or substantiate, to a maximum level of reliance where the Pu/U ratio is estimated (or verified) solely from established relationships and measured isotopic data. Some workers(12) recognize the robustness added by isotopic correlations; others(8) recognize the practical ease of verifying ratios and isotopic compositions as opposed to verifying element concentrations.

The balance between the reliance placed on isotopic correlations in verification and the reliance placed on direct Agency measurements is dictated by particular circumstances. For a well characterized spent fuel, a high degree of reliance on the correlation techniques is suggested. In other less characterized and more varied situations, less reliance is suggested.

B. Confirmation of Information

During the irradiation of low enriched uranium, the initial isotopic composition of uranium is changed as well as the isotopic composition of the plutonium produced in that process. Graphs of the ratios of the various uranium and plutonium isotopes from zero exposure to the end-of-life are a portrayal of the burnup (and buildup) paths of those isotopes. The isotopic compositions observed in spent fuel dissolver solutions represent the end point of those burnup paths. Extrapolation backwards along the burnup paths from end-of-life to zero exposure yields the initial isotopic composition of the spent fuel. Through such extrapolation techniques, it is possible to confirm the initial isotopic composition of the fuel with some degree of exactness. Correlations useful for this purpose include Pu/U versus final ^{235}U, Pu/U versus final ^{236}U, and published or measured alpha values for ^{235}U. These, of course, are not a substitution for direct verification at the time of fabrication.

The measured values on dissolver solutions of spent fuel for the Pu/U ratio and uranium and plutonium isotopic composition may be used to confirm with a high degree of exactness shippers' values for the Pu/U ratio, and isotopic compositions. The isotopic relationships themselves tend to be characteristic of a given reactor type and can be used to distinguish one ractor type from another.

Another area of safeguards concern is the possibility of substitution taking place after fuel fabrication. In this regard, the isotopic correlations and Pu/U ratio data can provide a degree of "after the fact" quantitative assurance that certain forms of substitution did not take place. In general, the use of all isotopic data makes substitutions that alter isotopic compositions difficult to conceal. The substitution of natural or depleted uranium for low enriched uranium would be detected through confirmatory tests of initial enrichment and comparison to historical values for relationships involving the initial enrichment. Substitution of a few rods which are identical but unirradiated into 11 irradiated assemblies would be detectable by relationships involving the plutonium isotopes such as Pu/U versus $^{240}Pu/^{239}Pu$. In this situation, the plutonium isotopes would retain their exposure dependent composition, e.g., a high $^{240}Pu/^{239}Pu$ ratio, whereas the Pu/U ratio and ^{235}U depletion would be reduced. This comparison would require "true" historical values as a basis for testing.

Other tactics such as the substitution of entire fuel bundles of a lower exposure for ones of higher exposure would be extremely difficult to detect by isotopic data alone. Reported reactor data would also have to be altered to the same extent.

Other confirmatory type tests which arise from use of isotopic correlations include 1) tests for the selective removal of plutonium (e.g., precipitation or plating) prior to the input accountability tank, and 2) tests for the introduction of recycle material into head-end tanks. The most important test is the test for the selective removal of plutonium. In this situation, comparisons to "true" historical values would detect plutonium removal within the technical reliability of pre-established values for Pu/U versus ^{235}D or Pu/U versus $^{240}Pu/^{239}Pu$.

C. Supporting Requirements

The full use of isotopic correlations in verification depends on certain supporting activities, process features, and availability of information. With the exception of inspection and evaluation activities, many of the conditions for (at least partial) use of the isotopic correlations currently exist. This conclusion is based on currently operating or proposed fabrication plants, reactors, and reprocessing plants and upon current measurement practices and availability of supporting information.

The main process requirement at reprocessing for the use of the Pu/U ratio method and for the isotopic correlations is that the input dissolver solutions be truly representative, e.g., do not contain significant quantities of extraneous nuclear materials such as recycle. Recycle materials can be handled without adverse effect in some situations, by taking specific actions. A second requirement is that it be possible to know the order of dissolution of fuel assemblies and also be possible to roughly relate input accountability batches to the particular fuel bundles from which the batch is made up.

The more important information to be obtained from or reported by plant operator include 1) the total U, total Pu, and U and Pu isotopic compositions of fuel bundles both before and after irradiation, 2) the Pu/U ratio and uranium and plutonium isotopic compositions of input dissolver solution batches, 3) head-end waste losses, and 4) the uranium and plutonium isotopic compositions of product batches. In case, complete isotopic data are not readily available, the power of the technique is reduced. The general order of importance of the isotopic compositions (decreasing importance) is for uranium: ^{235}U, ^{236}U, ^{238}U, ^{234}U; and for plutonium: ^{239}Pu, ^{240}Pu, ^{241}Pu, ^{242}Pu, and ^{238}Pu.

The correlation techniques have their maximum use in the verification of the Pu/U ratio when it is possible to apply them to successive lots of fuel from the same reactor. The validity with which they may be applied in this type of application is greatly enhanced when the irradiation conditions are well know. Thus, knowledge of those factors which influence the correlations (such as fuel design, cladding, fuel to moderator ratio (including void fraction for BWR's), initial enrichment, burnable poison level, operating mode etc.) allow accumulated data to be applied with high validity. In many cases, it is sufficient to know only that the conditions which influence the correlations are the same for successive reactor lots. However, in other cases when it is known quantitatively that certain conditions have changed, it is possible from theoretical considerations to predict with some exactness the effect of such changes.

Reactor statements for the end-of-life composition are important to the application of the correlation techniques. The reactor statements for total uranium serve as the shipper's value for total U in the application of the Pu/U ratio method. Shipper's statements for total Pu, ^{235}U, ^{236}U, the plutonium isotopic composition, and exposure are extremely useful in safeguards pre-planning activities. From those data, "predicted" correlation values can be computed and optimum sampling plans for Agency verification can be established in advance.

If isotopic data are also available on feed and product of uranium enrichment plants and from plants receiving product from the reprocessing plants, this ring of isotopic data makes the falsification of isotopic data a near impossibility, even under modest levels of verification.

III. SUMMARY AND CONCLUSIONS

A review was made of the work done to date on isotopic correlations of potential safeguards value. Particular attention was given to those correlations involving the uranium and plutonium isotopes and the Pu/U ratio of spent low enriched uranium fuels. Two applications which have potential value in Agency verification activities were considered in detail. These are:

- The use of isotopic correlations in supporting the Pu/U ratio method of verifying the plutonium input to reprocessing.
- The use of isotopic correlations and associated dissolver solution data in confirming safeguards information about spent fuels which is stated prior to receipt at the reprocessing plant. This aspect includes tests for substitution.

The following overall conclusions have been reached for the case of low enriched uranium fuels. It is believed that the above applications have been sufficiently demonstrated to be included in Agency verification activities. The technical basis and conditions of applications for use of the Pu/U ratio method are well established. The isotopic correlations of plutonium and uranium have also been demonstrated both experimentally and theoretically. The technical basis of supporting isotopic techniques such as the "across-the-plant" balance for isotopic composition at chemical reprocessing is also well established.

Several important questions must be resolved in regard to the application of isotopic correlations in safeguards. These are:

- The degree of reliance which should be placed on their use in verification.
- The nature of the information which will be required of plant operators for full use of the techniques and the manner in which that information is transmitted or made available to the Agency.
- The nature of the inspection procedures and activities required for use of the Pu/U ratio method and the correlations.
- The data evaluation procedures and staff capabilities which would be required for practical implementation.

This report addresses the questions of reliance and data evaluation. For the other points, it endeavors to provide a descriptive basis to assist the Agency in establishing information requirements and inspection activities.

The question of reliance is treated in two parts. First is the reliance placed on the Pu/U ratio method for verifying the total plutonium input to reprocessing. Second is the reliance placed on isotopic correlations as a means of independently estimating or verifying the Pu/U ratio of reprocessing plant input batches.

A major concern about the Pu/U ratio method is the validity of using the shipper's value for total U as the basis for converting the weighted or individual Pu/U ratios to total plutonium. The main conditions of validity are met when international independence exists or when the Agency is able to verify the total uranium content of fabricated fuel to the desired degree of exactness and maintain that verification through containment and/or surveillance until reprocessed. The concern in the verification of fabricated fuels is that the total uranium not be understated at fabrication, e.g., overload the rods.

It is apparent that for low enriched uranium fuels, the Agency can always use the shipper's value for total U to some advantage. Whether it uses that value as a primary means of verification or simply as a check, depends on its evaluation of validity (as discussed above).

Various degrees of reliance have been suggested for use of isotopic correlations to verify the Pu/U ratio. Suggestions range from a minimum level of their use as an "addition to" to a maximum level of use as the sole means of independently estimating or verifying the Pu/U ratio. The level of use depends on the relevant circumstances. The minimum level of using the correlations as a means of substantiating and reinforcing measurements made by the Agency will add to the robustness of the verification process. The maximum level of use is based on the situation where the correlations for a given fuel have been characterized to a point approaching the certainty of physical constants. The authors favor a balanced approach where the correlations are used in conjunction with some Agency measurements for the Pu/U ratio and isotopic compositions.