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Production of neutron-rich nuclides and radioactive beams by intermediate energy ^{238}U fission

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The yields of neutron-rich fission fragments from the interaction of 20 MeV/nucleon ^{238}U with ^{208}Pb have been measured. The production mechanism of these fragments is consistent with sequential fission following a quasielastic or deep inelastic collision. Substantial yields of very n-rich fragments are observed. The importance of these data for generation of n-rich radioactive beams by fission of intermediate-energy projectiles is discussed. [S0556-2813(97)50905-8]

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Many proposed radioactive beam facilities are based on the use of neutron-rich radioactive beams. These beams are to be generated by the fission of uranium induced by neutrons or energetic protons. Uranium is chosen as a target because of its large N/Z ratio (1.59 for ^{238}U) and the fact that in fission this N/Z ratio is largely preserved in the fragments, making them n-rich relative to the line of stability. Neutrons or energetic protons are chosen as projectiles because of the availability of high intensity beams and the large penetrating power of these beams, enabling the use of thick targets and high production rates. Such facilities are usually standard on-line isotope separator (ISOL) facilities, which have practical limits on the half-lives (1–20 s) of the n-rich species produced due to delays in release from the target matrix and ionization [1]. Projectile fragmentation (PF) facilities, while not having the overall beam intensities of ISOL facilities, may have some advantages that complement the standard ISOL facilities. The lack of a release or delay time in a PF facility will allow study and use of very short-lived nuclides.

Pioneering work done at GSI [2] has demonstrated that a PF facility can be a useful source of n-rich secondary beams. These experiments, utilizing the fragmentation of relativistic (750 MeV/nucleon) ^{238}U ions, demonstrated the production of more than 100 new n-rich radionuclides. The reaction mechanism for the production of these n-rich nuclei was low energy fission induced by electromagnetic excitation and peripheral nuclear interactions [2–4]. We report the observation of an alternative production mechanism for a number of these same very n-rich nuclei in the interaction of intermediate energy (20 MeV/nucleon) ^{238}U with ^{208}Pb. The reaction mechanism in these studies is consistent with quasielastic or deep-inelastic scattering of the ^{238}U *followed by sequential fission.* (It has been shown previously that deep inelastic scattering, by itself, can lead to the production of new very n-rich nuclei [5].) The ability to produce and study these n-rich nuclei at intermediate (and possibly low) energy facilities should open new opportunities for a broad and diversified program of studies.

The experiment was performed at the National Superconducting Cyclotron Laboratory at Michigan State University using the A1200 fragment separator [6]. The use of the A1200 for the production and identification of medium mass fragments has been discussed previously [7,8]. In these measurements, the A1200 was operated in the medium acceptance mode (angular acceptance 0.8 msr, momentum acceptance 3%).

A 20 MeV/nucleon $^{238}U^{35+}$ beam ($i=0.04$ particle nA) struck a production target of ^{208}Pb (5.8 mg/cm^2, 99.1% enriched) at the object position of the spectrometer at an angle of 1° relative to the optical axis of the spectrometer. Fission fragments from the decay of the scattered projectile, arriving at the first dispersive image of the spectrometer, passed through a slit defining a 3% momentum acceptance and an $X-Y$ position-sensitive parallel-plate avalanche counter (PPAC) (giving a ''start'' timing signal).

At the focal plane (14 m from the start detector), the fragments passed through a microchannel plate timing detector (giving a ''stop'' signal) and then into a four element (50, 50, 300, and 500 μm) Si detector telescope. For each event, time-of-flight (resolution 0.8 ns FWHM, typical TOF 240 ns), dE/dx, E, and magnetic rigidity ($B\rho$) were recorded.

The spectrometer and the detectors were calibrated using low intensity ^{238}U beam and a series of analog beams (^{197}Au, ^{156}Gd, ^{136}Xe, ^{129}Xe, ^{109}Ag, ^{95}Mo, ^{68}Zn, and ^{27}Al) covering a wide range of Z, A (at three different energies). From the measured quantities and the calibration data, the values of the atomic number Z, ionic charge q, mass number A, and velocity were calculated for each event. Details of the procedure are reported elsewhere [8]. The resolution for Z, q and A for fragments with mass around $A=100$ were 0.5, 0.4, and 0.7 units, respectively (Fig. 1). To cover the mass and velocity range of the fission fragments, data were taken at several overlapping magnetic rigidity settings of the spectrometer. The $B\rho$ settings were such that only fission fragments emitted forward in the rest frame of the scattered projectilelike fissioning nucleus were accepted by the spectrometer. Normalization of beam current for data taken

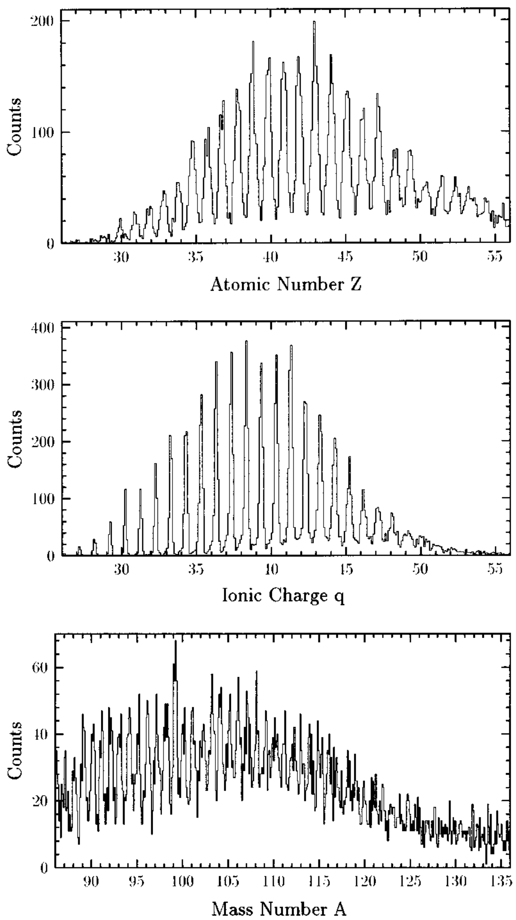

FIG. 1. Typical examples of measured Z, q, and A distributions for fission fragments from the reaction of 20 MeV/nucleon ^{238}U with ^{208}Pb at a single spectrometer setting.

at different settings was obtained with a set of four monitor detectors mounted around the target position. (These detectors were calibrated to the absolute beam current.)

To associate a given event with the formation of a fragment with a given Z, q, and A, we required that the calculated fragment Z, q, and A values were within ± 0.30, 0.25, and 0.50 units, respectively, of the corresponding integer values. No cuts were made on fragment velocity except those implied by the spectrometer $B\rho$ setting.

Applying the criteria for Z and q to the Mo ($Z=42$) fragments, as an example, and summing over all observed q values and velocities, gives the mass distribution shown in Fig. 2. For this element as for several others ($Z=38-48$), we have evidence for the formation of ''new'' n-rich nuclei. (By the term ''new,'' we mean these nuclei are not listed in current [9] compilations of nuclear data.) *These nuclei have been reported previously in the published or unpublished observations of relativistic projectile fission [2].* This work confirms that finding using a different production mechanism (see below). A list of the ''new'' nuclei observed in 24 hours of ''on-target'' beam time is given in Table I.

To obtain production cross sections, the observed counts were corrected for the yields of charge states missed by the focal plane detectors (due to charge changing of the ions passing through the PPAC at the first image of the A1200). The corrections were based on the ionic charge distribution

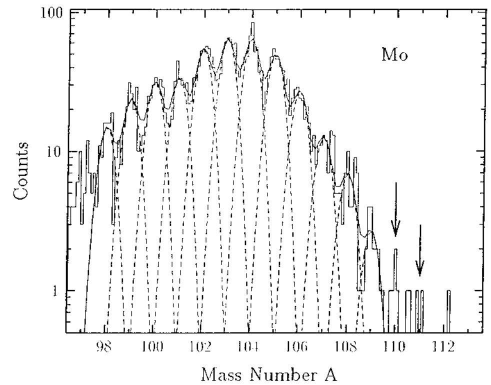

FIG. 2. Mass histogram for the $Z=42$ (Mo) fragments from the interaction of 20 MeV/nucleon ^{238}U with ^{208}Pb with arrows indicating the ''new'' nuclei. The dashed lines indicate the calculated spectrometer response function for fragments of given Z ($=42$) and A. The solid line is the summed response function.

data of Baron *et al.* [10] and were typically factors of 2–4. Subsequently, the yields were corrected for the limited angular acceptance of the spectrometer. The angular distribution of the fission fragments was assumed to arise from an isotropic emission in the frame of the fissioning nucleus (taken to be ^{238}U) deflected on average at an angle equal to the grazing angle. Finally, due to lack of absolute cross sections for this reaction, the cross sections were adjusted so that the integral of the mass yield curve equals the reaction cross section taken from Ref. [11].

The production cross sections for the ''new'' nuclei are given in Table I and compared to those observed in relativistic projectile fission. These cross sections are similar in magnitude. Given the similarity in ''(beam intensity $\times$ target thickness)'' products in the two experiments, lower production rates of these n-rich nuclei are observed in the present measurements, primarily due to the weaker forward focusing and charge state losses of the fission fragments at 20 MeV/nucleon, as compared to the very forward-focused and fully ionized fragments at relativistic energies [2].

The isobaric charge distributions for the ^{238}U + ^{208}Pb

TABLE I. List of the ''new'' nuclei and the number of counts observed in the present measurement (24 h data taking period). The cross sections are obtained as described in the text. Also listed are the production cross sections of these nuclei from the fragmentation of relativistic ^{238}U [2].

This work: Nuclide	Counts	$\sigma(\mu b)$	Ref. [2] $\sigma(\mu b)$
^{106}Zr	4	274	165
^{107}Zr	2	174	21
^{111}Mo	3	167	66
^{114}Tc	4	290	24
^{116}Ru	4	173	63
^{117}Ru	2	187	11
^{118}Rh	7	336	160
^{121}Pd	4	83	150
^{122}Pd	3	84	25
^{125}Ag	2	163	37

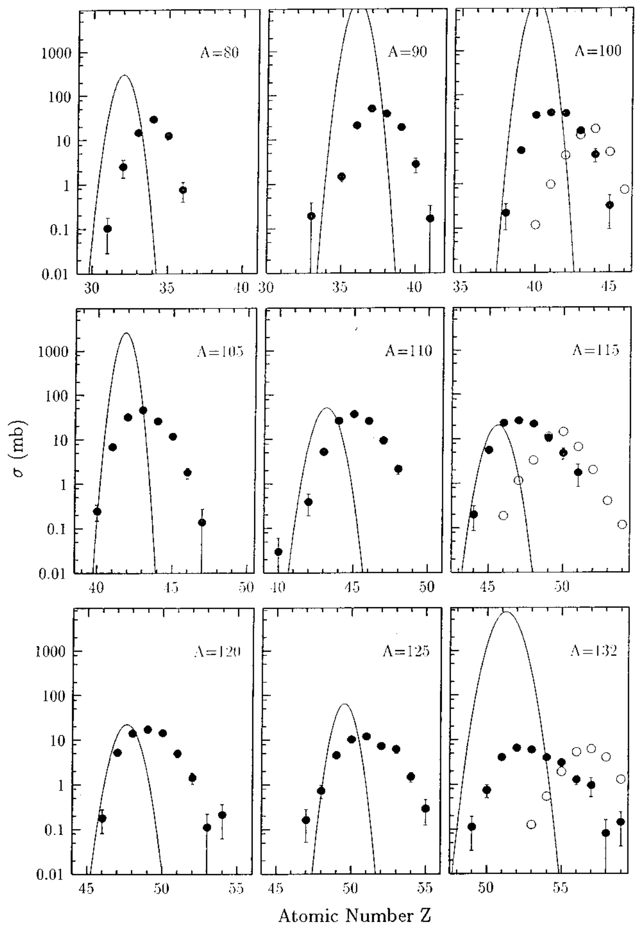

FIG. 3. Charge distributions for representative nuclei formed in the 20 MeV/nucleon $^{238}U + ^{208}Pb$ reaction (filled circles). Also shown are similar data for thermal neutron induced fission of ^{235}U (solid lines) and the 20 MeV/nucleon $^{238}U + ^{27}Al$ reaction (open circles). Statistical uncertainties are shown when they exceed the size of the plotting symbols.

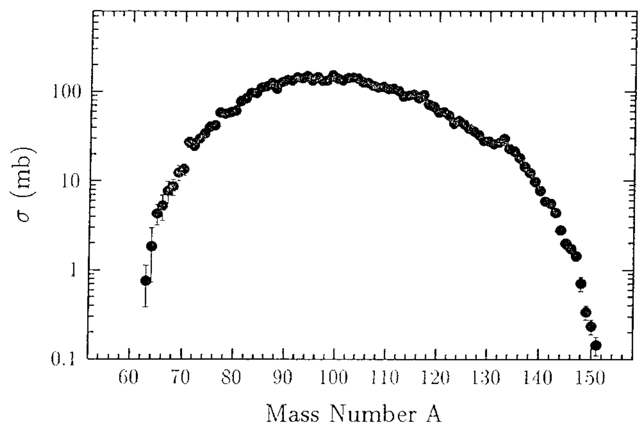

FIG. 4. Mass yield curve for the n-rich fission fragments from the 20 MeV/nucleon $^{238}U + ^{208}Pb$ reaction. Uncertainties are shown when they exceed the size of the plotting symbols.

reaction, as shown in Fig. 3, are very broad (typical $\sigma \simeq 1.2$), and centered at N/Z values larger than those reported in radiochemical measurements of the (20 MeV/nucleon) $^{12}C + ^{238}U$ reaction [12] and those observed in a parallel A1200 measurement of fission fragment distributions from the (20 MeV/nucleon) $^{238}U + ^{27}Al$ reaction [13]. Also shown in Fig. 3 are the corresponding charge distributions for the thermal neutron induced fission of ^{235}U [14], which are expected to be similar in shape to those observed in low energy fission processes induced by relativistic heavy ions [2].

The most probable primary fragment charge Z_p for fragments from neutron-induced fission is substantially lower, i.e., the fragments are more n-rich (although the widths for the higher-energy processes seen in this work are greater). This fact, along with the larger fission cross section for the neutron-induced reaction, makes ISOL facilities substantially better sources of longer-lived n-rich radioactive beams than PF facilities [with the possible exception of symmetric fission products ($A=110$–125), where the neutron-induced fission mass yield curve has a minimium and (as seen in Fig. 3) the cross sections are comparable to the ones obtained with a PF facility]. For n-rich nuclei on (or near) the r-process path as some of the ''new'' nuclei are, the predicted half-lives [15] are less than 1 second, making their production in ISOL facilities very difficult (due to delays in ionization and release times).

We believe that we can rule out any low energy fission process as the source of these n-rich nuclei by the symmetric shape of the fission mass distribution (Fig. 4). Similarly, the large shift in the centroids of the charge distributions as the target nucleus is changed from ^{27}Al to ^{208}Pb suggests a reaction mechanism in which the size and relative neutron richness of the target nucleus plays a role (Fig. 3). The relatively low c.m. energy of the fissioning ^{238}U of 9.3 MeV/nucleon would also argue against a ''fragmentation'' mechanism. Previous studies of the $^{238}U + ^{208}Pb$ or $^{238}U + ^{238}U$ reactions at somewhat lower energies [16] indicate that the fission fragments are due to sequential fission following a quasi- or deep-inelastic collision. Similar conclusions were reached in a related study of the reaction 24.3 MeV/nucleon $^{238}U + ^{197}Au$ [17].

It is interesting to evaluate what beam intensities one might expect for very short-lived n-rich nuclei at a second generation PF facility. Assuming a primary ^{238}U beam intensity of 2×10^{10} [18] at 50 MeV/nucleon, the same fragment production cross sections and charge distributions as obtained in this work, a production target thickness of 25 mg/cm^2 ^{208}Pb, a fragment separator angular acceptence of 10 msr and momentum acceptance of 6%, one can estimate secondary beam intensities of 1–500 particles/s at 50 MeV/nucleon. Such intensities would allow the study of the structure of these nuclei, but their use for the study of nuclear reactions would require substantially higher ^{238}U beam intensities.

For very neutron-rich nuclei along the r-process path, a counting rate of 10–100 per day should be enough to verify their stability and in the most favorable cases allow measurements of their decay properties. As an example, extrapolating the charge distributions of the present work (assuming a Gaussian shape), we can estimate the production cross sections for the r-process waiting point nuclei ^{129}Ag, ^{128}Pd, and ^{127}Rh ($N=82$ isobars) to be 3 μb, 0.3 μb, and 3 nb respectively, and predict (under the above mentioned assumptions for a PF facility) counting rates of 500, 50, and 2 particles per day, respectively.

In designing a PF radioactive beam facility to produce n-rich nuclei using the reaction mechanisms discussed in this work, one might note that one expects the angular distribution of the fissioning nucleus is centered near the grazing angle [17], which in the case of $^{238}U + ^{208}Pb$ is 10.5° at 20 MeV/nucleon (and 3.4° at 50 MeV/nucleon). Also one might speculate that if quasi- or deep-inelastic scattering followed by sequential fission is the mechanism for the production of the neutron rich nuclei at the energy of the present work, the higher beam intensities available at low energy (<10 MeV/nucleon) accelerators might compensate for the lack of forward focusing and smaller cross sections to allow use of these facilities to generate n-rich radioactive beams.

In summary, we have performed a high-resolution study of fission fragments from the reaction of 20 MeV/nucleon ^{238}U with ^{208}Pb. We find that this reaction leads to the production of a number of very neutron-rich nuclei. The production mechanism is consistent with sequential fission following a quasi- or deep-inelastic scattering event. The measured charge distributions and production cross sections indicate that useful intensities of very short-lived very n-rich nuclei could be produced at intermediate (and possibly lower) energy PF facilities which (along with the relativistic energy PF facility of GSI) would complement the more intense beams of longer-lived n-rich nuclei expected from ISOL facilities.

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