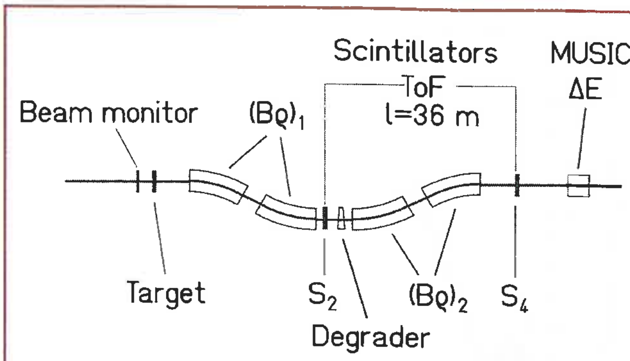
Comprehensive measurement and analysis of spallation in 1AGeV ^{238}U on protons

Monique Bernas
IPN-Orsay/CNRS-IN2P3 • *France*

The still increasing and developing world population demands more energy. Further burning of coal, oil and gas produces still more CO^2 producing deterioration of the climate. Regardless, oil and gas will run out in the middle of the century. Planning a decent future with a minimum supply of energy for those who will follow us, requires a re-examination of all possibilities of non-polluting, renewable, and sustainable energies including nuclear options. There are two nuclear reactions, which allow for a long-term exploitation: 1) converting H into He by fusion or 2) converting ^{238}U and ^{232}Th into ^{239}Pu and ^{233}U to be burnt by fission. In this paper, the fission option is the target. The real long-term reserves of fission energy lie in the even-even isotopes ^{238}U and ^{232}Th, as the supplies of ^{235}U, just like fossil fuels, are restricted, as well. The remaining ^{235}U and available ^{239}Pu could be burnt in a 3rd generation of reactors (EPR) using mixed U/Pu-fuel (MOX). Aiming at a contribution of fission energy to the world electricity production of one third, that is three times more nuclear power than today, reactors of a 4th generation should be envisaged, ready to offer wide technological applications in the middle of the century. Such an option should be foreseen as a necessary implement of a peaceful future.

Discussed in this scenario are improved systems of fast reactors, of high temperature gas-cooled reactors and of molten-salt reactors. The innovative vision for the 4th generation is Accelerator Driven reactor Systems (ADS). The nuclear waste is separated locally at the reactor sites into fissionable actinides (Th, U, Pu) to be burnt in the fuel-cycle of power reactors, and minor actinides (Np, Am, Cm) and long-lived fission products to be incinerated and transmuted in ADS. To this goal, a high intensity, 30 – 50 mA, proton beam at 1 GeV is coupled with a reactor core. The latter is run either with fast neutrons for U/Pu-fuels and minor actinides / fission products incineration or with thermal neutrons for Th/U-fuels. Two nuclear reactions are combined – spallation by protons producing cheap neutrons and nuclear fission releasing energy. Looking at the data still missing for the new systems, it is evident that spallation and fission at 1 GeV on a technological level is only poorly known. Knowledge of the chemical composition of the inventory, the material damages and the radioactivity induced by 1 GeV protons, are needed for the realisation of the spallation target of ADS.

The accelerator facility UNILAC/SIS at GSI gives access to 1 GeV beams of any stable element. A high-resolution spectrometer, the FRS, is available for investigations of reaction products. Spallation of the building materials of an ADS – U, Pb and Fe – as prototypes can be investigated in inverse kinematics. This method gives access to cross sections of almost all nuclides produced, as well as to their kinetic energies and the reaction mechanism of their production. In 1997 an experimental program was launched at GSI financed by the EU to provide data for the proposed prototype materials. A high accuracy aiming for a 10 % uncertainty in the isotopic cross sections of all elements is demanded for ADS-technology.

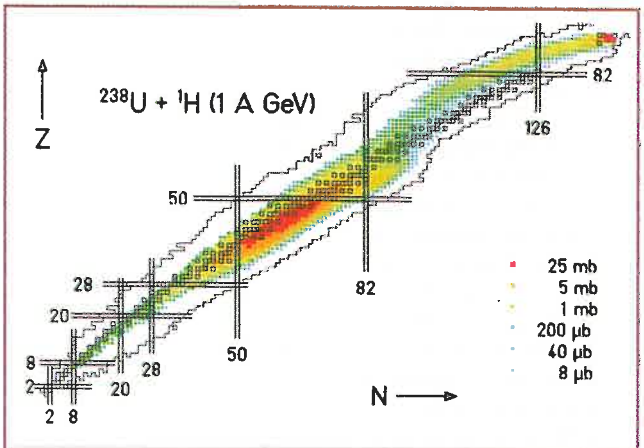


▲ Fig. 1: Schematic layout of the fragment separator FRS with the detector equipment.

In this article, we report on the campaign dedicated to the studies of spallation reactions in inverse kinematics. Most of the experimental results have been published in scientific journals, are documented in PhD-theses, or will be published soon. A comprehensive overview of the project and the results obtained can be found in Ref. [1]. The results were obtained by physicists from IPN-Orsay/CNRS-IN2P3, France, DAPNIA /CEA-Saclay, France, the University of Santiago de Compostela, Santiago, Spain in collaboration with the physicists of the GSI, Darmstadt, Germany. (Cf photo of physicists involved.)

Experiment

The experimental method and the analysis procedure have been developed and applied in previous experiments [2,3,4]. The heavy-ion synchrotron SIS at GSI, Darmstadt, can deliver the primary beams at energies between 0.2 – 1.5 A GeV. The dedicated experimental set up is shown in Fig. 1. A liquid hydrogen target was installed Ref. [5]. Heavy residues produced in the target were



▲ Fig. 2: The 1385 identified nuclides are shown on a chart of nuclei. Numerical values are available in [9]. The magnitudes (reducing by a factor of 5) of the experimental cross-sections are indicated by a colour scale. The stable isotopes are indicated by black squares.

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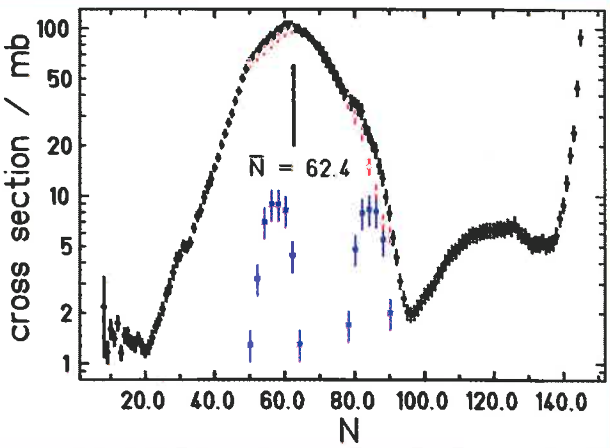


▲ **Fig. 3:** Measured N-distribution for all neutron numbers between N = 8 and 146. Summed cross sections (full points) from cross sections for low-energy asymmetric fission (blue squares) and for high-energy asymmetric fission (red circles) are reported separately. The minimum at N = 97 separates EVR and FF.

all strongly forward focused due to the inverse kinematics and the high velocity of the incoming beam. They were identified using the Fragment Separator (FRS) [6] and the associated detector equipment.

The FRS is a two-stage magnetic spectrometer with a dispersive intermediate image plane (S_2) and an achromatic final image plane (S_4), with a momentum acceptance of 3% and an angular acceptance of about 15 mrad around the beam axis. Two position-sensitive plastic scintillators placed at S_2 and S_4, respectively, provided the magnetic-rigidity ($B\rho$) and time-of-flight measurements, which allow the mass-over-charge ratio of the particles to be determined. For an unambiguous isotopic identification of the reaction products, the analysis was restricted to ions, which passed both stages of the fragment separator fully stripped. The losses in counting rate due to the fraction of incompletely stripped ions and the losses due to secondary reactions in the layers of matter in the beam line were corrected for.

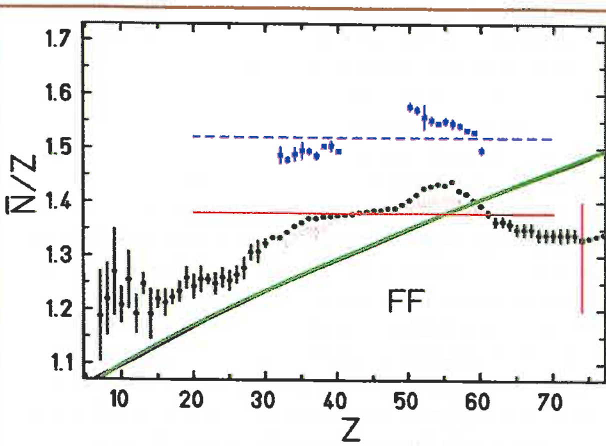
To identify all residues in the whole nuclear-charge range up to the projectile, it was necessary to use two independent methods in the analysis. The nuclear charges of the lighter elements, mainly produced by fission, were deduced from the energy loss in an ionisation chamber (MUSIC) with a resolution $Z/\Delta Z \approx 200$ obtained for the heaviest residues. Combining this information with the mass-over-charge ratio, a complete isotopic identification was performed. A mass resolution of $A/\Delta A \approx 400$ was achieved. Since part of the heavier reaction products was not completely stripped, the MUSIC signals were not sufficient for an unambiguous Z identification. Therefore, the identification of reaction products of heavier elements was performed with the help of an achromatic energy degrader [7] placed at the intermediate image plane of the FRS. Degrader thicknesses of about 5 g/cm^2 of aluminium were used. The nuclear charge of the products was deduced from the reduction in magnetic rigidity by the slowing down in the energy degrader. The MUSIC signal was still essential for suppressing events of incompletely stripped ions and from nuclei destroyed by secondary reactions in the degrader. The velocity of the identified residue was determined at S_2 from the $B\rho$ value with a relative uncertainty of $5 \cdot 10^{-4}$ and transformed into the frame of the beam. More than 100 different values of the magnetic fields were used in steps of about 2 % in order to cover all the produced residues and to construct the full velocity distribution of each residue in one projectile-target combination.

The re-construction of the full velocity distribution allows the reaction products formed in spallation and fission reactions to be disentangled due to their different kinematic properties. For isotopes produced by fission, only those emitted in a forward or backward direction with respect to the primary beam can be observed for given settings of the FRS because the angular acceptance is too small to observe sideward-emitted fragments. Isotopes produced as evaporation residues have a single narrow velocity distribution and are fully transmitted.

Reaction Mechanisms Cross Sections, and Recoil Velocities

The production of residual nuclides has been investigated for several systems, which are particularly relevant for the design of Accelerator-Driven Systems: $^{56}Fe+^{1}H$ at 0.2 - 1.5 *A* GeV, $^{208}Pb+^{1}H$ at 0.5 and 1 *A* GeV, $^{238}U+^{1}H$ at 1 *A* GeV [8]. For each system, the production rates of large numbers of final residues were measured. In the case of heavy systems, this amounts to more than a thousand nuclides per system. Moreover the velocity distributions of all these nuclides were determined. As an example, Figure 2 shows the measured production cross sections from the reaction 238U+1H at 1 A GeV [9-12] using a colour logarithmic scale. In this unique comprehensive "transmutation" of uranium, all elements from uranium to nitrogen, each with a large number of isotopes are observed. In the cross-section range down to 10 μb, 1385 nuclides are observed, as shown on the figure. The total cross-section of (1.97 ± 0.3) b divides into (1.53± 0.2) b for fission fragments (FF) and (0.44 ± 0.1) b for evaporation residues (EVR).

The spallation reaction is described as a two-step process: The collision of the fast proton on the heavy nucleus generates a cascade of nucleon-nucleon collisions in the target nucleus. Depending on the excitation energy deposited and the number of fast neutrons emitted, the remaining target nucleus gives up excitation energy by neutron-evaporation. In the process of de-excitation, the still excited nucleus can either undergo fission or continue to cool down by emitting still more neutrons. On the



▲ **Fig. 4:** The mean N̄/Z ratio plotted as a function of the atomic number. The different contributions are represented by different symbols (see Fig. 3). The valley of stability (green line) is shown. The mean N̄/Z ratio for the low-energy asymmetric process (dashed blue line) and for ^{107}Rh, representative of the high-energy process (red line), are indicated. The vertical line at Z = 74 separates fission fragments from evaporation residues.

chart of populated isotopes plotted on Fig. 2, the two de-excitation processes are well characterized. The EVR observed in the upper-right corner merge in the region of Z=73-77 with FF observed for elements below.

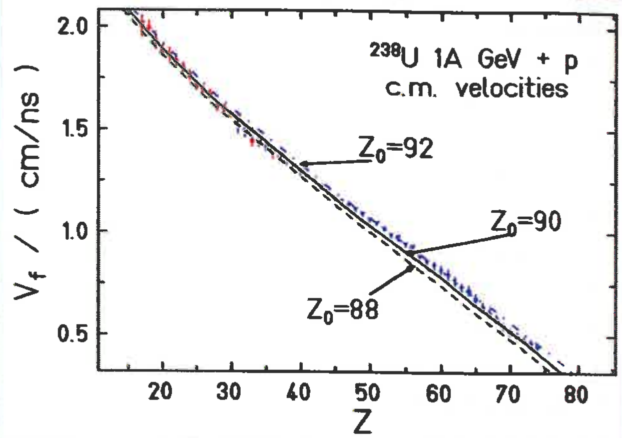
From U (red, in the upper right corner) the EVR-cross sections first fall down rapidly to N = 138 (Fig.3), covering 0.24b, the main part of the EVR cross section. Here one new isotope ^{235}Ac was discovered. The cross sections then show an extended plateau between N = 110 and N = 138 at a low level of 5.0 to 6.4 mb. At the upper limit, N = 138, highly fissionable nuclei are found being deformed and having small ground-state shell corrections. As N decreases further, spherical nuclei around N = 126 follow having large ground state shell corrections and being less fissionable. At the lower limit, N=110, fission barriers break down again and decreasing cross sections would be expected. It remains a challenge to understand the plateau of cross sections in the N-distribution. All we know about fission, level densities and their dependence on excitation energy is put to the test. At 1 GeV the excitation energy transferred in the primary cascade reaches an upper limit of about 500 MeV for a range of mass losses up to $\Delta A = 70$. The cross sections then decrease rapidly for N < 110.

The distribution of FF reveals a small (5%) contribution of the classical asymmetric low energy fission [11] as shown on Fig. 3. The underlying parent nuclei are relatively cold and cluster around ^{233}U. By subtracting the low-energy asymmetric fission, we obtain the high-energy symmetric distribution with a mean neutron number of 61.9. Combined with the mean proton number 44.9 obtained from the Z-distribution, a mean mass number of A = 106.8 is reconstructed for FF. $^{107}_{45}$Rh is the mean nuclide produced in high-energy symmetric fission.

The standard deviation of the high-energy symmetric Z- distribution of $\sigma_Z = 6.4$ a.u. is related via the curvature of the Liquid-Drop Model Potential Energy Surface (LDM-PES) to the excitation energy of the mean parent nucleus at the fission barrier [13]. With a fission barrier of 4 MeV, an energy above ground state of (58 ± 10) MeV is obtained, allowing for an emission of 6 neutrons. Adding these neutrons emitted to the neutron number of the mean pair of FF a neutron number of $N_0 = (130\pm1)$ follows for the mean parent nucleus. For high-energy symmetric fission ^{220}Th is the mean parent nucleus reconstructed from the isotopic distribution of FF. At very large asymmetries of the FF, N1/N2 < 22/102, the cross sections pass through a minimum and increase slightly for most extreme asymmetries. This was seen before [14] and is explained by the Businaro-Gallone peak in the LDM-PES beyond which fission barriers decrease again.

For the complete set of FF produced in the reaction, we obtain for each element the mean neutron to proton ratio $\overline{N}/Z$ and the width of its isotopic distribution $\sigma_N^{Z=\mathrm{const}}$. The $\overline{N}/Z$ values are shown in Fig. 4, separated into low-energy asymmetric and high-energy symmetric fission. They describe the isospin dependence of the cross sections. The $\overline{N}/Z$ -ratio of the mean fission fragment ^{107}Rh, 1.38 (red line), is found to be smaller than for low-energy fission where $\overline{N}/Z = 1.53$.(blue dashed line)

High-energy fission shows increasing $\overline{N}/Z$ –ratios in the range Z = 34 to 56. The slope observed agrees with a charge-polarisation expected for a smooth LDM-PES showing no nuclear structure effects. A new finding is the rapid decrease of the mean neutron density for isotopes at higher asymmetries. Taking the mean parent nucleus as ^{220}Th and the most asymmetric pair observed as $Z_1/Z_2 = 16/74$, mean neutron numbers $N_1/N_2 = 19/99$ are reached that add up to 118 neutrons present in the FF. Twelve neutrons are lost that indicates a high excitation energy of about 100 MeV. This energy is distributed in the high-energy regime between the pair of FF in proportion to their masses. The heaviest elements lose up to ten neutrons, and beyond erbium, Z > 68, all isotopes observed are stable or proton-rich. Neutron-rich isotopes in the wings of the Z-distribution will come with very low cross sections for the higher elements. It is the small contribution of low-energy asymmetric fission of (105±10) mb which remains the main source of neutron-rich isotopes for elements in the range Z = 28-64 [15].



▲ Fig. 5: The mean c.m. velocity of fission fragments measured as a function of Z, the atomic number. The three lines: dashed $Z_0 = 88$, full $Z_0 = 90$, and dashed-dotted $Z_0 = 92$, are calculated assuming Coulomb repulsion with the radius constant r_0 being fixed by taking the measured value of the velocity for symmetric fission of ^{220}Th as normalisation.

Figure 5 shows the mean velocities of FF in the uranium-rest frame. A comparison with calculated velocities, indicates that a small number of elements in the range $Z_0 = (88\text{-}92)$ dominates the family of parent nuclei. The systematic measurement of kinetic energies of spallation EVR, is achieved for the first time using our experimental method. Their kinetic energies are very small, on the average about 2.9 MeV. In this energy range slowing down of heavy ions mainly proceeds by elastic collisions.

The complete data set presented here represents the largest number of elements (from nitrogen to uranium) and isotopes (about 1400) ever observed in a single nuclear physics experiment. It is a main step forward [9], which stands for itself. Our work is not finished here. Tasks to be undertaken in the near future are open:

1) The energy dependence of the cross sections is hardly known. Further measurements at lower energies are needed for the prototype reactions selected.

2) Our data should serve as the benchmark for simulation codes of the complex physics of spallation reactions with the final goal being the prediction of intermediate systems.

3) An innovative measuring technique giving more and more precise and complete results generates a better understanding of the underlying physics. A number of surprising results are already published and can be found in our website [1].■

About the author

Monique Bernas is a physicist in experimental nuclear physics at the IPN d'Orsay, ORSAY France, bernas@IPNO.IN2P3.FR. Her lines of research are transfer reactions and fission studies aiming at unknown neutron-rich isotopes. She was involved in first investigations of ^{68}Ni and in the discovery of 107 fission products among which was the doubly magic nucleus ^{78}Ni.

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▲ **Fig. 6:** Members of the research team taken at the GSI experiment in 1998.
On the top row from left to right: P. Armbruster, M. Bernas, C. Stéphan, A. Brünle, F. Rejmund, T. Enqvist.
Kneeling from left to right: K. Burkard, J. Benlliure, L. Tassan-Got.