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Isotope Correlations for Safeguards and  
Accountability of LWR Fuel Cycles

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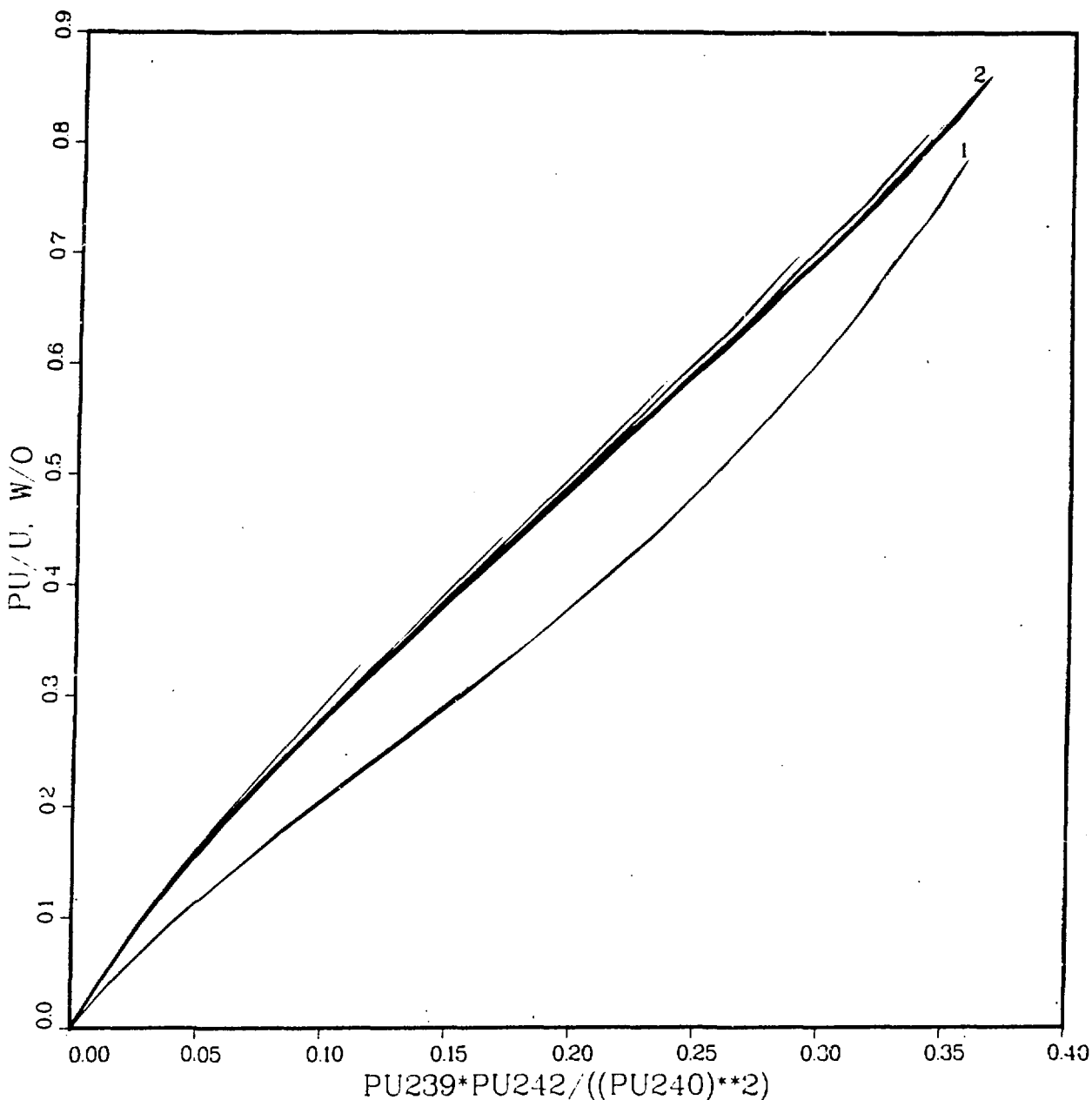
There exists a nuclear material accountancy gap of 4 or more years between the fabrication plant output and the reprocessing plant input. The closing of a material balance in any fuel cycle is of prime interest to safeguards and nuclear material accountability, both on the domestic and international level. The reactor is the source and sink of isotopes and consequently, in closing the material balance it is necessary to tie-in the input to the reprocessing plant to the reactor operations. There are two safeguards related questions that are to be addressed in the reprocessing plant to close the balance: a) What is the nuclear material content in the dissolver tank? and b) What should be the nuclear material content in the dissolver tank? The first question is addressed by the gravimetric method<sup>1</sup> involving isotopics and/or elemental assay. This is necessary but this is not sufficient to address the second question. The latter question is addressed by the isotopics and isotope correlations which relates to the reactor operations.

In this paper the two questions have been addressed in a comprehensive manner by the isotope correlation technique (ICT) study at the Argonne National Laboratory (ANL/ICT program). The results of the program include the following: (1) Twenty-nine heavy metal isotope correlations, some of which have been explored by other investigators<sup>2-5</sup>, were computed for each different fuel assembly of an operating (Zion-2) PWR using three-dimensional, two-dimensional and point reactor depletion calculations; the dependence of the computed correlations on the enrichment and the number of burnable poison rods in the assembly, and on the method of calculation has been studied (Fig. 1); the

isotope correlations obtained from three-dimensional depletion calculation have been compared with related and available measured reprocessing data for U, Pu isotopes for the Obrigheim PWR of the Federal Republic of Germany<sup>6</sup> (Fig. 2); (2) The sensitivity of these isotope correlations to anomalies in nuclear material flow (e.g., substitution of one unirradiated or natural uranium assembly for a spent assembly in a six assembly dissolver batch) and the sensitivity of the correlations to measurement errors of the dissolver solution isotopic composition have been studied (Fig. 3) and used in selecting some of the more effective correlations which were incorporated in an algorithm designed to determine the burnup and initial enrichment of the reprocessing batch for verification of the reported burnup and initial enrichment and to verify the internal consistency among the measured isotopic concentrations and the Pu/U ratio; (3) This cross-correlation algorithm has been programmed and executed on a completely self-contained portable ICT computer for infield implementation and assessment. For use after the integrity of the reprocessing batch has been verified by the cross-correlation algorithm, a modification of the current gravimetric method for measuring the plutonium and uranium content in the dissolver or accountancy tank has been obtained.

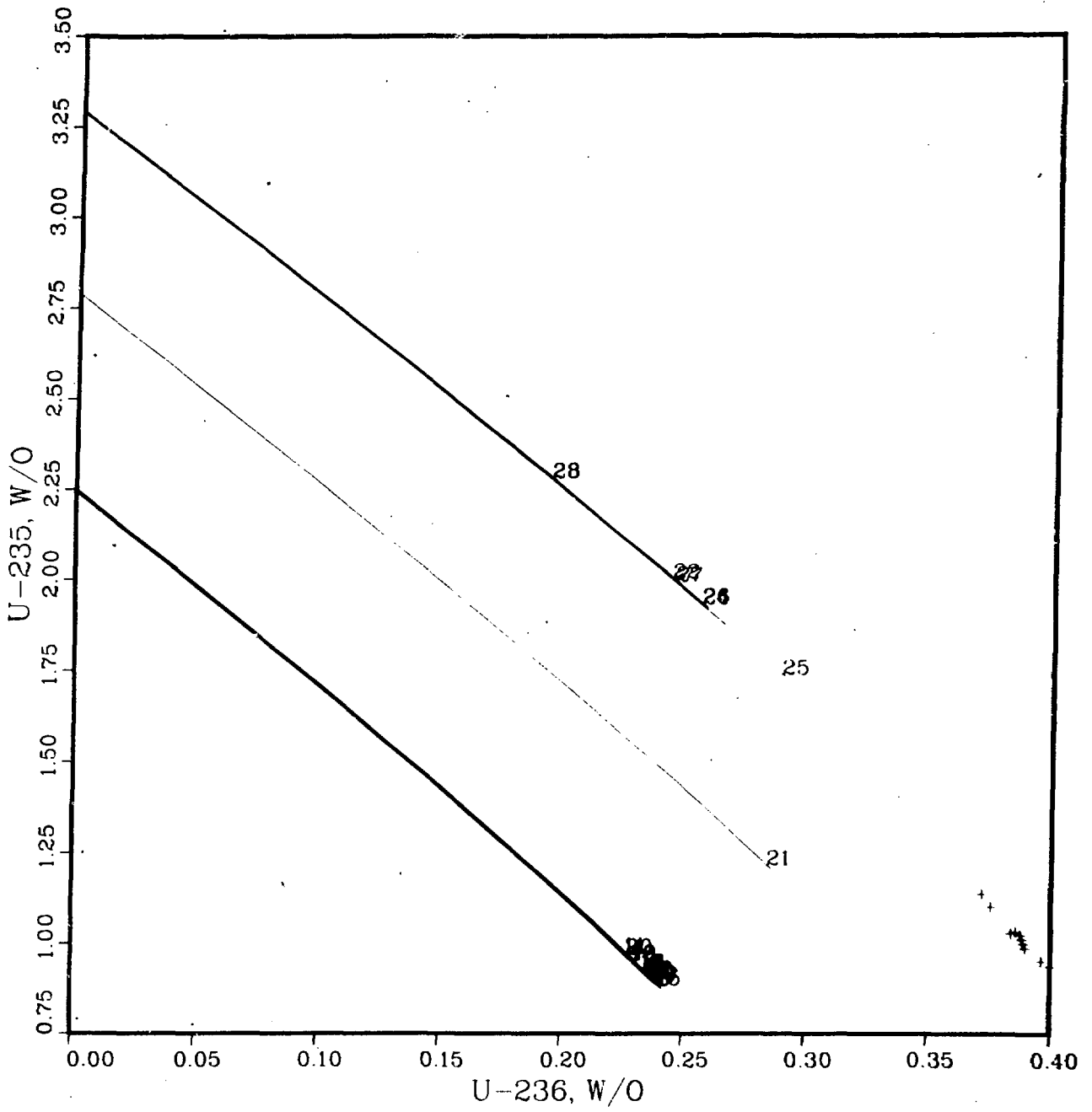
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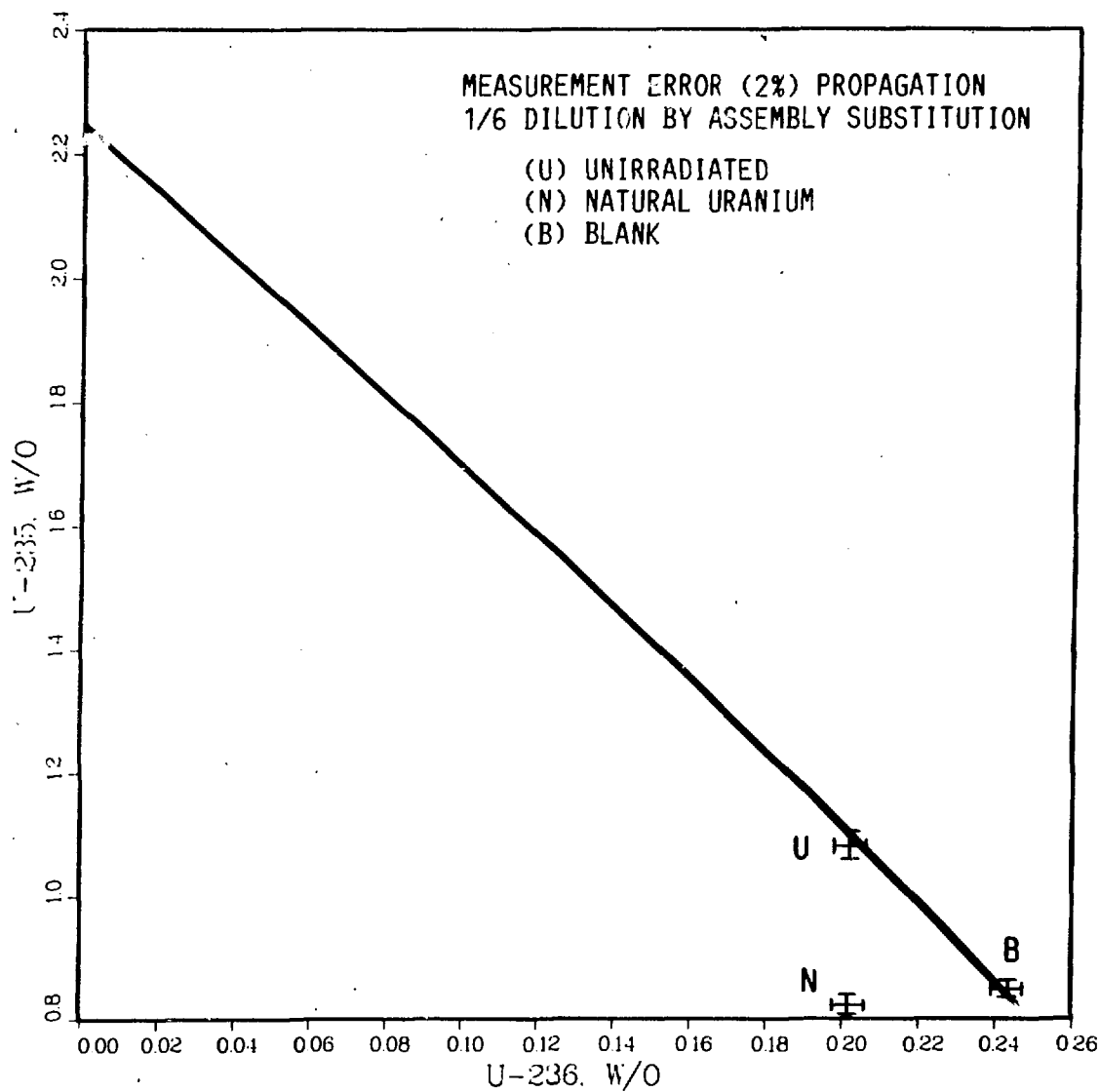
1 = EACH ASSEMBLY HOMOGENIZED OVER CORE HEIGHT (20 CURVES)  
 2 = THE 8 AXIAL SEGMENTS OF EACH ASSEMBLY (160 CURVES)

Fig. 1. Isotope Correlation No. 12 for Cycle 1 of Zion 2  
 for all Axial Segments of All Zone 1 Assemblies  
 (180 Curves, 3-D 2-Group Calculation)



+ Obrigheim Reprocessing Data for Assemblies of Enrichment 3.10 w/o.

Fig. 2. Isotope Correlation No. 3 for Cycle 1 of Zion 2 for all Assemblies having No BPR, Each Homogenized Over Core Height (28 Curves, 3-D 2-Group Calculation)



	X	% DIFF.	Y	% DIFF.
NO SUBST	$2.430 \times 10^{-1}$		$8.463 \times 10^{-1}$	
UNIRR SUB	$2.016 \times 10^{-1}$	$-1.704 \times 10^1$	$1.085 \times 10^0$	$2.822 \times 10^1$
NATUR SUB	$2.016 \times 10^{-1}$	$-1.704 \times 10^1$	$8.234 \times 10^{-1}$	$-2.704 \times 10^0$
BLANK SUB	$2.430 \times 10^{-1}$	0.000	$8.463 \times 10^{-1}$	0.000

Fig. 3. Measurement Error (2%) Propagation