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A new method to acquire nuclear fission data using heavy-ion reactions - a way to understand the fission phenomenon -

Contents of the announcement

- Using multi-nucleon transfer reactions with beams of heavy ions, nuclear data on 14 nuclei could be collected in one experiment
- Expect to improve the understanding of the nuclear fission in new regions of the nuclear chart

Through the joint research conducted by K. Nishio and K. Hirose (Advanced Science Research Center, Japan Atomic Energy Agency, hereafter referred to as JAEA), S. Chiba (Tokyo Institute of Technology), Y. Aritomo (Kindai University), important new data regarding the mass yield distributions¹⁾ of fission fragments produced in collisions between heavy ions, was obtained at JAEA. This measurement was made possible by the development of a new experimental technique exploiting multi-nucleon transfer reactions²⁾. Furthermore, a newly-developed dynamic model³⁾ of fission could successfully reproduce the measured data.

In the neutron-induced fission of actinide isotopes, a large number of nuclei are generated as fission fragments. The mass number yield distribution of fission fragments is related to nuclear reactor safety, since it constitutes important data to determine the decay heat⁴⁾ and the number of delayed neutrons⁵⁾ generated by the fissioning nucleus. In addition, knowledge of the fission-fragment mass distribution is needed in the case of transmutation of the long-lived minor actinide⁶⁾ (MA) nuclei via high-energy neutron-induced fission. Mass yields often cannot be measured directly because a sample target cannot be produced either due to lack of purity or due to the short MA half-life. Furthermore, high energy neutron-data is also experimentally difficult to obtain and rather scarce.

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In the present study, by irradiating a ²³²Th target with an ¹⁸O beam, accelerated at the JAEA Tandem Accelerator laboratory⁷, 14 different types of nuclei ranging from thorium to uranium were produced and could be studied in the same experiment. The mass yield distributions obtained in this work permit to deduce fission-fragment mass distribution in neutron-induced fission, for a corresponding neutron energy from 1 to 50 MeV. This technique will permit to collect nuclear data for an even larger number of additional actinides. Furthermore, by enabling the study of fission of neutron-rich nuclei, it is expect to lead to new developments in our understanding of nuclear fission in the region. The data were collected and published within the 2012-2015 project joint project "Comprehensive study of delayed-neutron yields for accurate evaluation of kinetics of high-burn up reactors".

Background

In the fission process, a variety of fission fragments are generated. The type and production probability of these isotopes determines the amount of decay heat generated by the used fuel after reactor shutdown and how it will evolve with time; it also affects the delayed-neutron yield which dominates the reactor dynamics during operation. In addition, in order to develop the technology to transmute long-lived MA and convert them into shorter-lived fission products, fission fragment distributions of neutron-induced MA fission are required, up to high neutron bombarding energies. Therefore, mass yield distributions constitute essential nuclear data for nuclear energy production. Neutron-induced fission data however is scarce, due to practical difficulties in producing actinide targets (because of short lifetimes, lack of availability and/or lack of isotopic purity). Furthermore, due to the difficulty of generating monochromatic high-energy neutron sources, the available data



Fig. 1 The principle of measuring the mass distribution of fission fragments via multi-nucleon transfer reaction. The ¹⁸O beam impinged on a ²³²Th target, to generate a compound nucleus ²³⁴Th by the transfer of 2 neutrons. By measuring the speed of the two fission fragments produced by the fission of the compound nucleus, the mass of the fragments can be determined.

are very limited. In the present study, we aim to solve the problem by irradiating a high-purity actinide target with an accelerated ¹⁸O beam, thereby generating a variety of actinides over a wide range of excitation energies, using multi-nucleon transfer reactions, and by studying their fission.

Research Method

A multi-nucleon transfer reaction is a process in which the heavy-ion beam and the target nucleus exchange in the reaction some of their constituent neutrons and protons. In the example of Fig. 1, the compound nucleus ²³⁴Th is generated after two neutrons are transferred from the ¹⁸O beam to the ²³²Th target. This kind of nuclear reaction is a statistical process in which different numbers of neutrons and protons can be transferred, and results in the production of a variety of compound nuclei. To obtain nuclear fission data for a variety of nuclei in the same experiment was never attempted before this work. The detector technology developed in this study served the purpose of identifying the compound nucleus produced in each reaction and to determine its mass distribution. The different light nuclei emitted in the reaction were identified using the ΔE -E Silicon detector telescopes⁸⁾ shown in the photograph of Fig. 2. By identifying the mass and proton number of the scattered light nuclei, it was possible to deduce the number of neutrons and protons transferred to the target and thus identify the compound nucleus. For example, in Fig. 1, the detection of ¹⁶O implies that ²³⁴Th was produced. To determine the mass number of the fission fragments generated in fission, time-of-flight analysis of the fission fragments kinematics was performed. For this reason, we have developed a position-sensitive multi-wire proportional counter⁹), to detect the fission fragments. The ²³⁴Th isotopes in the example of Fig. 1 can also be viewed as composite system made by a neutron absorbed by a ²³³Th nucleus. Such direct method of production is however unfeasible, due to the short half-life of ²³³Th (21.8 m). The present study is the first successful measurement of the fission-fragment mass distribution as a surrogate reaction.



Research results

The results obtained are shown in Fig. 3. As it can be seen, we succeeded in collecting data for 14 nuclei in one single experiment. Among these, in the case of 231,234 Th and for 234,256,236 Pa, no data was available prior to our measurement. Also, since in the experiment a wide range of compound-nucleus excitation energies were populated, we were able to investigate the excitation-energy dependence of the nuclear fission yield. From the point of view of the surrogate reaction, this is equivalent to the dependence on the incident neutron energy in neutron-induced fission. The spectra in Fig. 3 are arranged vertically according to the compound-nucleus excitation energy, which spans a range from near thermal neutron ~ 1MeV to approximately 50 MeV incident energy. Such range of excitation energy is one important advantage of the present experimental technique.

In our study, we also compared our measured data with theoretical calculations within the framework of the dynamic fission model. In this model, the time evolution of nuclear deformation is calculated to predict the mass of the fission fragments. As shown in Fig. 4, the shape of the fissioning nucleus affects the nuclear potential, especially at low excitation energies. The figure shown on the left corresponds to the energy felt by the nucleus when the excitation energy is high (corresponding for example to high incident neutron energy). If the excitation energy is low, however, the potential energy surface will change as shown by the figure on the right hand side. At low excitation energy, the energy needs to be corrected for the effects caused by the internal structure of the nucleus, in particular the level density of protons and neutrons energy levels in the nucleus (also known as shell structure). Such microscopic effects are incorporated and taken into account in the calculations. Based on this potential energy, the time evolution of the atomic nuclear shape was calculated using the Langevin's equation, and used to examine into which mass numbers each nucleus would divide. The results of these calculations, performed using the Monte Carlo method, are shown by the red curves of Fig. 3. These calculations incorporate the oscillatory motion of the nuclear shape caused by constituent protons and neutrons hitting the surface of nucleus. The evolution of the nuclear shape in the fission process advances along a mean trajectory, by largely fluctuated affected by the random forces, explaining the width of the fragment-mass distributions. As can be seen in Fig. 3, these calculations reproduce very well the distributions corresponding to less than 20 MeV equivalent neutron incident energy. This is the first time that such good agreement was obtained between data and mass yield calculations, which include the fundamental behavior of atomic nuclei.

Impact and Future Developments

In addition to the reaction using ²³²Th target nucleus, other measurements have and will be carried out with other high-purity actinide targets, such as ²³⁸U, ²³⁷Np, ²⁴³Am, ²⁴⁸Cm and ²⁴⁹Cf. By using this new experimental technique on a series of targets, not only we will collect all the nuclear data required for transmutation, but will also study the fission distributions of several unknown nuclei. By collecting data on several neutron-rich actinides, the fission properties of a new region of the nuclear chart will be studied. The theoretical model developed, which describes the nuclear fission process at a more fundamental level, independently of the nucleus or region of interest, is a highly versatile and useful model.



calculation based on the fluctuation-dissipation model, and reproduce the observed change from asymmetric to symmetric distributions as a function of excitation energy.



Fig. 4 Nuclear potential energy surface. The time evolution of the fission process in the fluctuation-dissipation model, represented by the continues line on the multi-dimensional surface. Random fluctuations (vibrations) can be observed around the average trajectories For high excitation energies (left) the nuclear fission proceeds towards a mass symmetric divide, while at low excitation (right), due to the shell structure of the nucleus, fission will follow the mass asymmetric route.

Article Information

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Glossary

1) Fission fragment mass yield distribution

When fission occurs, various types of nuclei are produced as fission products. The mass yield distribution is a plot of the yield of these nuclei as a function of mass number. It is usually normalized so that the sum of the yield is 200%.

2) Multi-nucleon transfer reaction

One of the nuclear reaction mechanisms that occur collisions between nuclei. By the exchange of protons and neutrons between the incident and target nuclei, different isotopes can be generated in such reaction. This kind of reaction permits to produce a variety of nuclei in a wide excitation-energy range.

3) Dynamical Model

The model developed in this study, which uses the equation of motions based on the fluctuation-dissipation theorem (Langevin equation). The fluctuation-dissipation theorem, which shows the relation between microscopic particles in thermal equilibrium, and the macroscopically observable motion, is a well known description of Brownian motion. The random fluctuations are described by the Einstein relation $g^2=\gamma T$, where g is the strength of the random force, γ the friction tensor, and T the temperature. This relation plays the role of a bridge between microscopic and macroscopic motion. In the fission model, microscopic motion refers to the motion of protons and neutrons in the nucleus, while macroscopic motion represents the temporal variation of the shape of the nucleus.

4) Decay heat

Decay heat is the heat released as a result of radioactive β-decay of fission products. In a nuclear reactor, even after shutdown the operation spent nuclear fuel continues to produce heat for a time dependent on the lifetime of the isotopes. The length of time and the amount of heat produced depends on the types and respective yields of the generated fission products.

5) Delayed neutron number

In some species of fission fragments generated by nuclear fission, some neutrons are emitted following beta decay, so called "delayed" neutrons. Half-life of these delayed-neutron emitting nuclei can be as long as 55 seconds. In nuclear reactors, delayed neutrons also contributed to maintain the criticality. However, unlike prompt neutrons, their longer timescale permits to keep the reactor in a safe subcritical state and to prevent the abrupt change in the reaction that would occur if only prompt neutrons were involved. Delayed neutrons grant a sufficient time margin to control the reactor and avoid super-criticality. The number and lifetime of delayed neutrons varies by the species and yield of the generated fission fragments.

6) Long-lived minor actinides

Transuranic actinides such as neptunium, americium and curium, with the exception of Plutonium, are usually referred to as minor actinides. Among these isotopes, ²³⁷Np, ²⁴¹Am and ²⁴³Am, which are produced in the combustion of the nuclear fuel in a reactor, are characterized by long half-lives and are hence called long-lived

minor actinides (LLMA). They constitute a major challenge in the disposal of nuclear waste. Nuclear transmutation is a technique proposed to diminish these LLMA via nuclear fission. Accelerator-driven subcritical reactors have also been proposed as a source of nuclear energy. The development of ADS (accelerator driven systems) is being carried out.

7) Tandem Accelerator

A high-voltage electrostatic accelerator in which by means of a charge exchange of the particles being accelerated, the same accelerating voltage is used twice. This versatile accelerator permits to supply a wide range of energies and ion species. Moreover, its features allow a precise control of the beam intensity and beam profile, which is very important for precise measurement in the field of nuclear research.



8) Silicon ΔE -E detector

The energy loss of charged particles in matter depends both on the mass and on the charge of the nuclei. The identification of different ion species carried out by measuring the partial (ΔE) and total (E) energy deposited by these ions in particle detectors is referred to as the ΔE -E method. In this study, we constructed a ΔE -E detector using silicon detectors with excellent energy resolution. In particular, we developed a 75µm-thick ΔE detector with good thickness uniformity, which permitted to clearly separate different oxygen isotopes.

9) Multi-wire proportional counter

A gas amplification detector for fission fragments. In this study we developed a 200×200 mm² detector with planar, wide electrodes. The electrodes consist of independent parallel wires, used to collect the electrons generated in gas multiplication. Two sets of perpendicular wires permit to record the position of the incident fission fragments.