Independent and cumulative yields of very neutron-rich nuclei in 20 MeV p- and 18–41 MeV d-induced fission of ^{238}U

M. Leino,[1] P. P. Jauho,[2] J. Äystö,[2] P. Decrock,[3] P. Dendooven,[3] K. Eskola,[1]
M. Huyse,[3] A. Jokinen,[2] J. M. Parmonen,[2] H. Penttilä,[2] G. Reusen,[3] P. Taskinen,[2] P. Van Duppen,[3]
and J. Wauters[3]

[1]*Department of Physics, University of Helsinki, SF-00170 Helsinki, Finland*
[2]*Department of Physics, University of Jyväskylä, SF-40100 Jyväskylä, Finland*
[3]*Instituut voor Kern- en Stralingsfysika, Katholieke Universiteit Leuven, B-3001 Leuven, Belgium*

(Received 19 March 1991)

Independent and cumulative yields in nearly symmetric fission have been measured for the first time by bombarding ^{238}U with 20-MeV protons and with 18-, 25-, and 41-MeV deuterons. Isobaric charge dispersion has been determined in the $A=110$, $A=112$, and $A=114$ mass chains and, from asymmetric fission, in the $A=80$ mass chain. The average isobaric charges in the $p+$U reaction were 44.01±0.20 ($A=110$), 45.15±0.15 ($A=112$), and 45.74±0.15 ($A=114$). The widths (standard deviations) of the charge dispersion curves were 0.82±0.08 ($A=112$) and 0.54±0.08 ($A=114$). The average charges are roughly 0.3 unit higher than those calculated on the basis of the unchanged charge distribution model. The ground-state branch in the beta decay of ^{80}Ge was determined to be 65%. Altogether some 15 new neutron-rich isotopes have been found, the newest one being ^{114}Ru with a half-life of 0.53±0.06 s. An ion guide facility was used to separate the isobaric chains on-line from other fission products. Typical cumulative yields in a mass chain were on the order of 1000 atoms/μC, but for new isotopes independent yields as low as a few atoms/μC could be observed.

I. INTRODUCTION

Spontaneous or induced fission of a heavy nucleus leads to the production of typically a few hundred different nuclides and is still a useful reaction for the study of new neutron-rich isotopes [1–3]. For a given mass split, the yield of different elements in an isobaric chain is influenced by the reaction Q values in such a way that only a few nuclides are observed in a given chain. The isobaric charge variance $\sigma^2_{Z(A)}$ is usually less than 1.0 in fission [4,5], also when induced by heavy ions [6]. In contrast, the isotopic mass variance $\sigma^2_{A(Z)}$ is larger by typically a factor of ≈ 5. The overall mass distribution is much wider and shows well-known structure caused by nuclear shells and depending on the fissioning system [7].

The measurement of independent fission yields $Y(Z,A)$ of the nuclides ^AZ allows the determination of the charge dispersion. Mass yield can be studied in a simpler way by determining the cumulative yields in isobaric chains. Extensive studies of thermal-neutron-induced and spontaneous fission yields have been made using, for example, radiochemical techniques [8] and on-line recoil spectrometers [4]. The study of the width of the charge distribution and of the odd-even effects observed in the yield curves has recently received much attention due to the improved experimental methods available at present [9]. In particular, the effects of excitation energy and fissility parameter Z^2/A of the compound system on the yields are of interest.

Analysis of the yields of fission products provides information on how shell structure and pairing correlations [4,7] influence the fission process. Systematic knowledge of the yields of very neutron-rich isotopes has, in addition, great practical value in the study of exotic nuclei. In particular, it is of interest to find out whether a given isotope has the highest effective yield in spontaneous fission or fission induced by light particles or heavy ions. This will be discussed briefly in Sec. IV C on the basis of the results of the present work.

From thermal-neutron-induced fission and spontaneous fission one cannot reliably determine charge-dispersion parameters for symmetric products because of their low production rates. When more energy is brought into the system, for instance, by charged particles, the symmetric component in the mass distribution is enhanced [7]. In heavy-ion-induced reactions the mass and charge distributions may be further modified by deep inelastic and transfer reactions competing with complete fusion followed by fission [10,11].

In this work, charge dispersion was studied for the first time in symmetric or nearly symmetric fission. Independent yields have previously not been determined for symmetric low-energy fission, although mass yields around symmetry have been published [5] for thermal-neutron-induced fission of some nuclides such as ^{235}U. This lack of experimental data is a result not only of small fission yields but also of the short half-lives of very neutron-rich fission products. Even fast on-line radiochemical separation methods are too slow when the half-life is in the region of seconds. On the other hand, use of on-line recoil spectrometers such as LOHENGRIN at Institut Laue-Langevin (ILL) requires an extensive experimental effort

due to the low efficiency and the need to integrate over charge state and energy distributions of the products [4]. The work described here was performed with on-line mass separators based on the ion guide [ion guide isotope separator on-line (IGISOL)] principle [12,13] which have a delay time of some milliseconds and efficiencies on the order of 0.1%.

Our measurements were performed in connection with studies on beta-decay properties and nuclear structure of very neutron-rich nuclei in the mass $A = 110-120$ region. Among the newest isotopes studied are ^{109}Tc, ^{112}Tc, ^{114}Ru, ^{117}Rh, ^{119}Pd, and ^{120}Pd (Refs. [14–19], respectively). Our new information concerning the decay schemes of these and other neutron-rich isotopes has made it possible to determine the charge distribution for some mass chains as well as cumulative isotopic yields for certain elements.

II. EXPERIMENT

The IGISOL (ion guide isotope separator on-line) facility [12,13] at the Department of Physics, University of Jyväskylä, was used to separate the wanted mass chain from other products from fission of ^{238}U induced by 20-MeV protons. The proton beam current from the MC-20 cyclotron was typically 500 nA and the intensity of the mass-separated beam was usually on the order of 1000 atoms/s. The separation of fission products in IGISOL is based on thermalization of the products as singly charged ions in helium gas. The ions are then guided by gas flow and an electric field directly to an ISOLDE II type isotope separator. The operation of IGISOL is practically independent of the chemical properties of nuclides and it can be assumed that the efficiency of IGISOL is the same for all elements in an isobaric chain. The problem associated with the formation of oxides will be discussed briefly in Sec. III. The mass-resolving power of the separator is around 400 under normal operating conditions. This separation method is particularly well suited for the study of the elements Zr, Nb, Mo, Tc, Ru, and Rh, which are difficult to separate using conventional mass separators with an on-line ion source. In Louvain-la-Neuve, the similar LIGISOL [20] facility operating at the heavy-ion accelerator CYCLONE was used in the study of deuteron-induced fission of ^{238}U.

A. Double chamber ion guide for fission studies

A new recoil chamber [13] has been designed specifically for studies of products from fission of uranium induced by low-energy ($\sim$20-MeV) protons. In the construction of this chamber, use has been made of the nearly isotropic distribution of fission fragments. The new fission chamber is shown in Fig. 1. A similar chamber was used in the LIGISOL experiments.

In the new design, fragments are stopped in a plasma-free subchamber, which is separated from the beam and four 15-mg/cm^2 metallic ^{nat}U targets by a 0.9-mg/cm^2 Ni foil. The purity of the U targets was 99.9%. The presence of plasma produced by the primary beam in the

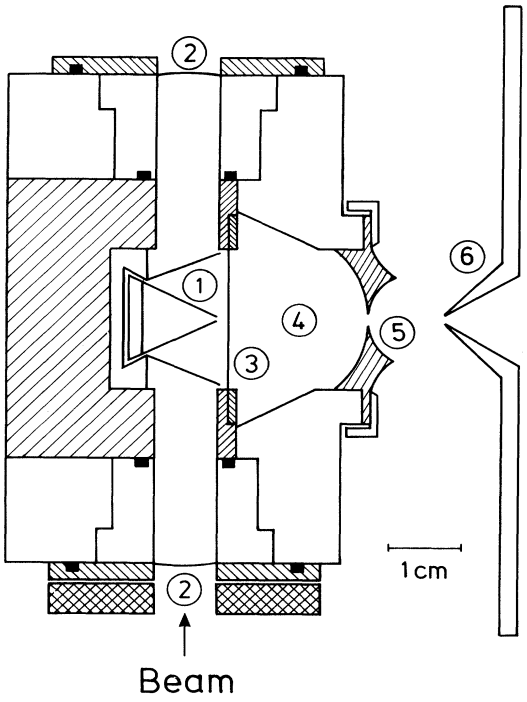

FIG. 1. Cutaway view of the double-chamber fission ion guide. To optimize the yield the four uranium targets are mounted so that the angle between the beam and the plane of the target is 20–25°. A nickel foil prevents the plasma produced by the beam from extending into the stopping chamber. Helium enters from the left and exits with the thermalized ions to the right. (1) Uranium targets, (2) beam windows, (3) nickel foil, (4) stopping chamber, (5) exit aperture, (6) skimmer electrode.

stopping gas increases recombination losses of the thermalized fragments. The separating foil does not completely eliminate recombination since the fragments also ionize the stopping gas. The effect of ionization, however, is negligible with the fragment yields which we observe in our experiments [13]. There is some elemental variation in the stopping efficiency due to differences in average kinetic energies and average charge states. This effect is not expected to affect our results significantly since yields of elements with nearly equal proton numbers are compared.

The stopping volume is about 8 cm^3 and it is filled with He at a pressure of typically 10 kPa. The gas pressure is limited by pumping capacity. The chamber has to be evacuated in a short time in order to minimize diffusion and recombination losses. A drawback of the present system is inefficient stopping of fission fragments in the He gas. Stopping efficiency is on the order of 1%. The main cause for loss of thermalized ions is diffusion to the walls of the stopping chamber. Further losses are caused by geometry and extraction efficiency. Total efficiency for collecting fission products is on the order of 10^{-3}. Typical delay times are on the order of a few milliseconds and decay losses during thermalization and separation

are insignificant even for the most neutron-rich nuclides studied in the present work.

B. Determination of cumulative and independent fission yields

In order to determine independent yields of the nuclides in a mass chain separated by IGISOL one has to know in detail the beta decay and level schemes of the nuclear species. Conventional nuclear spectroscopic studies in Jyväskylä were performed using 20% and 25% Ge detectors and a 1.4-cm^3 planar Ge detector for γ-ray and x ray singles and coincidence measurements, a 1.0-mm plastic transmission scintillator for β-γ coincidence measurements, a plastic telescope for beta end-point energy measurements, and a conventional conversion electron measurement system. In some cases, the novel magnetic conversion electron spectrometer ELLI [21] (electron lens for IGISOL) was used in the determination of internal conversion coefficients. The separated nuclei were implanted on a collection tape which was moved periodically to reduce the buildup of long-lived activities. In most cases, the cyclotron beam was turned on and off in synchronization with the tape movements to facilitate half-life measurements. In Louvain-la-Neuve, a 70% Ge detector was used in coincidence with a 1.0-mm-thick plastic beta transmission scintillator.

A major problem in finding out independent as well as cumulative yields is the determination of the intensity of the ground-state branch in the beta decay of a nuclide. The method used to solve the problem varied from nuclide to nuclide. In some cases nuclear-structure considerations exclude the existence of a strong ground-state branch. For example, from spin and parity systematics and from beta-decay properties of even Tc isotopes it was concluded that the spin of the ground state of ^{110}Tc is ≥ 3 so that beta decay to the 0^+ ground state of ^{110}Ru is forbidden. On the other hand, in the case of fast Gamow-Teller beta decays from 0^+ parent nuclides, one of the strongly populated 1^+ daughter states could be the ground state. A careful study of the whole isobaric decay chain is then necessary to determine the intensity of this branch. As an example, the decay of ^{110}Ru will be discussed in detail in Sec. III. In some cases, the sum beta spectrum of all the members in a mass chain was decomposed on the basis of the half-lives and other known data to extract the ground-state branches. The use of this method is sometimes hampered by the presence of beta-decaying impurity activities in the form of oxides.

The determination of cumulative isotopic yields benefits from stable operation of the IGISOL which has been achieved by use of the double fission chamber described above. The total beam dose was determined by integrating the beam current downstream from the target chamber. This method may introduce a systematic error, in which case all absolute yields given in the following are too small by a constant factor. Cumulative yields were determined from long continuous measurements in which most activities had reached saturation. The collection tape was moved only when changing the mass to be separated.

III. RESULTS

In the following, cumulative isotopic yields in saturation as well as independent isobaric yields and charge-dispersion parameters will be given. Details of our spectroscopic studies can be found elsewhere [1,15,22,23]. When there are two isomeric states for one nuclide, yields of each isomer were added together. The total thickness of uranium target material was 60 mg/cm^2 in the Jyväskylä experiments and 50 mg/cm^2 in the Louvain-la-Neuve experiments, and so the results should be considered averaged over the spread in proton or deuteron energy.

A. Cumulative isotopic yields of neutron-rich Tc, Ru, Rh, Pd, and Ag isotopes

The cumulative yields in saturation of $_{43}$Tc, $_{44}$Ru, $_{45}$Rh, $_{46}$Pd, and $_{47}$Ag isotopes from the 20-MeV proton-induced fission of ^{238}U are shown in Fig. 2. In most cases, the error caused by efficiency calibration and statistics is $\leq 10\%$. However, there is some variation in the IGISOL efficiency caused for example by different locations of the beam spot on the target in different experiments. This leads to varying stopping and collection efficiencies. A typical resulting uncertainty of $\pm 50\%$ is marked on some data points in Fig. 2. Another possible source of error is in the determination of the ground-state beta branch. When it was not known, zero branching was assumed. This gives lower yield limits, as shown in Fig. 2. The cumulative yield for the most neutron-rich isotopes decreases by a factor of ~ 5 per unit mass number. The lowest yields, a few atoms/μC, observed in our studies are those of ^{117}Rh [17] and ^{120}Pd [19]. If approximately 2.5 neutrons are emitted from each fragment in symmetric fission (Sec. IV A), the corresponding products have mass numbers around 116–117. From Fig. 2 it can be seen that mass yields stay nearly constant and may even decrease slightly with decreasing A in the range studied.

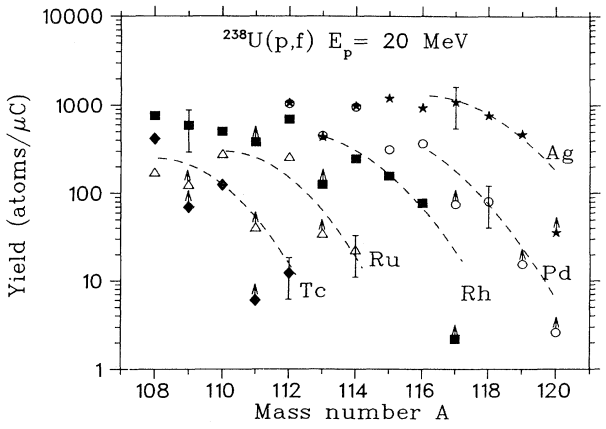

FIG. 2. Cumulative yields in saturation of Tc, Ru, Rh, Pd, and Ag isotopes. The dashed lines are drawn to guide the eye. Lower limits are given when the ground-state beta-decay branch is unknown, in which case zero branching was assumed.

B. Independent $A=110$, 112, and 114 nuclide yields from $p+^{238}$U and $d+^{238}$U

In the following, the determination of independent yields in the $A=110$, $A=112$, and $A=114$ chains will be described. As an example, the $A=110$ chain will be discussed in some detail while only an outline of the analysis will be given for $A=112$ and 114. The results of our analysis for the three mass chains from the 20-MeV $p+^{238}$U reaction studied in Jyväskylä are given in Fig. 5 (Sec. IV). In Fig. 3 the independent yields determined at $A=110$, 112, and 114 using the reaction $d+^{238}$U are shown. The bombarding energies were 18, 25, and 41 MeV.

1. The A=110 mass chain

The short half-life [15], 0.92±0.03 s, of ^{110}Tc allowed a straightforward determination of its production rate in saturation by means of the strong $2^+\rightarrow 0^+$ 241-keV γ transition in ^{110}Ru. As mentioned in Sec. II, zero beta branching to the ground state of ^{110}Ru was assumed. The absolute intensity of this transition was measured in our decay scheme study [15]. No feeding from ^{110}Mo decays was assumed. However, we did observe two coincident gamma transitions with a ~0.2-s half-life which might result from beta decay of the new isotope ^{110}Mo. The nominal yield was 2 orders of magnitude lower than the yield of ^{110}Tc. Such a low value may partly result from strongly converted low-energy transitions or from a large ground-state branch or it could be connected with a possible enhancement of Tc yield (see Sec. IV A). Molybdenum has a tendency to form oxides but this has been observed to cause maximally a loss of roughly 50%.

Our results on the beta decay of the two ^{110}Rh species have been published recently [22] and were used in the following analysis. The next step was the determination of the direct production rate of the 28-s ^{110}Rh. The decays of this state and of the other ^{110}Rh state with 3.2-s half-life both contribute to the strong 374-keV gamma transition in ^{110}Pd. The production rate of the 28-s ^{110}Rh was obtained from the observed intensity of the 905-keV gamma transition in ^{110}Pd, which can only result from the decay of the directly produced 28-s ^{110}Rh [22]. Since the half-life, 11.6±0.6 s, of ^{110}Ru is considerably longer than the half-life of the 3.2-s ^{110}Rh daughter, it was possible to determine the contribution to the intensity of the 374-keV ^{110}Pd γ line from the decay chain ^{110}Tc$\rightarrow$ ^{110}Ru$\rightarrow$ ^{110}Rh (3.2 s) and from the directly produced ^{110}Ru in a measurement with a collection cycle of 20-s beam on and 30-s beam off. Since the half-life of ^{110}Tc is short, the total number of Ru nuclei at the beginning of the beam-off (decay) period could be determined by following the falloff of the intensity of the 112-keV transition [23] in the 3.2-s ^{110}Rh. The intensity of the ^{110}Ru$\rightarrow$ ^{110}Rh (3.2-s) ground-state branch needed for this was determined by comparing the intensities of the 112-keV transition and the 374-keV transition which follows the Rh decay. The value deduced for the ground-state branch is 70±10%. To complete the determination of the production rates it was then a straightforward matter to estimate the yield of the 3.2-s ^{110}Rh. No yield value could be determined for the stable ^{110}Pd. The half-lives of ^{110}Tc and ^{110}Ru given above are from the recent work of Äystö *et al.* [15] and Jokinen *et al.* [23], those of the two beta-decaying states in ^{110}Rh from Ref. [24].

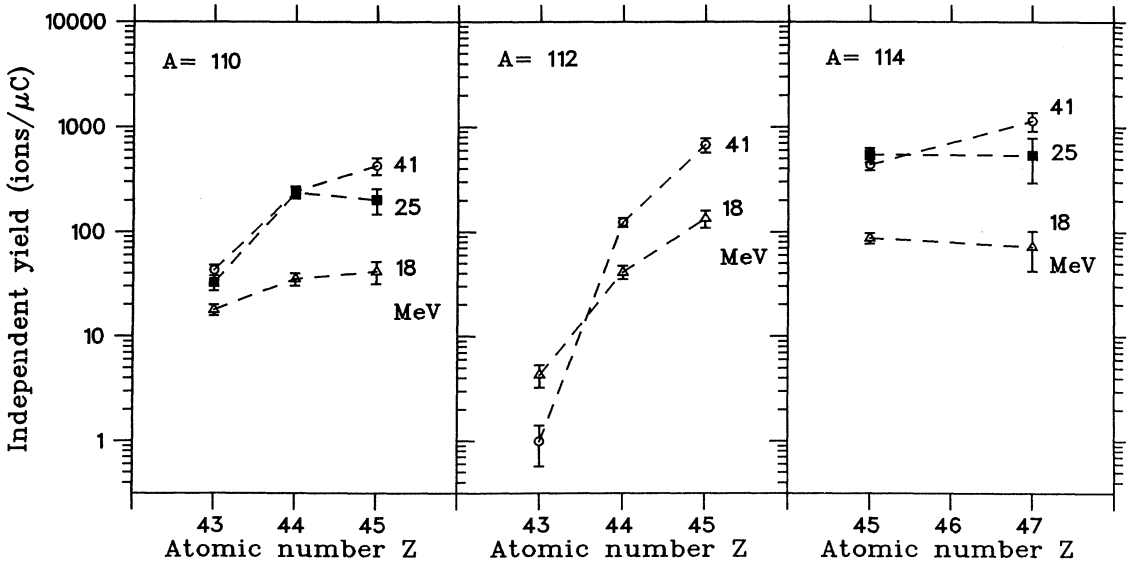

FIG. 3. Independent yields for $A=110$, 112, and 114 from the $d+^{238}$U reaction determined at three bombarding energies of 18, 25, and 41 MeV as indicated in the figure. At $A=112$, no measurements were performed with $E_d=25$ MeV. The ^{114}Pd data are not shown because of the large uncertainty in the γ branch of the 232-keV transition [27].

2. *The A=112 mass chain*

Yields of ^{112}Tc, ^{112}Ru, and ^{112}Rh were extracted from a γ-β coincidence spectrum measured using continuous proton beam of 20-MeV energy. The absolute intensity of the 236.8-keV gamma transition which follows the beta decay of ^{112}Tc [15] was used in determining the yield of ^{112}Tc. The beta-decay branch to the 0^+ ground state in ^{112}Ru was taken to be zero just as in the case of ^{110}Tc decaying to ^{110}Ru. The three gamma transitions with energies 82.3, 244.8, and 327.0 keV, which follow beta decay of ^{112}Ru, all gave compatible results for the yield of ^{112}Ru. Absolute γ intensities from Ref. [23] were used. The two γ transitions, with energies 560.5 and 777.5 keV, which only follow the beta decay of ^{112}Rh (6.8 s) and ^{112}Rh (3.8 s) [22], respectively, gave the yields of the isomeric and the ground state separately. The production ratio $Y(^{112}\text{Pd})/Y(^{112}\text{Rh})$ was measured with the planar Ge detector by making use of the 18.5-keV γ transition [25], which follows beta decay of ^{112}Pd ($T_{1/2}=21.03$ hr).

Two overlapping gamma peaks with 617.4-keV energy assigned to parent nuclides ^{112}Ag ($T_{1/2}=3.13$ hr) and $^{96}\text{Y}^m$ ($T_{1/2}=9.6$ s [26]) appear, because Y is produced as a small oxide impurity in the IGISOL ion beam. To separate the two components, a source was collected on the implantation tape using continuous beam and the decay curve of the 617.4-keV γ peak was measured in γ-β coincidence mode. The yield of ^{112}Ag relative to the cumulative yield of ^{112}Pd was determined by fitting two exponential components in the curve. The yield ratio was solved using the known half-lives.

3. *The A=114 mass chain; beta decay of the new isotope ^{114}Ru*

Prior to the present work, no experimental data on the decay of ^{114}Ru was available. Using IGISOL the $A=114$ mass chain was studied in an experiment in which the 4.0-s data-acquisition cycle consisted of a 1.6-s-long collection period and a 2.4-s-long decay period. Four γ rays with energies of 53, 88, 127, and 128 keV were seen in coincidence with 20.1-keV Rh $K\alpha$ x rays and with beta particles. Therefore they are assigned to the decay of ^{114}Ru. The half-life of ^{114}Ru was determined from the decay of gamma activity in coincidence with Rh $K\alpha$ x rays. The result was 0.53±0.06 s. The decay of ^{114}Ru will be discussed in more detail in a forthcoming publication [16].

The relative yields of ^{114}Ru and ^{114}Rh were determined from the growth-in of the 333-keV γ transition [22] in the decay of ^{114}Rh during the beam-off period. In the case of 2.4-min ^{114}Pd, use was made of its long half-life compared with those of ^{114}Ru and ^{114}Rh. Its yield was determined from the decay period in a run with a 270 s+530 s collection cycle. The decay of ^{114}Pd to levels of ^{114}Ag has been studied by us recently [27], and the 127-keV γ transition in ^{114}Ag and the level scheme from Ref. [27] were made use of in the present work. Finally, the yield of ^{114}Ag was determined by following the decay of the 558-keV γ line in ^{114}Cd during the decay period of the 270 s+530 s bombardment. The well-known [28] decay properties of ^{114}Ag were used in this analysis.

C. Independent yields in very asymmetric fission

In the reaction $d+^{238}$U, independent yields of three elements, $_{31}$Ga, $_{32}$Ge, and $_{33}$As, were determined for the charge dispersion curve at bombarding energies of 18, 25, and 41 MeV. The results are given in Fig. 4. The branch for the decay of ^{80}Ge to the 1^+ ground state of ^{80}As was determined in Jyväskylä using the reaction $p+^{238}$U. The gamma intensities in γ-β coincidence mode and the decay curve of β singles with the ΔE-E telescope were measured using periodic acquisition. The result for the ground-state branch was 65%. It was determined on the basis of the 27±5% absolute gamma intensity of the 265.3-keV transition and of the intensities of the other gamma transitions to the ground state which follow beta decay of ^{80}Ge. Our value for the gamma intensity of the 265-keV transition differs from that of Ref. [29], which is 48% but is compatible with Ref. [28], which gives 25%.

The yield of the stable nuclide ^{80}Se cannot be determined with our method and the yield of the important r-process waiting-point nucleus ^{80}Zn was too low to be measured. One can, however, estimate that the average charge $\bar{Z}$ in the $A=80$ chain is 32.3±0.1 and is independent of the bombarding energy in this energy range. The influence of ^{80}Se on $\bar{Z}$ was estimated by assuming that $\sigma_z=0.75$.

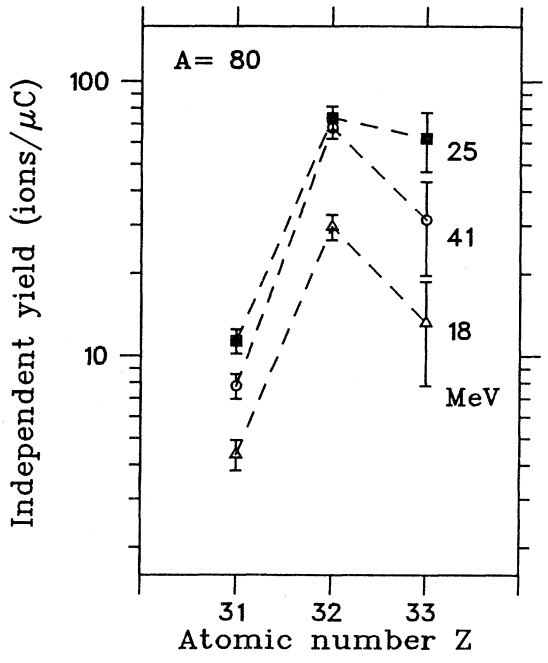

FIG. 4. Independent yields for $A=80$ from the $d+^{238}$U reaction determined at three bombarding energies of 18, 25, and 41 MeV as indicated in the figure.

IV. DISCUSSION

A. Charge dispersion in the $A=110$, 112, and 114 mass chains

The mean isobaric charge $\bar{Z}$ and the standard deviation σ_Z in $A=112$ and $A=114$ mass chains were determined using the 20 MeV $p+^{238}$U reaction. In the case of $A=110$ only the mean isobaric charge can be given with adequate accuracy due to certain identification of only three nuclides.

Our data from the $d+^{238}$U bombardment are generally insufficient for the determination of charge-dispersion parameters. Some conclusions are possible, however. In the same way as for $A=80$ in asymmetric fission (Sec. III C), the $A=110$, 112, and 114 yields seem to be constant when energy is increased above 25 MeV. The yield for $Z>43$ increases by roughly a factor of 6 as energy is changed from 18 to 25 MeV, about twice as much as for $A=80$. Tc yields will be discussed below.

It is of interest to compare our values from the $p+$U reaction with those of Wahl [5], determined for thermal-neutron-induced fission of ^{235}U. The situation is complicated by the fact that in our case the compound nucleus ^{239}Np is formed with 25 MeV of excitation energy, and more than one fissioning Np isotope is expected to contribute. According to Ref. [30], first-, second-, and third-chance fission contribute 45, 34, and 21%, respectively, at 20-MeV bombarding energy. A calculation based on these values and on the unchanged charge distribution (UCD) formula [7] was made using a basic charge-dispersion curve with a constant width of 0.52. The result was that fissions from ^{239}Np, ^{238}Np, and ^{237}Np produced a composite charge-distribution curve at $A=110$ with an only slightly increased width of 0.55. Pairing may, however, change this result in the real situation.

A comparison of our results from the $p+$U reaction and values based on the recommended independent yields given by the Z_p model [5] for the reaction ^{235}U(n_{th},f) is shown in Table I. In calculating the average charge number Z_{UCD} on the basis of the UCD model, it was assumed that the average number of neutrons emitted from the fragments $\bar{\nu}=2.5$. This value is in approximate agreement with values extrapolated from the data of Tracy *et al.* [31], in which fission of ^{238}U induced by 40–60-MeV protons was studied. It is also compatible with the recent results of Strecker, Wien, Plischke, and Scobel [32]. In the literature, charge distribution has often been characterized with parameters of a Gaussian distribution. Here, instead, the results for both reactions are presented in terms of the standard deviation σ_Z calculated directly from the independent yield values.

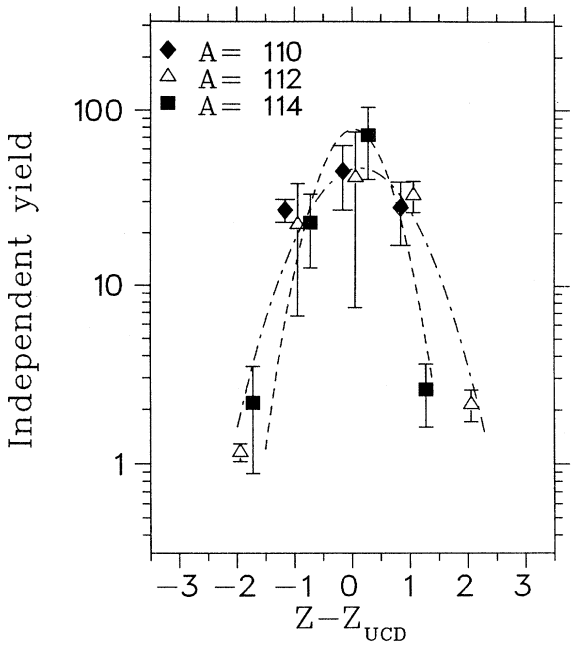

FIG. 5. Relative independent yields (in percent) of isotopes with $A=110$, $A=112$, and $A=114$ from the 20 MeV $p+^{238}$U reaction. The data for each isobarich chain was shifted using the respective Z_{UCD} values. Charge-dispersion curves with widths from Table I are drawn for $A=112$ and $A=114$.

It is well known that closed shells have an effect on low-energy fission charge-dispersion curves [33]. In $n_{th}+^{235}$U fission the yield of Sn ($Z=50$) nuclei is enhanced and consequently also the yield of the complementary Mo ($Z=42$) nuclei. In the case of $p+$U or $d+$U, shell effects, if significant, should increase the yield of Tc ($Z=43$). Possible signs of this enhancement are seen in our results. From Figs. 3 and 5 it can be seen that

TABLE I. Mean isobaric charges $\bar{Z}$ and standard deviations σ_Z in the $A=110$, $A=112$, and $A=114$ mass chains. The experimental data from the reaction 20 MeV $p+^{238}$U are from this work. Corresponding data calculated on the basis of recommended yields [5] in the reaction $n_{th}+^{235}$U are given for comparison (no experimental data are available; see text for details).

	$p+^{238}$U			$n_{th}+^{235}$U	
A	$\bar{Z}$	σ_z	Z_{UCD}[a]	$\bar{Z}$	σ_z
110	44.01±0.20	b	43.92	43.16	0.58
112	45.15±0.15	0.82±0.08	44.70	44.21	0.62
114	45.74±0.15	0.54±0.08	45.48	45.17	0.60

[a]The values based on the UCD assumption were calculated for the reaction $p+$U assuming that on the average 2.5 neutrons are emitted from the fragment [5,31,32].
[b]Not available (see text).

the charge-dispersion curves at $A=110$ are wide both in the $p+U$ reaction and in the 18-MeV $d+U$ reaction. A possible explanation is that the independent yield of Tc is enhanced. Also in the 18-MeV $d+U$ reaction with $A=112$, yield of Tc seems to be relatively high. If these effects are real, the absence of anomalously high Tc yields at 25- and 41-MeV bombarding energies can be understood as a result of excitation energy washing out shell effects. The degree of possible enhancement of Tc yield is difficult to estimate; increasing excitation energy should lead to a decrease of the yield of Tc as compared with yields of Ru and Rh, since more neutrons are emitted at higher excitation energy. However, there is only little change in the relative yields of Rh and Ag at $A=114$ as energy is increased from 18 to 41 MeV.

It can be seen from Table I that for all three mass chains the mean isobaric charge is roughly 0.8 unit higher in proton-induced fission of ^{238}U than in thermal-neutron-induced fission. One should note, however, that the mean charge in the reaction $n_{\text{th}}+^{235}$U is based on model estimates [5], since no experimental data are available for these mass numbers. When $A=114$, the standard deviations are comparable for the two reactions. In Fig. 5 the data from all three isobaric chains studied are combined by shifting the curves according to the UCD assumption [7]. Our results, especially combined with the fairly high mass yields for symmetric fission, make the ^{238}U(p,f) reaction attractive in the study of extremely neutron-rich isotopes although the dispersion curves seem to be slightly more favorable in thermal fission.

At 18-MeV bombarding energy, the compound nucleus ^{240}Np is produced at the same excitation energy in the $d+U$ reaction as ^{239}Np in the 20-MeV $p+U$ reaction. The mean isobaric charges are 46.0 (45.74±0.15) for $A=114$ and 44.5 (44.01±0.20) for $A=110$, where the values in parentheses are the corresponding results in the $p+U$ reaction. The mean charges are similar for the two reactions. The idea behind the use of the $d+U$ reaction was that a significant part of the total reaction cross section would lead to direct reactions in which a proton leaves the ^{239}U nucleus behind at low excitation energy. The result would be an increased N/Z ratio of the fission fragments. This hypothesis seems not to be supported by our experimental result since the observed independent yields (ions/μC) are smaller for ^{110,112}Tc and ^{112}Ru in the $d+U$ reaction than in the $p+U$ reaction (see Figs. 2 and 3; independent and cumulative yields are approximately equal for these very neutron-rich isotopes).

B. Charge dispersion in the $A=80$ mass chain

The mass yield at $A=80$ seems to reach a maximum around 30-MeV bombarding energy (Fig. 4). The dependence of $\bar{Z}$ on deuteron energy is weak. The excitation energy of the compound nucleus ^{240}Np is 33.0 MeV at 25 MeV deuteron energy. From this it can be estimated that fission of four nuclides, $^{237-240}$Np, contributes to the observed distribution at 25-MeV bombarding energy. According to Ref. [34], approximately 0.3 neutrons on the average are emitted in thermal-neutron-induced fission of ^{235}U from fragments with mass number 80. Taking into account the additional excitation energy brought into the system by 25-MeV deuterons the average number of neutrons emitted from fragments with $A=80$ was estimated to be 1.3. If the average mass number of the fissioning nuclei is taken to be 238.5 the average charge number given by the UCD model is 31.70. The difference $\Delta Z=Z_{\text{UCD}}-Z_L=-0.6$, where Z_{UCD} is the average isobaric charge given by the UCD model and Z_L is the experimental value for the light fragment. In thermal-neutron-induced fission of ^{233}U, approximately the same difference was observed [4].

C. Comparison with fission from the reaction ^{22}Ne+^{238}U

Spontaneous fission and a number of nuclear reactions leading to fission have been used for the purpose of studying very neutron-rich nuclei. The independent fission yields leading to a given region of the chart of nuclei depend on the reaction. Of the low-energy reactions, thermal-neutron-induced fission is still competitive in the production of new isotopes, for example, in the Cu-Ni region [2]. To study nuclei in the mass valley of low-energy fission, heavy-ion-induced fusion-fission as well as direct reactions and multinucleon transfer leading to fission can in principle be used. An example of heavy-ion-induced fission will be discussed briefly.

The independent isotopic distributions of Rb and Cs have been studied [35] earlier using an on-line mass spectrometer and the reaction 6 MeV/nucleon ^{22}Ne+^{238}U. The variance of the distribution was essentially the same for both elements and had a value of $\approx$4.25. The distributions were still roughly Gaussian at this bombarding energy which corresponds to 64.5-MeV excitation of the compound system. If one assumes that the $\bar{N}/Z$ ratio for an element depends linearly on Z and that the width of the isotopic distribution is constant, one can calculate the expected isotopic yield curves from the Ne+U reaction for the elements studied in the present work. Here $\bar{N}$ is the average neutron number [35] of an element. Assuming that the peak isotopic yield for an element is 10 mb and taking the IGISOL efficiency to be 10^{-3} the yields given in Table II were calculated.

Yields of fission products from the reaction $\sim$5–11 MeV/nucleon ^{20}Ne+^{238}U have recently been studied using the Gesellschaft für Schwerionenforschung (GSI) on-line mass separator [36]. The most neutron-rich Pd isotope observed was ^{118}Pd with a yield of 5000 atoms/s particle μA. For this isotope, the yield was 2 orders of magnitude higher than in our experiment because of the relatively long half-life [27] of 1.8 s, the higher efficiency of the separator, and the favorable hot-ion source chemistry of palladium. Gaussian isotopic yield distributions with a width of 5.6 mass units were found to fit the data in mass region $A\approx120-130$. When the bombarding energy in the work of de Saint-Simon *et al.* [35] was increased to 162 MeV ($\approx$7.4 MeV/nucleon), the variance of the isotopic distribution for Cs was increased significantly. This effect was interpreted to be a consequence of the onset of fission following direct reactions. However, it has been pointed out [10] that the yields of the neutron-rich Cs isotopes from this reaction corre-

TABLE II. Estimated yields for the reaction 132-MeV $^{22}Ne+^{238}U$ compared with experimental yields from the 20-MeV $p+^{238}U$ reaction (present work) in units of atoms/s particle μA.

Reaction	^{117}Rh	^{118}Pd	^{120}Pd	^{122}Ag
20-MeV $p+^{238}U$	>2	80	>3	Not available
132-MeV $^{22}Ne+^{238}U$	22	250	11	27

spond to those observed in 40 MeV $p+^{238}U$ fission [31]. Thus it appears that heavy-ion-induced fission is not dramatically better as a production method for neutron-rich isotopes than low-energy proton-induced fission.

V. SUMMARY AND CONCLUSIONS

The IGISOL facilities in Jyväskylä and Louvain-la-Neuve based on the ion guide principle were found suitable for performing yield measurements in both the symmetric and asymmetric mass split regions in fission induced by low-energy light charged particles. For the first time, independent fission yields were measured near symmetry for p- and d-induced fission of U with the excitation energy of the compound system around 25 MeV. Some 15 new neutron-rich isotopes in the mass region $A=110-120$ have been produced for spectroscopic studies. Our cumulative yield determinations are currently being extended over a wide range of products with masses $A=96-120$. The yields are also being studied as a function of proton energy in the range 14–20 MeV using thin U and Th targets [37].

Heavy-ion accelerators will provide new ways to reach neutron-rich isotopes. In particular, fusion-fission and deep-inelastic reactions might be productive together with (a new generation of) IGISOL. Not very much is known about charge distribution from heavy-ion fusion and deep-inelastic reactions [10,11,38] in the product mass range $A\approx100-130$ although much effort has been put into the determination of yields of heavy elements, in particular, from deep-inelastic reactions [39,40]. Whether new regions of the nuclear chart will be reached remains to be seen. Our estimates based on results from the $^{22}Ne+^{238}U$ reaction are not very promising. However, population of high-spin isomers is expected to be enhanced. Already in the 20-MeV p-induced fission of U, exotic isomers with half-lives on the order of a few milliseconds have been observed in our studies [1]. For example, in the case of ^{114}Ag, the population ratio of the isomeric versus ground state was 0.04.

ACKNOWLEDGMENTS

The authors are grateful to P. Armbruster for illuminating discussions. This work has been supported by the Academy of Finland. One of us (M.L.) would like to thank the Wihuri Foundation for a travel grant.

[1] H. Penttilä, J. Äystö, P. Jauho, A. Jokinen, J. M. Parmonen, P. Taskinen, K. Eskola, M. Leino, P. Dendooven, and C. N. Davids, Phys. Scr. **T32**, 38 (1990).
[2] M. Bernas, P. Armbruster, J. P. Bocquet, R. Brissot, H. Faust, Ch. Kozhuharov, and J. L. Sida, Z. Phys. A **336**, 41 (1990).
[3] N. Kaffrell, P. Hill, J. Rogowski, H. Tetzlaff, N. Trautmann, E. Jacobs, P. de Gelder, D. de Frenne, K. Heyde, S. Börjesson, G. Skarnemark, J. Alstad, N. Blasi, M. N. Harakeh, W. A. Sterrenburg, and K. Wolfsberg, Nucl. Phys. **A470**, 141 (1987).
[4] U. Quade, K. Rudolph, S. Skorka, P. Armbruster, H.-G. Clerc, W. Lang, M. Mutterer, C. Schmitt, J. P. Theobald, F. Gönnenwein, J. Pannicke, H. Schrader, G. Siegert, and D. Engelhardt, Nucl. Phys. **A487**, 1 (1988).
[5] A. C. Wahl, At. Data Nucl. Data Tables **39**, 1 (1988).
[6] Yu. Ts. Oganessian and Yu. A. Lazarev, in *Treatise on Heavy-Ion Science,* edited by D. A. Bromley (Plenum, New York, 1989), Vol. 8.
[7] R. Vandenbosch and J. R. Huizenga, *Nuclear Fission* (Academic, New York, 1973).
[8] T. M. Semkow, A. C. Wahl, and L. Robinson, Phys. Rev. C **30**, 1966 (1984).
[9] J. P. Bocquet and R. Brissot, Institut Laue Langevin report.
[10] W. Reisdorf, M. de Saint-Simon, L. Remsberg, L. Lessard, C. Thibault, E. Roeckl, and R. Klapisch, Phys. Rev. C **14**, 2189 (1976).
[11] S. A. Karamyan, Yu. Ts. Oganesyan, Yu. E. Penionzhkevich, and B. I. Pustyl'nik, Yad. Fiz. **9**, 715 (1969) [Sov. J. Nucl. Phys. **9**, 414 (1969)].
[12] J. Ärje, J. Äystö, P. Taskinen, J. Honkanen, and K. Valli, Nucl. Instrum. Methods Phys. Res. Sec. B **26**, 384 (1987).
[13] P. Taskinen, H. Penttilä, J. Äystö, P. Dendooven, P. Jauho, A. Jokinen, and M. Yoshii, Nucl. Instrum. Methods Phys. Res. Sec. A **281**, 539 (1989).
[14] H. Penttilä, H. Salminen, J. Äystö, K. Eskola, Z. Janas, P. P. Jauho, A. Jokinen, M. E. Leino, and J. M. Parmonen, University of Jyväskylä, Department of Physics, Annual Report 1990.
[15] J. Äystö, P. P. Jauho, Z. Janas, A. Jokinen, J. M. Parmonen, H. Penttilä, P. Taskinen, R. Béraud, R. Duffait, A. Emsallem, J. Meyer, M. Meyer, N. Redon, M. E. Leino, K. Eskola, and P. Dendooven, Nucl. Phys. **A515**, 365 (1990).
[16] A. Jokinen *et al.* (unpublished).
[17] H. Penttilä, P. P. Jauho, J. Äystö, P. Decrock, P. Dendooven, M. Huyse, G. Reusen, P. Van Duppen, and J. Wauters, University of Jyväskylä, Department of Physics, Annual Report 1990.
[18] H. Penttilä, J. Äystö, K. Eskola, Z. Janas, P. P. Jauho, A. Jokinen, M. E. Leino, J. M. Parmonen, and P. Taskinen, Z. Phys. A **338**, 291 (1991).
[19] Z. Janas, J. Äystö, K. Eskola, P. Jauho, A. Jokinen, J. Kownacki, M. Leino, J. M. Parmonen, H. Penttilä, J. Szerypo, and J. Żylicz, University of Jyväskylä, Department of Physics, Annual Report 1990.
[20] K. Deneffe, B. Brijs, E. Coenen, J. Gentens, M. Huyse, P.

Van Duppen, and D. Wouters, Nucl. Instrum. Methods Phys. Res. Sec. B **26**, 399 (1987).
[21] J. M. Parmonen, Z. Janas, W. H. Trzaska, J. Äystö, J. Kantele, P. Jauho, A. Jokinen, and H. Penttilä, Nucl. Instrum. Methods Phys. Res. Sec. A (in press).
[22] J. Äystö, C. N. Davids, J. Hattula, J. Honkanen, K. Honkanen, P. Jauho, R. Julin, S. Juutinen, J. Kumpulainen, T. Lönnroth, A. Pakkanen, A. Passoja, H. Penttilä, P. Taskinen, E. Verho, A. Virtanen, and M. Yoshii, Nucl. Phys. **A480**, 104 (1988).
[23] A. Jokinen, J. Äystö, P. Dendooven, K. Eskola, Z. Janas, P. P. Jauho, M. E. Leino, J. M. Parmonen, H. Penttilä, K. Rykaczewski, and P. Taskinen, Z. Phys. A (in press).
[24] P. de Gelder, E. Jacobs, and D. de Frenne, Nucl. Data Sheets **38**, 545 (1983).
[25] D. de Frenne, E. Jacobs, and M. Verboven, Nucl. Data Sheets **57**, 443 (1989).
[26] H. W. Mueller, Nucl. Data Sheets **35**, 281 (1982).
[27] V. Koponen, J. Äystö, J. Honkanen, P. Jauho, H. Penttilä, J. Suhonen, P. Taskinen, K. Rykaczewski, J. Żylicz, and C. N. Davids, Z. Phys. A **333**, 339 (1989).
[28] *Table of Isotopes,* 7th ed., edited by C. M. Lederer and V. S. Shirley (Wiley, New York, 1978).
[29] B. Singh and D. A. Viggars, Nucl. Data Sheets **36**, 127 (1982).
[30] J. R. Boyce, T. D. Hayward, R. Bass, H. W. Newson, E. G. Bilpuch, and F. O. Purser, Phys. Rev. C **10**, 231 (1974).
[31] B. L. Tracy, J. Chaumont, R. Klapisch, J. M. Nitschke, A. M. Poskanzer, E. Roeckl, and C. Thibauldt, Phys. Rev. C **5**, 222 (1972).
[32] M. Strecker, R. Wien, P. Plischke, and W. Scobel, Phys. Rev. C **41**, 2172 (1990).
[33] A. C. Wahl, Phys. Rev. C **32**, 184 (1985).
[34] V. F. Apalin, Yu. N. Gritsyuk, I. E. Kutikov, V. I. Lebedev, and L. A. Mikaelian, Nucl. Phys. **71**, 553 (1965).
[35] M. de Saint-Simon, L. Lessard, W. Reisdorf, L. Remsberg, C. Thibault, E. Roeckl, R. Klapisch, I. V. Kuznetsov, Yu. Ts. Oganessian, and Yu. E. Penionshkevitch, Phys. Rev. C **14**, 2185 (1976).
[36] W.-D. Schmidt-Ott, F. Meissner, P. Koschel, U. Bosch-Wicke, R. Kirchner, O. Klepper, H. Folger, E. Roeckl, A. Plochocki, K. Rykaczewski, and Z. Preibisz, Nucl. Phys. **A522**, 610 (1991).
[37] P. P. Jauho *et al.* (unpublished).
[38] G. D. Adeev, L. A. Filipenko, and P. A. Cherdantsev, Yad. Fiz. **33**, 90 (1981) [Sov. J. Nucl. Phys. **33**, 47 (1981)].
[39] H. Keller, R. Bellwied, K. Lützenkirchen, J. V. Kratz, W. Brüchle, H. Gäggeler, K. J. Moody, M. Schädel, and G. Wirth, Z. Phys. A **328**, 255 (1987).
[40] K. Lützenkirchen, J. V. Kratz, W. Brüchle, H. Gäggeler, K. Sümmerer, and G. Wirth, Z. Phys. A **317**, 55 (1984).