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A Reappraisal of Experimental Relative Conversion Ratios for Early Cores in DIMPLE

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A REAPPRAISAL OF EXPERIMENTAL RELATIVE CONVERSION RATIOS FOR EARLY CORES IN DIMPLE

M F Murphy

SUMMARY

Recent measurements of the neutron capture rates in U238 in DIMPLE assemblies have highlighted an error in earlier DIMPLE benchmark data. Although the source of the error was identified many years ago, revised data have never been formally published. To remedy this omission, corrections for the early results have been derived to obtain a recommended set of values. On average the corrected results are (3.3±0.5)% larger than the original values. It is shown that the corrected results are in good agreement with other early measurements, which were rejected at the time, and with those obtained using current techniques.

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1 INTRODUCTION

A series of measurements in 3% uranium oxide water moderated lattices in DIMPLE (1) have been used for many years to validate the WIMS codes. At the time the measurements were made it was noted that there was a discrepancy between the U238 capture results quoted and those obtained from an independent chemical method.

Subsequently a possible source of the discrepancy between the two experimental methods was identified. However, this was not widely published and the original results were again used for WIMS validation in 1981 (2).

Recent measurements in DIMPLE, in a re-build of one of the earlier lattices, have confirmed that the original U238 capture results were in error. On the basis of this work and the earlier studies a corrected set of data for the early DIMPLE benchmarks is derived below.

2 EARLY STUDIES

In 1965 a series of regular 3% uranium oxide water moderated lattices were studied in DIMPLE (1). Extensive reaction rate measurements were made, including determinations of the Relative Conversion Ratio (RCR). This is defined as:-

$$\text{RCR} = \frac{\dfrac{\text{U238 capture rate in the lattice}}{\text{U235 fission rate in the lattice}}}{\dfrac{\text{U238 capture rate in a thermal spectrum}}{\text{U235 fission rate in a thermal spectrum}}}$$

The determinations of the U238 capture rates were carried out by two methods, in order to indicate the presence of possible systematic errors. The main method (3) involved detecting the neutron capture induced Np239 activity from a thin slice of the uranium oxide fuel using a sodium iodide γ-X-ray coincidence system. As an independent check, a smaller number of determinations were made using a chemical separation method (4). This involved dissolving an irradiated uranium oxide fuel pellet, separating the Np239 from the fission products and uranium by organic solvent extraction, and producing a deposit of Np239 by electro-deposition. This deposit, which included Np237 as a tracer, was then counted for α and γ activities. In reporting the results obtained (1) it was noted that the chemical separation method gave values that were consistently higher than the coincidence method by about 3%. It was stated in Reference 1 that, "In view of the internal inconsistency of these chemical results and the early state of development of the technique, the results have not been included in the main assessment of the U238 captures in these lattices".

Before and after the publication of Reference 1 in 1967, investigations were carried out to try to identify the reason for the discrepancy between the two techniques. Eventually it was found that the method of correcting for fission product interference in the coincidence technique was incorrect. This information was published as an internal note in January 1968 and an edited version is included as an appendix here. It was also discussed in Reference 5.

It was found that, when the fission product correction was made correctly, the discrepancy between the chemical technique and the coincidence technique was very much reduced (5).

3 RECENT MEASUREMENTS

In 1983 DIMPLE was recommissioned for a new programme of reactor physics and criticality experiments. To re-establish measuring and operating procedures it was decided to rebuild assembly R1/100H, reported in Reference 1. In the present series of DIMPLE experiments this assembly is known as S01. Most of the reaction rates previously reported were remeasured, including the RCR. The original measurements were made over twenty years ago and, not surprisingly, the techniques of measuring the U238 capture rate have changed in the intervening years.

The chemical separation technique has, in effect, been discontinued. It is a complicated and time consuming process that has served its purpose as an indicator of systematic errors.

The original coincidence counting method has been modified. Firstly, an aliquot of a dissolved fuel pellet is now counted, as a liquid source, rather than counting a solid disc of uranium oxide. The discs, which needed to be matched very precisely in terms of flatness and diameter to the adjacent pellets, were both costly and fragile. It was also necessary to mask the edges of the discs during counting, since the high activity due to resonance neutron capture at the edges would otherwise have been overemphasised by the lack of gamma attenuation.

The second change to the coincidence counting method is that the fission product interference correction is now made as part of the main measurement, rather than by using the results of a subsidiary measurement. The early method is outlined in the Appendix. It consisted essentially of measuring the coincidence count-rates from two fuel samples with different enrichments and solving the two simultaneous equations to obtain the rates due to fission and those due to U238 capture. The present method uses 97% enriched uranium oxide powder to provide a source of fission products with a negligible Np239 contribution to the coincidence counting. Since the correction predominates in the thermal column, the powder is irradiated there, sufficiently far removed from the fuel disc to avoid any perturbation. As with the fuel capsule in the reactor, both

the disc and the powder are converted to liquid sources prior to counting. The fission rates in all three samples are related by counting with a conventional sodium-iodide detector above a bias of 0.6MeV. This method eliminates the need to know the relative flux depressions in the thermal column irradiations, which was the problem with the earlier work. A full description of the current technique is given in Reference 6.

To check on possible systematic errors in the current technique, the Np239 activity from the lattice and thermal liquid sources is also measured by gamma spectroscopy using a germanium semi-conductor detector. This detector can exclude interfering fission product activities by virtue of its good energy resolution. Thin discs of uranium oxide from the fuel, which are used to establish the U235 fission rate, are also measured on this detector, to check for gross errors in the dissolution process and in the dispensing of the liquid sources. In the measurements in assembly S01, accurately machined discs identical to those used for the RCR work in R1/100H, as well as specially machined pellets, were irradiated to provide a check of all the techniques.

The results obtained for core S01 are given in Table 1 for the coincidence technique and in Table 2 for the gamma spectroscopy technique. Figure 1 shows typical pellet packs used for the irradiations. It can be seen from the tables that the results for each technique are consistent within the experimental errors. In addition the mean values of RCR from the coincidence counting and the gamma spectroscopy are in good agreement, ie 4.28±0.5% and 4.26±0.5% respectively. The results are also in good agreement with the value of 4.31±0.7%, obtained by chemical separation, reported for R1/100H. However, the coincidence counting result reported for R1/100H, of 4.15±0.7% (1), is 3% lower than the current results for RCR in S01.

4 REVISED VALUES FOR EARLIER DIMPLE CORES

4.1 Justification for Revisions

It is certain that the coincidence counting RCR values reported in Reference 1 are incorrect. This statement is justified by the following:

(i) A discrepancy of about 3% was found between the coincidence technique results and the chemical technique results in R1/100H(1).

(ii) A definite error was later found in the coincidence counting analysis (Appendix to this note).

(iii) A range of measurements, with a correct analysis of the coincidence fission product corrections, gave agreement to about 1% between the coincidence and chemical techniques (5).

(iv) Recent measurements of RCR using coincidence and gamma-spectroscopy techniques have confirmed the earlier chemical results.

Accepting that the coincidence counting results reported in Reference 1 are incorrect due to a known error, then it is possible to make reasonable estimates of what the values of RCR should have been. There are a number of ways of estimating the magnitude of the adjustments required to the results. These are detailed below.

4.2 Use of Chemistry Data

The chemical separation results for each core could be simply accepted as the best values. This has the disadvantage of rejecting all the information contained in the coincidence results.

To try to make use of the coincidence information, the mean ratio between the chemical and coincidence methods for the series of cores can be evaluated and applied as a normalisation to the coincidence results. The justification for this is that the self consistencies of the coincidence results are better and a renormalisation improves the absolute values, assuming that the chemical results are correct. It should be noted here that most of the fission product correction (~75%) is applied to the thermal column sample, so that the total correction is relatively insensitive to the variations in capture rate from core to core in DIMPLE. Considering the results in Appendix 8 of Reference 1, it is found that the weighted mean ratio of chemical results to coincidence results is 1.033±0.6%.

4.3 Revision of Fission Product Corrections

There is an approach that does not rely on using the chemical results at all. This involves estimating the differences between the coincidence results evaluated firstly with the old fission product corrections and secondly with the later corrections. This is not completely straightforward, as the fission product correction is time dependent and the exact times of the measurements are not now known. However, it is probable that all the measurements were made over approximately the same time span after irradiation. Normally the coincidence measurement starts about one day after the irradiation and continues for one or two days. At one day after irradiation the fission product correction that would have been applied to the early results would have been 5.8% and at two days after irradiation it would have been 2.3%. Using fission product data that were available later, it has been deduced that these corrections should have been 9.3% and 5.8% respectively. The effect of applying the later correction is therefore to increase the U238 capture ratios by (3.5±1.0)% and hence to increase the RCRs by the same amount. The uncertainty has been set to cover the assumptions in this approach, typically the uncertainty in the fission product correction is an order of magnitude smaller than that quoted.

4.4 Recommended RCR Values

Using the mean ratio approach, as discussed in Section 4.2 indicates that the coincidence RCR values reported in Reference 1 should be increased by (3.3±0.6)%. Applying later fission product correction data to the coincidence values, as discussed in Section 4.3, indicates that the RCR values should be increased by (3.5±1.0)%. The weighted mean of the adjustments produced by these two approaches is +(3.3±0.5)%. This normalisation has been applied to the coincidence counting results reported in Reference 1 and the old and new values are given in Table 3 of this note. Also included in Table 3 are the chemical separation results and the recent coincidence counting results. The recommended results are those in the final column of the table, with the S01 result being used in preference to R1/100H because of the smaller uncertainty on the recent measurement.

5 SUMMARY

The RCR results reported for early DIMPLE benchmark assemblies have been reviewed in the light of results from recent measurements and taking into account known errors in the original experimental analysis. It is recommended that the original coincidence counting results, and hence the RCRs, should be increased by (3.3±0.5)%. Recommended results for DIMPLE Assemblies R1/100H, R1/100H/H, R1/83H and R3/100H consistent with this approach have been derived.

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4 G A Barnett et al. The Separation and Precise Comparative Determination of Np239 in Irradiated Low Enrichment UO_2 Pellets and Foils. AEEW - R 202 (1962).

5 G A Barnett et al. Measurement of Relative Conversion Ratio and Fast Fission Ratio in Low Enrichment Oxide Lattices. AEEW - R 648 (1969).

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Table 1

RCR in DIMPLE Assembly S01, Coincidence Counting of Liquid Sources for Np239 Activity

Irradiation No	Lattice Position	F_5 L/T	% Error	C_8 L/T	% Error	RCR	% Error	Mean RCR	Overall Mean RCR
	A	0.1236	0.25	0.5277	0.35	4.269	0.45		
	B	0.1226	0.25	0.5290	0.35	4.315	0.45	4.276	
1	C	0.1040	0.25	0.4425	0.35	4.255	0.45	±0.3%	
	D	0.1212	0.25	0.5147	0.35	4.247	0.45		
	E	0.1107	0.25	0.4756	0.35	4.296	0.45		4.284 ±0.3%
	D	0.1188	0.5	0.5114	0.7	4.305	0.9		
2	D	0.1194	0.7	0.5141	0.35	4.306	0.8	4.310	
	D	0.1182	0.7	0.5107	0.35	4.321	0.8	±0.4%	
	E	0.1093	0.5	0.4711	0.35	4.310	0.6		

Notes

1. F_5 L/T is the lattice to thermal U235 fission ratio.

2. C_8 L/T is the lattice to thermal U238 capture ratio.

3. RCR is the ratio $(C_8\ L/T)/(F_5\ L/T)$.

4. All the lattice positions, A to E, were nominally identical and located adjacent to the lattice centre.

5. The random errors only are shown in the table above. There is also a systematic contribution of ±0.4%, arising mainly from holder calibration and sample dispensing (5).

Table 2

RCR in DIMPLE Assembly S01, Ge(Li) Counting for Np239 Activity

Irradiation No	Lattice Position	Sample Form for C_8 Measurements	F_5 L/T	% Error	C_8 L/T	% Error	RCR	% Error	Mean RCR	Weighted Mean RCR
1	C D E	0.127mm discs	0.1040 0.1212 0.1107	0.25 0.25 0.25	0.441 0.510 0.475	1.6 1.1 1.8	4.24 4.21 4.29	1.6 1.1 1.8	4.23 ± 0.8%	4.26 ±0.5%
2	D E	0.127mm discs	0.1188 0.1093	0.5 0.5	0.514 0.468	1.2 1.3	4.32 4.28	1.3 1.4	4.30 ± 1.0%	
2	D E D D	Liquid Sources	0.1188 0.1093 0.1194 0.1182	0.5 0.5 0.7 0.7	0.506 0.465 0.515 0.510	1.7 2.0 1.4 1.8	4.26 4.25 4.31 4.31	1.8 2.1 1.6 1.9	4.28 ± 0.9%	

Table 3

Comparison of RCR Values

Core	Old Coinc RCR	% Error	Chem RCR	% Error	New Coinc RCR	% Error
R1/100H	4.149	0.7	4.314	0.7	4.285	0.9
S01	-	-	-	-	4.284	0.5
R1/100H/H	4.263	1.1	4.323	1.2	4.404	1.2
R1/83H	4.508	1.1	4.610	2.0	4.657	1.2
R3/100H	4.789	1.1	4.945	1.2	4.947	1.2

APPENDIX

Discrepancies in Early RCR Measurements

(An edited version of an internal note dated January 1968)

1 INTRODUCTION

The physics and chemistry methods of U238 capture rate determinations in low enrichment lattices were described in References 2 and 3.

The two methods gave good agreement (within 2%) when the fuel enrichment was 1.1% but when the enrichment was increased to 3% a consistent discrepancy of about 3% (with the chemistry method higher than the physics) clearly emerged. As shown in Table A.1 further measurements indicated that the discrepancy increased rapidly with enrichment.

The discrepancy caused both the physics and chemistry methods to be looked at very closely. No fault was found in the chemistry method, but investigations into the physics method in 1968 revealed an error which accounted for the major part, if not all, of the observed discrepancy.

Table A.1

Magnitude of Discrepancy as a Function of Fuel Enrichment

Fuel Enrichment	RCR		% Discrepancy
	Physics	Chemistry	
0.04%	2.154±0.008	2.153±0.006	Nil
3%	many measurements		3%
9%	10.73 ±0.25%	12.042±0.5%	12%

2 ERROR IN PHYSICS METHOD

The experimental evidence indicated that the discrepancy was associated with activity from fission products, since their contribution to the total induced activity increases with enrichment. These fission products generate coincident radiations that are recorded with the required coincident radiations that are emitted when Np239 decays into Pu239 thus recording a U238 neutron capture. A correction is made to allow for the fission product coincidences and it is the estimation of this correction that was found to be in error. To obtain the required Np239 coincidences the measured

coincidence rate is reduced using a time dependent correction function Q(t), where:

$$Q(t) = \frac{\text{Fission product coincidences count rate per U235 atom}}{\text{Np239 coincidences count rate per U238 atom (after correction for } \gamma \text{ and X-ray absorption)}}.$$

To measure Q(t), two foil packs were irradiated in a uniform thermal spectrum for the standard time of two hours. One pack (E) used approximately 18% enriched uranium oxide, the other (B) approximately 2% and each consisted of a 0.125mm thick uranium oxide measuring foil guarded on each side by similar foils to eliminate fission product losses. An hour after the end of the irradiation, the foils were counted for U235 fission product activity. The ratio of U235 fission product activity per U235 atom in the two foils was then derived; let the value of this ratio be K, say, and

$$K = \frac{\bar{\phi}_E}{\bar{\phi}_B}$$

where $\bar{\phi}_E$ is the average flux intensity in foil E and $\bar{\phi}_B$ that in foil B. K was found to be about 0.82.

After about 24 hours from the end of irradiation the foils were counted for coincidence activity and, if F_5 is the coincidence count rate from fission products per U235 atom per unit flux and A_8^T the neptunium coincidence count rate per U238 atom per unit flux after correction for transmission losses; the coincidence count rates C_E and C_B may be written as:-

$$C_E = \bar{\phi}_E N_5^E F_5 + \bar{\phi}_E N_8^E A_8^T a_E \qquad (1)$$

and

$$C_B = \bar{\phi}_B N_5^B F_5 + \bar{\phi}_B N_8^B A_8^T a_B \qquad (2)$$

where N_5^E is the number of U235 atoms in foil E and a_E is the correction for transmission losses in foil E, and similarly for foil B.

putting $\bar{\phi}_E = K\bar{\phi}_B$

and solving (1) and (2) for $Q(t) = \frac{F_5}{A_8^T}$

gives:

$$Q(t) = \frac{C_B\ N_8^E\ a_E K\ \left(1 - \dfrac{C_E\ N_8^B\ a_B}{C_B\ N_8^E\ a_E K}\right)}{C_E\ N_5^B\ \left(1 - \dfrac{C_B\ N_5^E\ K}{C_E\ N_5^B}\right)} \qquad (3)$$

Since the amount of U235 in the two foils differs by a factor of ten the ratio of the flux in the two foils, K, is significantly different from unity. This is obvious and elementary and it was assumed that the computer code used to determine Q(t), made an appropriate correction. A consideration of the input data to this programme, however, showed that it could not possibly make such a correction and a detailed inspection of the programme confirmed this. This then was the error in the physics method; the function Q(t) was wrong because no account was taken of the relative thermal flux perturbations in the measuring foils. In the example given this results in an error of about 50% in Q(t).

This can be seen from a consideration of (3) with values inserted appropriate to an 18% and a 2% disc at one day after irradiation:

$$Q(t) = 16.2 \times K \left[\frac{1-1.91/K}{1-5.62xK}\right]$$

With K=1 this gives Q(t)=3.19 and with K=0.82 (the flux perturbation of the 18% disc relative to that of the 2% disc, described previously) it gives Q(t)=4.89.

The magnitude of the net correction for fission product coincidence is about 2½% for 1.14% enriched fuel and about 7% for 3% enriched fuel. Thus an error of approximately 50% in Q(t) only becomes obvious when 3% and higher enriched fuel is used.

3 COMPARISON OF PHYSICS WITH CHEMISTRY METHODS USING CORRECT DATA

The application of the corrected values of Q(t) indicates that the discrepancy has largely if not completely vanished. This is illustrated below in Tables A.2 and A.3 for 3% enriched measurements in two early DIMPLE assemblies (ME 2/1 and 2/2).

Table A.2

U238 Capture Measurements in DIMPLE (ME 2/1)

Meas No	Physics Coincidence Foil	Chemistry				Chemistry (Foil)	Chemistry (Pellet)
		Foil	Error %	Pellet	Error %	Coincidence	Coincidence
1	0.7036±(0.22r,0.6s)%	0.7146	±0.5%	0.7037	±0.5%	1.015	1.00
2	0.7032±(0.22r,0.6s)%	0.7146	±0.5%	0.7148	±0.5%	1.016	1.016
3	0.7036±(0.22r,0.6s)%	0.7151	±0.5%	0.7184	±0.3%	1.016	1.021
4	0.7037±(0.22r,0.6s)%	0.7094	±0.5%	0.7068	±0.1%	1.008	1.004
					mean	1.014	1.010

Table A.3

RCR Measurements in DIMPLE (ME 2/2)

Meas No	Physics Coincidence Foil	Chemistry		Chemistry (Foil)
		Pellet	Error %	Coincidence
1	1.97±(0.46r,0.6s)%	1.98	±0.25%	
2	1.98±(0.45r,0.6s)%			1.005
3	-	1.98	±0.25%	
4	1.96±(0.47r,0.6s)%			

It will be seen that the results in Table A.2 show a discrepancy of 1.2% and in Table A.3 a discrepancy of 0.5% giving a mean value comparable to the 0.6% systematic error in the coincidence measurement, ie the discrepancies obtained using the correct values of Q(t) are now hardly significant.

To demonstrate the effect of the change in the value of the Q(t), the results from Table A.2 are juxtaposed with the results of the same measurements corrected with the old value of the function in Table A.4.

Table A.4

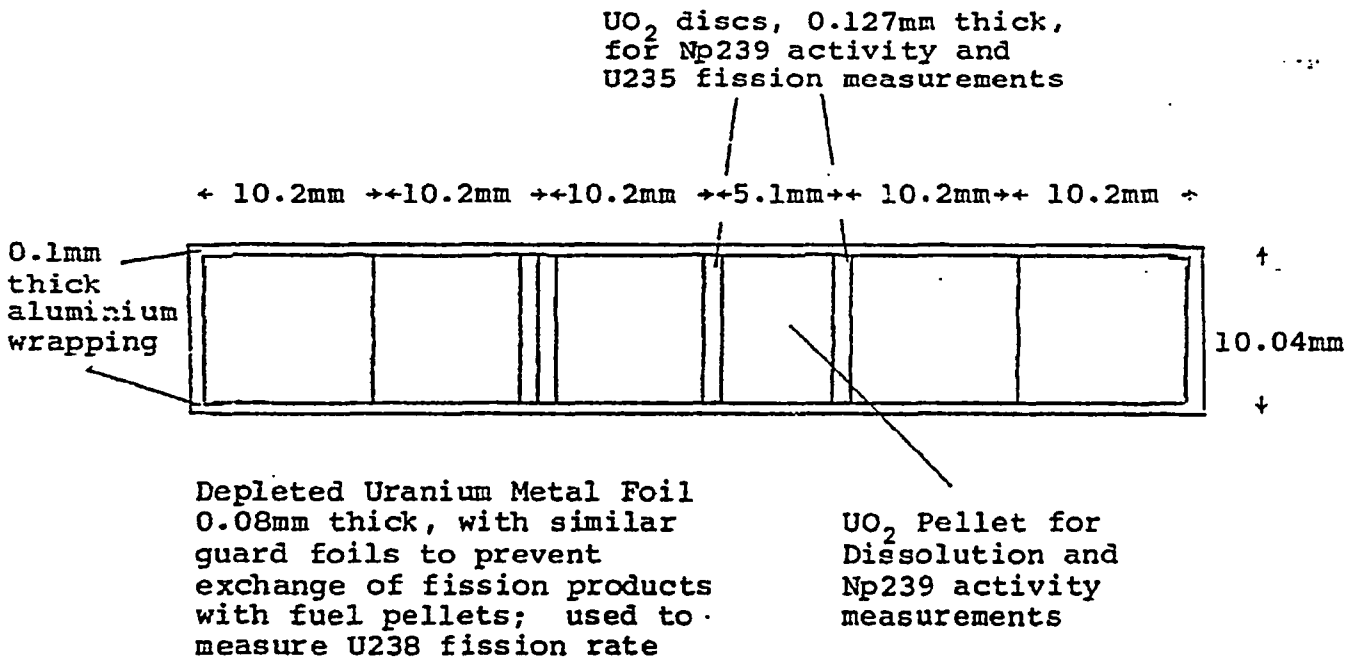
Comparison of Old and New Evaluations of RCRs

Meas No	Physics using old Q(t) values	Physics using new Q(t) values	Chemistry Pellet	Chemistry / Physics old values
1	0.688	0.704	0.704	1.023
2	0.688	0.704	0.715	1.039
3	0.688	0.704	0.718	1.044
4	0.689	0.704	0.707	1.026

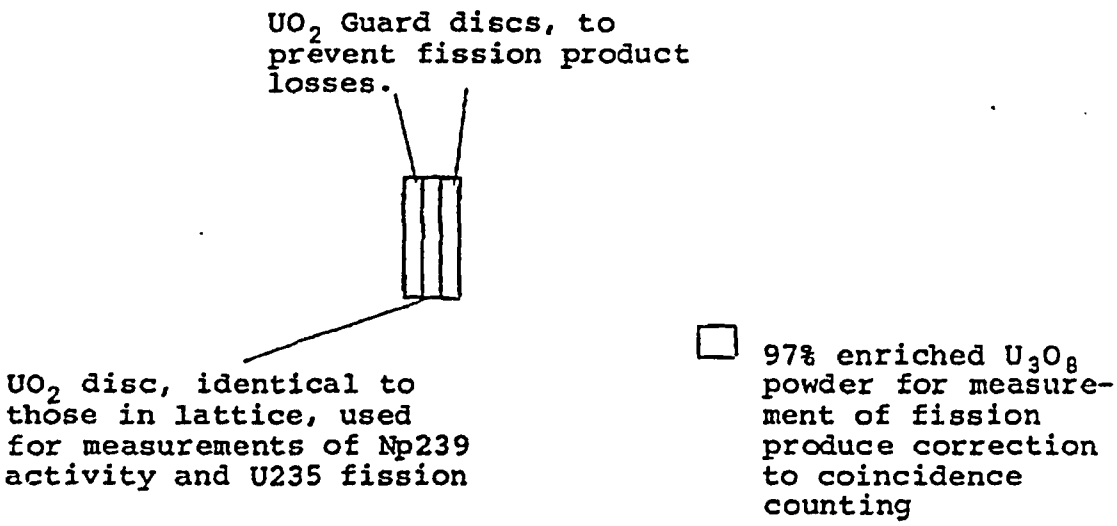
It will be seen the familiar ~3% discrepancy reappears when the old value of the Q(t) function is used.

5 CONCLUSIONS

A source of error was found in the coincidence technique which became significant when the enrichment of the fuel increased above 2%. Measurements with the corrected coincidence technique produced results that agreed with chemistry to better than 1%, ie comparable to the systematic error of the coincidence measurements.



RCR Lattice Pack of UO_2 Pellets and Foils



Thermal Column Pack

FIGURE 1 - TYPICAL FOIL PACKS USED IN IRRADIATIONS