Deutsch[6] determined the lifetime of *ortho*-positronium as observed in freon. He obtained

$$1/{}^3\tau = (6.8 \pm 0.7) \times 10^6 \text{ sec}^{-1}.$$

Therefore,

$$\sigma_{2k}/\sigma_{3k} = \tfrac{1}{3}\,{}^3\tau/{}^1\tau = 398 \pm 40,$$

which is in agreement with our value.

The theory of Ore and Powell for the three-photon annihilation of a positron-negatron pair has therefore been directly confirmed. The numerical results of Lifshitz and Ivanenko and Sokolov are definitely in disagreement with our experimental results.

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Fission Yield Fine Structure in the Mass Region 99–106*

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Fission yield measurements were carried out in the mass region 99 to 106 to investigate previously postulated fine structure related to nuclear closed shells. Radiochemical data were obtained in this mass region in the fission reactions $U^{235}(n,f)$, $U^{238}(\gamma,f)$, $U^{235}(d,f)$, and $U^{238}(d,f)$ for the yields of Mo^{99}, Mo^{101}, Mo^{102}, Ru^{103}, Ru^{105}, and Ru^{106}. Fine structure spikes were found in all reactions. It was found that the mass position of the spikes can be explained by the assumption that primary fission fragments with 82 neutrons or 50 protons and their complements have an enhanced yield as a result of selectivity in the fission process itself. Agreement was also found with two previously known reactions: $U^{233}(n,f)$ and Cm^{242} spontaneous fission. Fine structure is also predicted for several other fission reactions.

I. INTRODUCTION

SINCE the discovery of fission in 1939, a great deal of work has been done by nuclear chemists to investigate the problem of the distribution of mass between the fission fragments. It was found quite early that fission is asymmetric, and that the distribution of mass could be described,[1] at least approximately, by a smooth curve with two broad maxima. This fact has been verified many times, for many fission reactions, and although the shape and mass position of the yield-mass curve may change somewhat, all cases of low- and medium-energy fission seem to follow the same type of mass distribution.

Several significant departures from the smooth distribution curve have been discovered, however, and in certain mass regions the smooth curve is found to be totally inadequate. The earliest indication of anomalous yields was reported by Thode and Graham,[2] who found that the measured yields of Kr^{84} and Xe^{134} were about 35 percent too high to lie on any continuous smooth curve. Stanley and Katcoff[3] found the cumulative yield of I^{136} in the thermal-neutron fission of U^{235} to be 3.1 percent, while the next member of the same chain, Xe^{136}, whose independent fission yield was estimated[4] to be about 0.4 percent, had been found by Thode and Graham[2] to have a cumulative yield of 6.1 percent. This indicated a much higher independent yield for Xe^{136} than could easily be explained. Still another anomalous fission yield was found by Macnamara, Collins, and Thode,[5] in that the fission yield of Xe^{133} was found to be 6.3 percent instead of the expected 5.0 percent. Recent studies[6,7] have led to somewhat higher absolute values for the xenon fission yields.

These so-called fine-structure phenomena, as known up to that point, were explained satisfactorily by Glendenin.[8] He postulated that in all cases where a primary fission fragment contains one neutron in excess of a closed shell (i.e., contains 51 or 83 neutrons) this extra loosely bound neutron is immediately lost. This neutron loss, considered to be entirely independent of

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[1] See, for example, C. D. Coryell and N. Sugarman, editors, *Radiochemical Studies: The Fission Products* (McGraw-Hill Book Company, Inc., New York, 1951), National Nuclear Energy Series, Vol. 9, Div. IV.

[2] H. G. Thode and R. L. Graham, Can. J. Research **A25**, 1 (1947).

[3] C. W. Stanley and S. Katcoff, J. Chem. Phys. **17**, 653 (1949).

[4] Glendenin, Coryell, and Edwards, Paper 52 of reference 1.

[5] Macnamara, Collins, and Thode, Phys. Rev. **78**, 129 (1950).

[6] S. Katcoff and W. Rubinson, Phys. Rev. **91**, 1458 (1953).

[7] A. C. Pappas, Laboratory for Nuclear Science, Technical Report No. 63, Massachusetts Institute of Technology, September, 1953 (unpublished).

[8] L. E. Glendenin, Laboratory for Nuclear Science and Engineering, Technical Report No. 35, Massachusetts Institute of Technology, August, 1949 (unpublished).

the known prompt neutron emission, results in a decrease in yield at the mass of the shell-plus-one fragment, and an equal increase in yield at the mass of the closed-shell fragment. The high fission yields of masses 133 and 134, and the anomalous yields at mass 136 were thus explained semiquantitatively as the influence of the 82-neutron shell on neutron emission. It should be noted that although this postulate has been modified by Wiles,[9] who considered the amount of neutron loss to be a function of the neutron-proton ratio as compared with the same ratio for stable nuclei, and by Pappas,[7] who extended the postulate to the third, fifth, and seventh neutrons in excess of a closed shell, based on studies of shell effects in neutron-binding energies and the emission of neutrons from fission fragments, the underlying principle of neutron loss remains essentially as conceived by Glendenin.[8]

This shifting of mass yield, as explained by Glendenin, requires that a depression in fission yields occur at about masses 135–137, to balance the excess at masses 133 and 134. Wiles,[9] in a mass-spectrometer study of the fission-produced cesium isotopes of masses 133, 135, and 137, found that no such dip occurs. To explain this, it was proposed[9] that in addition to the neutron-loss mechanism of Glendenin, an additional effect occurs, namely, that primary fission fragments with a closed shell of 82 neutrons have a somewhat enhanced yield. This "extra" primary yield or selectivity was postulated to be some function of the expected unperturbed yield of that fragment, as calculated using the postulate of equal charge displacement.[4] Fission-yield calculations based on this proposal, and on a slightly modified form of Glendenin's original hypothesis, were found to fit the experimental data quite well from mass 132 to mass 137. Pappas[7] found good agreement between the predicted and the measured values in the mass region 130 to 150 except at masses 134 and 135, where the observed values were somewhat higher than expected. In order to account for this, he, too, found it necessary to include some enhanced yield for 82-neutron fragments, as well as a lower probability for neutron emission from these fragments.

It is a necessary consequence of this proposed selectivity in the fission process that the fragment which is complementary to the 82-neutron fragment must also have an enhanced yield. It was predicted by Wiles,[10] and at about the same time independently discovered by Glendenin, Steinberg, Inghram and Hess,[11] that a fine-structure spike occurs at mass 100 in the thermal-neutron fission of U^{235}. Primary selectivity expected for 50 neutrons would occur near masses 84–85 and 147–149 in the thermal-neutron fission of U^{235}, where the fission yields are low and changing rapidly with mass. Thus the effect will be harder to interpret. Primary selectivity for 50 protons would be expected near masses 129–131 and the complementary masses 103–105.

The present work is a study of this primary selectivity in the fission process, and of its relation to nuclear closed shells, $N=82$ and $Z=50$. The region of the complementary fine structure, i.e., masses 99–105, was chosen for this investigation to avoid the complications introduced by the superposition of the two types of fine structure that occur between masses 132 and 137. A study of the complementary fine structure is somewhat complicated by the uncertainty in the number of prompt neutrons lost by each fission fragment before it becomes an experimentally observable fission product. However, this was found not to be a very serious obstacle.

Fission yields can be determined by the mass-spectrometric method only for samples which have undergone very large amounts of fission. Radiochemical determinations permit study of a much greater variety of fission processes, but pains must be taken to compare β activities on an absolute basis. Gaps occur in the information where species are too short lived to be determined quantitatively, such as at mass numbers 96, 98, 100, and 104.

II. EXPERIMENTAL

Fission yields have been determined for 14.6-min Mo^{101} and 11.0-min Mo^{102} relative to 67-hr Mo^{99}, and for 67-hr Mo^{99}, 43-day Ru^{103}, 4.5-hr Rh^{105}, and 1.0-yr Ru^{106}, all relative to 12.8-day Ba^{140}. These yields were measured in the thermal-neutron fission of U^{235}, the 13-Mev photofission of U^{238}, and in the 15-Mev deuteron fission of U^{235} and U^{238}.

Irradiation Procedures

The thermal-neutron irradiations were done at the M.I.T. cyclotron, where neutrons are produced by the Be(d,n) reaction. The neutrons were thermalized by the 4-inch thick walls of a paraffin block which held the sample. The samples were irradiated either as solid UO_3 or as a solution of UO_2Cl_2 in 12M HCl.

For the photofission experiments, irradiations were made in the 13.5-Mev electron beam of the M.I.T. linear accelerator. The maximum energy of the electrons was 13.5 Mev, the most probable electron energy was 13.0 Mev. The electrons were stopped in a $\frac{1}{4}$-inch lead plate, placed immediately in front of the sample, which gave rise to a complete spectrum of bremsstrahlung with maximum energy just over 13 Mev. Irradiations were made on UO_3.

Deuteron irradiations were made in the external beam of the M.I.T. cyclotron. The energy of the deuterons striking the target was 15 Mev. Irradiations were made on natural UO_3 (U^{238}), and on nearly pure

[9] D. R. Wiles, M.Sc. thesis in chemistry, McMaster University, Hamilton, Ontario, Canada, October, 1950 (unpublished); Wiles, Smith, Horsley, and Thode, Can. J. Phys. **31**, 419 (1953).

[10] D. R. Wiles, Laboratory for Nuclear Science, Quarterly Report, Massachusetts Institute of Technology, August, 1951 (unpublished).

[11] Glendenin, Steinberg, Inghram, and Hess, Phys. Rev. **84**, 860 (1951).

U^{235}, in the form of U_3O_8, kindly loaned by the U. S. Atomic Energy Commission.

In some cases, the samples to be irradiated were so small (~10 mg) as to involve possible loss of fission fragments by recoil, or loss of fission-produced xenon by gaseous effusion. In such cases, the samples were tightly wrapped in several thicknesses (~12 mg/cm² total) of aluminum foil, and this foil was dissolved and processed with the sample.

Chemical Procedures

Molybdenum was separated from the fission mixture by extraction into diethyl ether from 6*N* HCl, after addition of Te^{IV}, Te^{VI}, and Mo^{VI} carriers. After back extraction into water, impurities such as Ga, Sb, Te, and other elements which are extracted slightly under these conditions were removed by scavenging with $Fe(OH)_3$. Technetium was found to follow the molybdenum almost completely up to this point, and rhenium was here added to act as hold-back carrier. The molybdenum was finally precipitated from the hot, buffered solution by 8-hydroxyquinoline (oxine). When the short-lived molybdenum isotopes were being studied, it was found that the separation could be done in about fifteen minutes. For this fast separation, in which only a small amount (~1 mg total weight) of molybdenum oxinate was separated, drying of the sample was done by washing the sample, on the filter paper, first with boiling water, then with dioxane, and finally with ether. The chemical yield was of no importance in these cases, because Mo^{99} was used as an internal standard for fission. The samples were mounted as the oxinate. Tracer tests showed that separation from technetium in this fast procedure is better than a factor of 200, and that separation from tellurium is a factor of 10^3 or better. Separation from all other fission products is expected to be much better than 10^3.

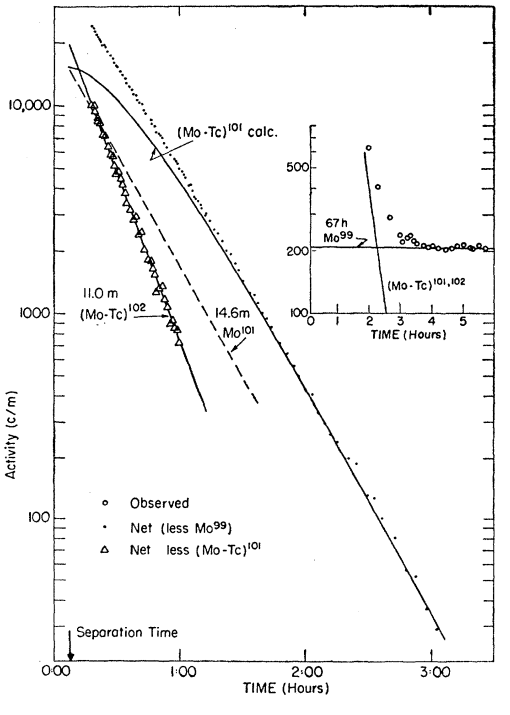

FIG. 1. Analysis of Mo-Tc decay curve.

When 67-hr Mo^{99} was being studied relative to Ba^{140}, the separation was done by the same method, but care was taken to get a stoichiometric compound, without coprecipitated reagent. Drying was done in an oven at 110° for fifteen minutes. The carrier was standardized by the same procedure.

Ruthenium separation was done by distillation of the tetraoxide with carrier from a perchloric acid solution of the fission mixture. Separation from technetium was done by precipitating the hydrous oxide, RuO_2, by reduction of the alkaline solution with alcohol. This precipitate was dissolved in HCl, and ruthenium metal was precipitated by reduction with powdered magnesium. The metal precipitate was filtered and washed with water, alcohol, and ether, and dried at 110° for ten minutes. When essentially weightless sources were needed, an aliquot, containing <20 micrograms of ruthenium, was evaporated to dryness on the source mounting. The chemical yield was determined colorimetrically by the thiourea method, using an identical aliquot.

Barium was separated by precipitation at 0°C as $BaCl_2 \cdot H_2O$ by HCl-ether reagent (1 part diethyl ether, 5 parts concentrated HCl). The precipitate was dissolved in water, and a ferric hydroxide scavenging was performed. The chloride precipitation was repeated twice, and the final precipitate was filtered and dried with alcohol, ether, and suction.

Beta Counting

The standard mounting for samples consisted of a 21-mm diameter filter paper disk (14.6 mg/cm²) onto which the sample was filtered, supported by Scotch Tape (8.5 mg/cm²), in a 1-inch hole in a $2\frac{3}{4}$-inch by $3\frac{1}{4}$-inch card. The sample was covered with a film of polystyrene (2.5 mg/cm²). For counting, this sample was supported by an aluminum shelf arrangement, about 2 cm below an end-window G. M. counter (Tracerlab TGC-1). The whole counting assembly was in an aluminum-lined lead shield with $1\frac{1}{2}$-inch walls.

In order to compensate for scattering and absorption of the beta radiation by the counting assembly, by the air gap, and by the sample itself, appropriate corrections were made. The corrections used were nearly identical with those of Pappas,[7] which are derived from the work of Zumwalt,[12] Engelkemeir *et al.*,[13] and

[12] L. R. Zumwalt, Unclassified Report MonC-397 (unpublished); Atomic Energy Commission Report AECU-507 (unpublished).

[13] Engelkemeir, Seiler, Steinberg, and Winsberg, Paper 4 of reference 1.

TABLE I. Decay data used. Values marked with an asterisk (*) are from references 9 and 14; all others are from reference 15.

Species	Half-life	β-energies (Mev)
Mo^{99}	67 hr	1.23, 80%; 0.45, 20%
Mo^{101}	14.6 min*	1.2*, 70%*; 2.2*, 30%*
Tc^{101}	14.3 min*	1.4*
Mo^{102}	11.0 min*	1.2*±0.3
Tc^{102}	<25 sec	~3.5*
Ru^{103}	43 days	0.217
Ru^{105}	4.5 hr	1.15
Rh^{105m}	45 sec	~0.11 (e^-)
Rh^{105}	36.5 hr	0.57
Ru^{106}	1.0 yr	0.04
Rh^{106}	30 sec	3.3 (weighted average)
Ba^{140}	12.8 days	1.02, 60%; 0.48, 40%
La^{140}	40.0 hr	1.32, 70%; 1.67, 20%; 2.3, 10%

Pappas.[7] Corrections could only be estimated for the equilibrium pair, 11.0-min Mo^{102}→<25-sec Tc^{102}, because of the uncertainty in the β energies. The error is felt to be small, however, because all of these samples were very thin (~1 mg/cm²), so that the self-scattering and self-absorption corrections were small and not strongly dependent on the beta energy. The counting correction factors (by which the observed counting rates were divided) for these thin molybdenum samples were 1.03, 1.06, and 1.04, respectively, for Mo^{99}, Mo^{101}, and Mo^{102}.

All counting rates were corrected for coincidence losses (determined as 1.2 percent per 100 counts/second), for chemical yield, for the size of aliquot used, and for the fraction of saturation activity produced in the irradiation.

In determinations of the fission yields of the short-lived molybdenum isotopes, counting of samples was begun 25-35 minutes after the end of the irradiation, and continued every minute for about two hours. After this time, counting was done every five minutes for 2–3 hours more, and then every fifteen minutes for several hours to establish the slope of the 67-hr Mo^{99}–6.0-hr Tc^{99m} growth-decay curve, and give the initial activity of the Mo^{99}. The long-lived molybdenum could then be subtracted from the gross activity. The tail of the remaining curve was a pure growth-decay curve of the 14.6-min Mo^{101}–14.3-min Tc^{101}.[14] A calculated curve for the total activity of this pair, computed using the calculated counting efficiency of Tc^{101} equal to 0.98 times that of the Mo^{101}, was fitted to the tail of the observed curve after subtraction of Mo^{99}. The upper end of this fitted curve gave the activity of the Mo^{101} at separation time. Subtraction of this computed curve from the observed points (less Mo^{99}) gave the decay curve of Mo^{102}–Tc^{102}. Considerable confidence in the precision of this method is given by the fact that the slope of this last decay curve was in every case found to be within ±0.3 minute of the half-life of 11.0 minutes found[14] for Mo^{102}. These successive subtractions are shown in Fig. 1 for a typical analysis.

[14] D. R. Wiles, Phys. Rev. **93**, 181 (1953); D. R. Wiles and C. D. Coryell (to be published).

Counting of Ru^{103}, which has a 217-kev β, was done from a <20-microgram sample, evaporated on a ~50 microgram/cm² film of Parlodion. A careful β absorption curve taken of this sample was extrapolated to zero absorber according to a 4.5-mg/cm² initial half-thickness, to give the total activity.

The half-lives and β energies used in this work are given in Table I.[9,14,15]

The half-lives of the zirconium and niobium isotopes of masses 101 and 102 are not known, so parent corrections could not be made at these masses. A careful estimate of the half-lives, using shell-specialized parameters[16,17] for the semiempirical mass formula, and the energy—half-life correlations of beta-decay theory, showed that Zr^{102} and Nb^{101} could have half-lives as long as 2 minutes, or as short as about one second. From this calculated maximum half-life, the maximum possible error can be calculated. In all experiments, separation times were such that the precursors would have completely decayed before separation. The possible error, then, would result only from failure to correct for production, by decay of the parent, of the desired species after the end of the irradiation. The error, which is always positive, can be derived directly from the standard decay laws, and amounts to

$$\text{Error}=\frac{T_{\frac{1}{2}}(\text{parent})}{T_{\frac{1}{2}}(\text{daughter})-T_{\frac{1}{2}}(\text{parent})}.$$

Using a parent half-life of 2 minutes, and assuming no independent formation of the daughter, the maximum error is computed to be 16 percent for Mo^{101} and 22 percent for Mo^{102}. This error is a constant, and is independent of the nature and the time of the irradiation. An appreciable independent yield of the daughter (which is very likely in both these cases) would decrease the error. It must be emphasized that these surprisingly large errors represent maxima only, and it is probable that the real errors are much smaller, perhaps negligible. (For example, the highly likely process of branched β^- decay would greatly decrease the calculated half-life and correspondingly reduce the error.)

TABLE II. Fission yields obtained in this work. Precision is expressed as standard deviation of the mean value. Italicized values were used as reference. Parentheses indicate values of doubtful validity.

Type	Mo^{99}	Mo^{101}	Mo^{102}	Ru^{103}	Ru^{105}	Ru^{106}	Ba^{140}
$U^{235}(n,f)$	5.9 ±0.2	5.4 ±0.2	4.1 ±0.2	1.4	0.83±0.2		*6.2*
$U^{238}(\gamma,f)$	6.8 ±1.0	6.5 ±1.0	5.0 ±0.8	(1.4)	3.15±0.3		*5.77*
$U^{238}(d,f)$	1.27±0.02	1.30±0.04	1.06±0.03	0.22	1.16±0.06	0.31	*1.00*
$U^{235}(d,f)$	1.42±0.09	1.27±0.12	0.86±0.05	0.44	0.90±0.04	0.60	*1.00*

[15] Hollander, Perlman, and Seaborg, Revs. Modern Phys. **25**, 469 (1953).

[16] Coryell, Brightsen, and Pappas, Phys. Rev. **85**, 732 (1952); and Laboratory for Nuclear Science Quarterly Report, Massachusetts Institute of Technology, August, 1952 (unpublished).

[17] C. D. Coryell, Ann. Revs. Nuclear Sci. **2**, 305 (1953).

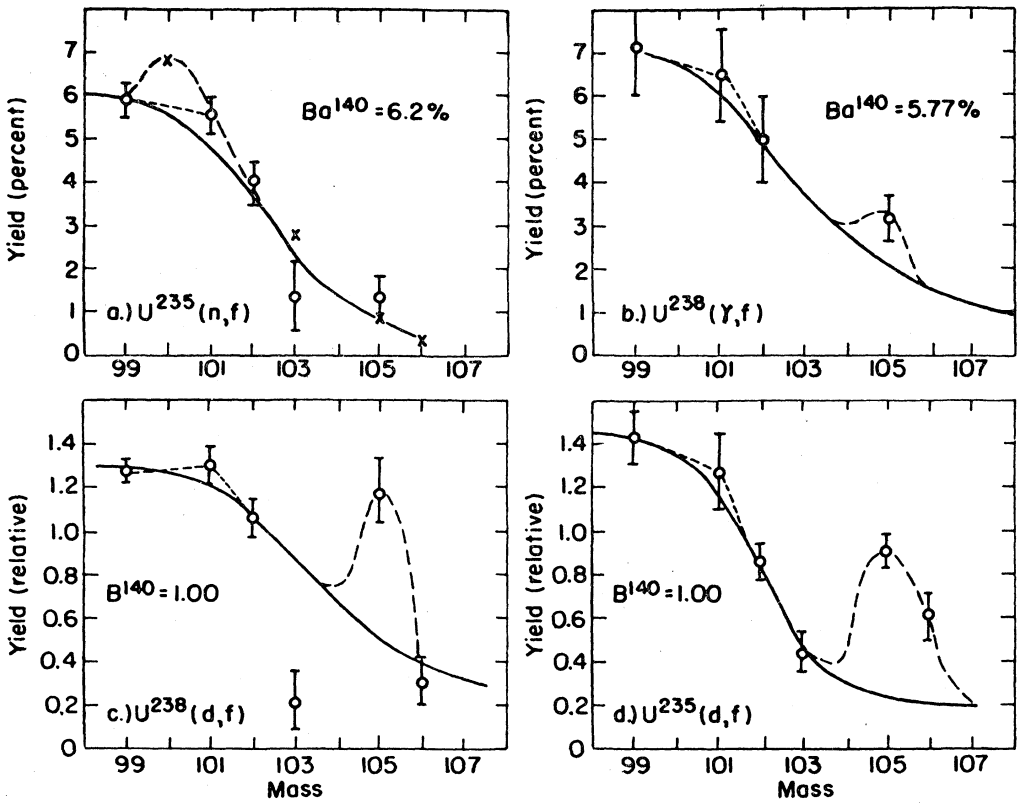

FIG. 2. Fission yields determined in this thesis. (o = this work; × = others.)

The fission yield values obtained in this work are given in Table II and are shown graphically in Fig. 2. The value of 6.2 percent is taken[1,7] for Ba^{140} in $U^{235}(n,f)$ and that of 5.77 percent is taken for Ba^{140} in $U^{238}(\gamma,f)$ according to Richter.[18] The fission yields for $U^{238}(d,f)$ and $U^{235}(d,f)$ are compared to Ba^{140} as unity. The value for Mo^{99} in $U^{235}(n,f)$ is in good agreement with the absolute value, 6.14±0.16 percent, reported by Terrill *et al.*[19] The value for Ru^{103} in $U^{235}(n,f)$ is less reliable than that of Hardwick[20] (2.85 percent), and little weight can be given to the values for Ru^{103} and Ru^{106} in Table II and Fig. 2.

In Fig. 2, the observed yields are shown as open circles, and the values given by Glendenin *et al.*[11] and by Hardwick[20] as crosses. The points for Mo^{99}, Mo^{101}, and Mo^{102} are joined by a dotted line to indicate that the relative values for the fission yields of these isotopes are known with greater precision than the absolute values, because of having been determined relative to Mo^{99}. The solid line is a proposed hypothetical "smooth curve" of fission yields, drawn with some personal prejudice. The dashed line indicates the observed fission yield curve. The difference between the observed fission yields (dashed line) and the smooth curve (solid line) is considered to be the "fine-structure yield." The magnitude of this fine-structure yield is of only qualitative interest, because of the uncertainty in the position of the smooth curve. The mass location of the fine structure, on the other hand, is felt to be quite reliable. There may be some doubt as to whether the fine structure indicated at mass 101 in parts (b), (c), and (d) of Fig. 2 is real, or whether it might be an error resulting from the parent correction discussed earlier. Moreover, the minimum shown in the dashed curve of Fig. 2b [$U^{238}(\gamma,f)$] is perhaps exaggerated. This, however, is not very important to the following discussion of fine structure.

Two other cases of complementary fine structure have been reported—in the thermal-neutron fission reaction $U^{233}(n,f)$, a fine-structure spike occurs[21] at mass 99, and in the spontaneous fission reaction $Cm^{242}(f)$, a fine-structure peak is found[22] at masses 105–106.

[18] H. G. Richter, Ph.D. thesis in Chemistry, Massachusetts Institute of Technology, August, 1952 (unpublished); H. G. Richter and C. D. Coryell, Phys. Rev. **95**, 1550 (1954).

[19] Terrill, Scott, Gilmore, and Minkkina, Phys. Rev. **92**, 1091 (1953).

[20] W. H. Hardwick, Phys. Rev. **92**, 1072 (1953).

[21] Steinberg, Glendenin, Inghram, and Hayden, Phys. Rev. **95**, 867 (1954).

[22] E. P. Steinberg and L. E. Glendenin, Phys. Rev. **95**, 431 (1954).

III. DISCUSSION

In an attempt to interpret the nature of this fine-structure effect, the following assumptions were made:

(1) The fine structure observed in the mass 99–106 region is caused by a primary selectivity in the fission process which favors[9] certain nuclei having a closed shell of 82 neutrons.

(2) It is assumed that those closed-shell nuclei are favored for which a high independent yield is predicted by the postulate of equal charge displacement.[4,7] The observations would thus not show up the yield increases for closed-shell nuclei of low smooth-curve yields.

(3) It is assumed that on an average, the fragment complementary to the closed-shell fragment will lose 2 prompt neutrons.

(4) A similar treatment should be applicable to the 50-proton shell.

Calculation of the expected unperturbed yield, as required by the second assumption, was done according to the method of Pappas,[7] based on the principle of equal charge displacement from stability for the most probable member of fission chains. The effect of shells on the variation of most stable charge, Z_p, with mass number, A, is included in Pappas' work.[7]

TABLE III. Closed-shell nuclei having high independent yields in fission, and their complements.

	$N=82$ Closed shell			$Z=50$ Closed shell		
		Complements			Complements	
Type of fission	Favored nuclei	Before neutron loss	After neutron loss	Favored nuclei	Before neutron loss	After neutron loss
$Th^{232}(\gamma,f)$	Sn^{132}	Zr^{100}	Zr^{98}	Sn^{130}	Zr^{102}	Zr^{100}
	Sb^{133}	Y^{99}	Y^{97}	Sn^{131}	Zr^{101}	Zr^{99}
				Sn^{132}	Zr^{100}	Zr^{98}
$Th^{232}(n,f)$	Sn^{132}	Zr^{101}	Zr^{99}	Sn^{131}	Zr^{102}	Zr^{100}
	Sb^{133}	Y^{100}	Y^{98}	Sb^{132}	Zr^{101}	Zr^{99}
$U^{233}(n,f)$	Sb^{133}	Nb^{101}	Nb^{99}	Sn^{129}	Mo^{105}	Mo^{103}
	Te^{134}	Zr^{100}	Zr^{98}	Sn^{130}	Mo^{104}	Mo^{102}
$U^{235}(n,f)$	Sb^{133}	Nb^{103}	Nb^{101}	Sn^{130}	Mo^{106}	Mo^{104}
	Te^{134}	Zr^{102}	Zr^{100}	Sn^{131}	Mo^{105}	Mo^{103}
$U^{238}(\gamma,f)$	Sn^{132}	Mo^{106}	Mo^{104}	Sn^{131}	Mo^{107}	Mo^{105}
and	Sb^{133}	Nb^{105}	Nb^{103}	Sn^{132}	Mo^{106}	Mo^{104}
$U^{238}(f)$	Te^{134}	Zr^{104}	Zr^{102}			
$U^{233}(d,f)$	Sb^{133}	Mo^{102}	Mo^{100}	Sn^{128}	Nb^{107}	Nb^{105}
	Te^{134}	Nb^{101}	Nb^{99}	Sn^{129}	Nb^{106}	Nb^{104}
	I^{135}	Zr^{100}	Zr^{98}			
$U^{235}(d,f)$	Sb^{133}	Mo^{104}	Mo^{102}	Sn^{129}	Tc^{108}	Tc^{106}
	Te^{134}	Nb^{103}	Nb^{101}	Sn^{130}	Tc^{107}	Tc^{105}
				Sn^{131}	Tc^{106}	Tc^{104}
$U^{238}(d,f)$	Sn^{132}	Tc^{108}	Tc^{106}	Sn^{131}	Tc^{109}	Tc^{107}
	Sb^{133}	Mo^{107}	Mo^{105}	Sn^{132}	Tc^{108}	Tc^{106}
$Pu^{239}(n,f)$	Sb^{133}	Tc^{107}	Tc^{105}	Sn^{129}	Ru^{111}	Ru^{109}
	Te^{134}	Mo^{106}	Mo^{104}	Sn^{130}	Ru^{110}	Ru^{108}
				Sn^{131}	Ru^{109}	Ru^{107}
$Cm^{242}(f)$	Te^{134}	Ru^{108}	Ru^{106}	Sn^{129}	Pd^{113}	Pd^{111}
	I^{135}	Tc^{107}	Tc^{105}	Sn^{130}	Pd^{112}	Pd^{110}

The distribution of yield along the chain is the same as that first proposed by Glendenin *et al.*[4] This calculation has been performed for all cases for which there are fine-structure data available for comparison. The predictions of favored nuclei under these assumptions are given in Table III.

It is evident that the predicted mass location of the complementary fine structure is dependent on the number, ν, of prompt neutrons emitted by the complementary fragment. The number $\nu=2$ was chosen partly for the sake of convenience, and is quite likely not accurate. There is evidence[7] that the total number of prompt neutrons emitted in near-symmetric fission may be as low as 1.4. The region of present study is further from symmetrical fission, where ν is believed to be considerably larger. The number of neutrons lost by the complementary fragment must probably be between 1.5 and 2.0.

Table IV gives the comparison of the mass numbers observed for fission spikes with those predicted as in Table III. We are grateful to Dr. L. E. Glendenin and Dr. E. P. Steinberg for advance communication of some of their mass-spectrometric yield data[21] for $U^{233}(n,f)$ and radiochemical yield data[22] for $Cm^{242}(f)$. The remaining identifications of fine structure spikes come from Fig. 2. The fission yield of Mo^{100} in thermal-neutron fission of U^{235} is from reference 11.

It is to be noted that the experimental data reported in Table IV encompass spreads of 4 charge units and 5 neutrons in nuclei undergoing fission, and in addition a broad spread of excitation energies from spontaneous fission through fission induced by 15-Mev deuterons. There is a fairly good correlation between observed masses of fine-structure spikes and those predicted from the assumption of a primary selectivity of closed-shell structures when the mass numbers for closed-shell

TABLE IV. Comparison of observed and calculated complementary fine structure.

Type of fission	Mass location of predicted fine structure		Mass of observed fine structure
	$N=82$ shell	$Z=50$ shell	
$Th^{232}(\gamma,f)$	97–98	98–100	···
$Th^{232}(n,f)$	98–99	99–100	···
$U^{233}(n,f)$	98–99	103[a]	···
$U^{235}(n,f)$	100–101	104[a]	100–101
$U^{238}(\gamma,f)$			101[b]
	102–104	105	105
$U^{238}(f)$	102–104	105[a]	···
$U^{233}(d,f)$	98–100	104–105	···
$U^{235}(d,f)$	101–102		101[b]
		104–106	105–106
$U^{238}(d,f)$			101[b]
	105–106	106–107	105
$Pu^{239}(n,f)$	104–105	107–109[a]	···
$Cm^{242}(f)$	105–106	110–111[a]	···

[a] Fine-structure effects are expected to be quite small here because of low yield.
[b] The apparent fine structure at this mass is small.

effects occur in regions of substantial smooth-curve yields. In $U^{235}(n,f)$, the effect at mass 100 (mass spectrometric yield determination) is more prominent than that at mass 101. In the next two entries in Table IV, the mass 101 effect seems to be small, but for these and $U^{238}(d,f)$ mass 100 data are not available. The rapid fall of yields for thermal-neutron fission and spontaneous fission on approaching symmetrical fission[1,22] tends to minimize effects of the 50-proton shell.

Attempts have been made to define an empirical yield enhancement curve for closed-shell species which would be a function of smooth-curve independent yields. Such a treatment serves to support the correlations of Table IV, but is at present on too speculative a basis for detailed confidence. The accumulation of more experimental data on independent fission yields in selected cases should help ground such treatment. Treatment at the level presented in this paper predicts a relatively large separation in the $U^{233}(d,f)$ curve for the spikes complementary to $N=82$ (masses 97–99) and complementary to $Z=50$ (masses 103–105).

The statistical theory of Fong[23] provides one mechanism for selectivity of closed-shell species in the fission process. The enhanced stability of closed-shell configuration in one of the two fragments implies greater internal energy in the fission mode, thus giving higher statistical weights and fission yields for these modes. This enhancement should be slowly diluted out with increasing energy in the fission process.

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γ Rays from the $C^{13}(p,\gamma)N^{14}$ and $Na^{23}(p,\gamma)Mg^{24}$ Reactions

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The γ rays from the 554-kev resonance of $C^{13}(p,\gamma)N^{14}$ and the 310-kev resonance of $Na^{23}(p,\gamma)Mg^{24}$ have been measured with a three-crystal pair spectrometer. Energies and intensities were measured and cascades detected by coincidence measurements. The 5.7-Mev γ-ray from $C^{13}(p,\gamma)N^{14}$ is 0.7 ± 0.6 percent of the intensity of the main 8.05-Mev radiation. The most intense radiation from $Na^{23}(p,\gamma)Mg^{24}$ was the 1.38-Mev line, and no 2.76-Mev radiation was observed.

THE three-crystal pair spectrometer[1,2] is well suited to measure weak γ rays in the presence of more intense radiation of higher energy. We have used it to investigate the γ rays from the 554-kev resonance of the $C^{13}(p,\gamma)N^{14}$ and the 310-kev resonance of $Na^{23}(p,\gamma)$-Mg^{24} reactions.

$C^{13}(p,\gamma)N^{14}$

The pair pulse-height energy distribution curve is shown in Fig. 1. The energy calibration is based upon RdTh (2.62 Mev) and Po—Be (4.45 Mev) and the assumption that the main component has an energy of 8.05 Mev.[3] The intensities of the lines, obtained from the pair cross section values of Mann, Meyerhof, and West,[4] was found to be:

Energy (Mev)	8.05	5.7 ± 0.15	4.05 ± 0.05	2.35 ± 0.02	1.66 ± 0.03
Relative intensity	100	0.7 ± 0.6	12.5 ± 2	15 ± 5	17 ± 4.

The shape of the pair peak was estimated for each energy. The asymmetrical shape of the peaks arises partly because of bremsstrahlung and electron escape and partly because all the triple coincidences do not correspond to pair formation in the central crystal. It is possible to obtain a triple coincidence when the Compton-scattered γ ray is captured in one side crystal and the bremsstrahlung from the Compton electron is captured in the other. The assumptions made in calculating the shape of pair spectrum peaks were checked by comparison with the measured shape of the known γ rays from the $F^{19}(p,\gamma)O^{16}$ and the $B^{11}(p,\gamma)C^{12}$ reac-

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