Radiochemical study of sub-barrier fusion hindrance in the $^{19}\text{F} + ^{209}\text{Bi}$ reaction

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(Received December 22, 2009; accepted in final form November 12, 2010)

*Charged particle induced reaction / Fusion / Fission /
Cross sections / Excitation function / Gamma spectrometry*

Summary. Production cross sections of fission product ^{99}Mo produced in the reaction of $^{19}\text{F} + ^{209}\text{Bi}$ have been measured at the incident energies 135–83 MeV near and below the fusion barrier by radiochemical methods. The excitation function for the fusion-fission cross sections in this reaction were deduced down to 80 μb from the measured production cross sections. A systematic analysis of the fusion-fission excitation function shows that sub-barrier fusion hindrance in the heavy mass system $^{19}\text{F} + ^{209}\text{Bi}$ is different from that in the medium-heavy mass systems but similar to that in the medium-light mass system and in the heavy mass system. This work is the first radiochemical study of sub-barrier fusion hindrance in heavy mass systems.

1. Introduction

An unexpectedly steep falloff of fusion cross sections has recently been observed in heavy-ion fusion reactions at deep sub-barrier energies [1]. This unexpected steep falloff of fusion cross sections, the so-called sub-barrier fusion hindrance, was studied experimentally [2–8] and theoretically [9–12]. However, the underlying mechanism of sub-barrier fusion hindrance is still an open question. Sub-barrier fusion hindrance has been studied mainly in medium-heavy and -light mass systems [2–4, 6–8] because improvements in the instruments to detect fusion residues, for example, the Fragment Mass Analyzer [13], make it possible to determine the fusion-evaporation cross sections in sub-milli-barn level precisely. In contrast, only a limited number of experimental studies was carried out in heavy-mass systems in which fused nuclei decay predominantly by fission (fusion-fission) and not by neutron evaporation (fusion-evaporation reactions) [5].

In this work, fusion-fission cross sections for the $^{19}\text{F} + ^{209}\text{Bi}$ reaction at sub-barrier energies were determined by a radiochemical method in order to study sub-barrier fusion hindrance in heavy-mass systems.

2. Experiments

The ^{209}Bi targets with a thickness of 0.15–0.22 mg/cm^2 were irradiated with 135–83 MeV $^{19}\text{F}^{6+}$ ions using the 20 MV tandem accelerator at JAEA-Tokai. Beam current was collected by a Faraday cup and was monitored on-line by using a current integrator module equipment connected to a multi-channel scaler (MCS) which was controlled by a personal computer. The MCS data were recorded to calibrate beam fluctuations. The beam current was controlled to be approximately 300 nA. The irradiation time was changed from 30 min to 20 h according to beam energies of 135–83 MeV. Fig. 1 schematically shows the two types of irradiation setups which were installed in the Faraday cup. Fission fragments were collected in the backing and the catcher foil of 5.4 mg/cm^2 aluminum. The solid angle of the catcher and the backing foil in Fig. 1a is 99.7% for 4π geometry.

The irradiation setup in Fig. 1b was mainly used to measure the cross sections of some high-yield fission products from reactions at beam energies above 90.9 MeV by γ -ray spectrometry without chemical separation procedures. At

Fig. 1. Schematic view of the irradiation setups.

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lower beam energies intense activities of nuclides produced in the reactions between ^{19}F ions and nuclides in the catcher make it impossible to measure γ -rays from fission products with smaller cross sections. Similar irradiation setups have been used on a regular basis by radiochemical experiments. In contrast, the setup presented in Fig. 1a was used in this work to reduce the systematic errors at low beam energies. Both the setups in Fig. 1a and b were used to measure the cross sections of ^{99}Mo below beam energies of 90.8 MeV by radiochemical separations. The procedure of radiochemical separations of ^{99}Mo is based on that reported in Ref. [14]. After irradiation, the target, the backing and the catcher foil were dissolved in 6 M HCl solution involving 0.1 ml of Mo carrier (10 mg Mo/ml). The radiochemical separations of molybdenum (VI) from fission products mixtures were carried out by using ion-exchange techniques. Molybdenum (VI) was adsorbed on the anion exchange resin, Dowex1-X8 (50–100 mesh), from hydrochloric acids solutions of concentration 6 M. Resin washed with 1 ml of 6 M HCl, 5 ml of 0.1 M HCl and 2.5 ml of 3 M HN_4OH . Molybdenum was eluted with 10 ml of hot 6 M $\text{NH}_4\text{C}_2\text{H}_3\text{O}_2$. Additional decontamination of the Mo elute was carried out by an iron hydroxide scavenge. Precipitation of Mo with α -benzoin-oxime was applied to the preparation of samples for γ -ray spectrometry. The average chemical yield was 65%. Chemical yields of 49–80% were determined by a neutron activation analysis performed after the measurements of the fission product ^{99}Mo with a 66 h half-life, that is, after more than 35 d from the experiments at the tandem accelerator. Standards were prepared by infiltrating 0.1 ml of the Mo carrier solution to a paper filter and by drying it. The samples and the standards put in the same capsule were irradiated with neutron flux of approximately $5 \times 10^{13} \text{ n/cm}^2/\text{s}$ for duration of 30 s by using the JRR-3 PN-1 and PN-2 equipments at JAEA-Tokai. The chemical yields were obtained from the ratio of specific activity between the sample and the standard.

The measurements of the fission products as well as the samples and the standard irradiated with neutrons were carried out by a Ge detector whose detection efficiency and resolution for 140 keV photopeak of ^{99}Mo -Tc were 18.0% and 1.6 keV FWHM, respectively.

The cross section of fission products, σ , was calculated by the following equation,

$$\sigma = \frac{C_\gamma}{\varepsilon_c \varepsilon_\gamma I_\gamma N \phi (1 - e^{-\lambda T})}, \quad (1)$$

where C_γ is the counting rate of the photopeak area at the end of irradiation, ε_c the chemical yield, ε_γ the photopeak detection efficiency, I_γ the emission probability of the γ -ray, N the number of target atoms, ϕ the beam flux, λ the decay constant and T the irradiation time. The corrections for the growth and decay of parent-daughter nuclides in the β -decay chain were applied to the 140 keV photopeak of ^{99}Mo -Tc.

3. Results and discussion

The production cross sections of ^{99}Mo , $\sigma(^{99}\text{Mo})$, are listed in Table 1. The excitation function of $\sigma(^{99}\text{Mo})$ is shown in Fig. 2. The solid circles and gray squares indicate the

Table 1. Production cross sections of ^{99}Mo , $\sigma(^{99}\text{Mo})$, in the 114.4–82.8 MeV $^{19}\text{F} + ^{209}\text{Bi}$ reaction. Beam energy, E_{lab} , refers to that at the center of the target in the laboratory system. Type of irradiation setup corresponds to (a) or (b) in Fig. 1.

E_{lab} (MeV)	$\sigma(^{99}\text{Mo})$ (mb)	Type of irradiation setup
114.4 ± 0.5	26.9 ± 2.4	(b)
102.9 ± 0.5	14.3 ± 1.6	(b)
94.9 ± 0.5	4.65 ± 0.43	(b)
91.0 ± 0.5	1.54 ± 0.15	(b)
90.8 ± 0.2	1.77 ± 0.19	(a)
88.8 ± 0.2	0.727 ± 0.083	(a)
87.8 ± 0.2	0.490 ± 0.058	(a)
87.0 ± 0.5	0.361 ± 0.026	(b)
86.8 ± 0.2	0.252 ± 0.030	(a)
85.8 ± 0.2	0.0970 ± 0.0164	(a)
84.9 ± 0.5	0.0646 ± 0.0063	(b)
83.83 ± 0.2	0.0112 ± 0.0016	(a)
83.79 ± 0.2	0.00890 ± 0.00095	(a)
82.8 ± 0.2	0.00329 ± 0.00038	(a)

Fig. 2. Cross sections of fusion-fission and fission products ^{99}Mo as a function of incident beam energies in laboratory system. Open circles represent the fission cross sections measured by solid surface barrier detectors, taken from Ref. [16]. Solid circles and gray squares were obtained by using the respective irradiation setups in Fig. 1a,b.

present results obtained by using the respective irradiation setups in Fig. 1a and b. The energy loss in the catcher foil (5.4 mg/cm^2 aluminum) put on the upstream of beam as shown in Fig. 1b was calculated to be approximately 24 MeV for $E_{\text{lab}} = 85$ and 87 MeV [15]. Therefore, the systematic errors of the energy loss calculation probably result in the small shifts of the data (gray squares) towards lower energies compared with the data shown by solid circles. It is noted that the data obtained by the irradiation setup in Fig. 1a involve much smaller systematic errors due to incident beam energies than those obtained by that in Fig. 1b. Errors of incident beam energies in Table 1 represent calculated energy fluctuations in the targets that include the energy straggling due to passing through the target for the irradiation setup in Fig. 1a or the catcher and the target for that in Fig. 1b. It should be noted that the production cross sections of ^{99}Mo are determined in a wide range between 26.9–0.00329 mb.

Fig. 3 shows the mass yield curve at $E_{\text{lab}} = 115 \text{ MeV}$. A large number of data result in fractional cumulative and independent yields as shown by open circles in Fig. 3. The compound nucleus ^{228}U in heavy-ion induced fission $^{19}\text{F} +$

Fig. 3. Mass yield curves of fission products at $E_{\text{lab}} = 115$ MeV. The solid curve represents a mass yield curve which was obtained by fitting a Gaussian curve to the experimental cumulative yields (solid circles). Fractional cumulative and independent yields are shown by open circles. Width parameters of Gaussians are 13.1 u for the solid curve, 10.4 u for the dashed and 15.7 u for the dotted Gaussian.

^{209}Bi produces neutron-deficient fission products compared with those in the thermal-neutron induced fission of actinides, $^{233,235}\text{U}$ and ^{239}Pu . In the light mass region, the nuclides ^{95}Zr , ^{99}Mo , ^{103}Ru and ^{112}Pd provide complete cumulative yields which make it possible to provide the characteristics of the mass yield curve. In the fissioning system the fissioning nucleus splits into two mass-symmetric fragments [16, 17]. Therefore, the mass yield curve as shown by the solid curve in Fig. 3 was obtained by fitting a Gaussian curve to the complete cumulative yields:

$$\sigma = \frac{\sigma_f}{2\sqrt{2\pi}\sigma_A} \exp\left[-\frac{(A - A_p)^2}{2\sigma_A^2}\right], \quad (2)$$

where σ_f is the fission yield, A the fragment mass, A_p the most probable mass and σ_A the width. The fission cross section was determined to be 650 ± 68 mb. The obtained $A_p = 110.1 \pm 1.3$ u is reasonably explained by neutron emission from excited primary fragments [17, 18]. The obtained $\sigma_A = 13.1 \pm 1.3$ u is comparable to 14.0–15.4 for $^{16,18}\text{O} + ^{209}\text{Bi}$ [19]. The dashed and dotted curves represent the Gaussian curves with $\sigma_A = 10.4$ and 15.7 u that are normalized to the yields of ^{99}Mo . The curves give respective fission yields $\sigma_f = 636$ and 698 mb. From this analysis it was found that fission yield σ_f was nearly independent of the Gaussian parameters, not only of the width σ_A but also of the most probable mass A_p . The analysis was carried out for the complete cumulative yields not only at $E_{\text{lab}} = 115$ MeV but at $E_{\text{lab}} = 103, 95$ and 91 MeV. In addition, the widths of the mass yield curves are nearly independent of the incident beam energies in the heavy-ion induced fission of ^{209}Bi [19]. These facts show that fission cross sections σ_f can be obtained from experimental $\sigma(^{99}\text{Mo})$ by using the fractional yield of ^{99}Mo to the total fission yield, $\sigma(^{99}\text{Mo})/\sigma_f = 0.043$, which is obtained from the mass yield curves of solid line in Fig. 3. The fusion-fission cross sections in Fig. 2 were deduced from $\sigma(^{99}\text{Mo})$ and their uncertainty of the fractional yield was estimated to be approximately 11%.

The open circles in Fig. 2 represent the fission cross sections measured by silicon surface barrier detectors [16]. In the experiments using silicon surface barrier detectors, the

total fission cross sections are determined by integrating the different cross sections over all angles. Thus, in general, scattered beam ions with larger cross sections prevent precise measurements of fission fragment angular distributions with smaller cross sections at low energies. Such difficulties in the measurements probably causes a slightly faster decrease of fission cross sections below 91 MeV compared with the present one. It should be noted that in this work the excitation function for the fusion-fission reaction was determined down to nearly two orders of magnitude smaller than the data reported. In this reaction evaporation cross sections are expected to be much smaller than fission cross sections [20]. Therefore, the observed fission cross sections can be regarded as the fusion cross sections for $^{19}\text{F} + ^{209}\text{Bi}$.

The experimental data deviate from the calculation by a one-dimensional barrier penetration model [21] (solid curve) below $E_{\text{lab}} = 86$ MeV. The energy at the point of steep falloff is in good agreement with $V_{\text{KNS}} = 86.8$ MeV, which is the touching point of the projectile and the target nucleus for $^{19}\text{F} + ^{209}\text{Bi}$ estimated with the Krappe-Nix-Sierk potential. The energy V_{KNS} systematically predicts the threshold incident energy for deep sub-barrier fusion hindrance [12]. This suggests the fusion hindrance occurs in the present system. The systematic analysis which was used in Ref. [22] were applied for the present data to reveal the characteristics of fusion hindrance as follows.

Fig. 4a shows the logarithmic derivative of energy-weighted fusion cross section, L , as a function of the center-

Fig. 4. (a) Logarithmic derivative of fusion-fission cross section for $^{19}\text{F} + ^{209}\text{Bi}$. The solid circles were obtained from two data points and the open circles were derived from least-square fits to three points. The logarithmic derivative L_{CS} for a constant S factor is shown by the solid line. (b) S -factor representation for the same data with a value $\eta_0 = 55.7$ is shown.

of-mass energy E ,

$$L = \frac{d \ln(E\sigma_f)}{dE}. \quad (3)$$

The experimental logarithmic derivative (solid and open circles) increases around $E = 81$ MeV with decreasing incident energy but a saturation seems to occur below 79 MeV. The solid line in Fig. 4a represents the logarithmic derivative L_{CS} for a constant S -factor which corresponds to s -wave transmission for a pure point charge Coulomb potential [22]. The value of L_{CS} is expressed by the Sommerfeld parameter η as,

$$L_{CS} = \frac{\pi\eta}{E} = \frac{\pi Z_1 Z_2 e^2}{E^{3/2}} \sqrt{\frac{m_N}{2} \frac{A_1 A_2}{(A_1 + A_2)}}, \quad (4)$$

where Z_i , A_i ($i = 1$ and 2) and m_N are the atomic and mass numbers of the projectile and target nucleus and the nucleon mass. It is clear that the experimental L does not reach L_{CS} at $E = 76$ MeV.

In the reactions $^{58}\text{Ni} + ^{58}\text{Ni}$, $^{60}\text{Ni} + ^{89}\text{Y}$, $^{90}\text{Zr} + ^{89}\text{Y}$, $^{90}\text{Zr} + ^{90}\text{Zr}$ and $^{90}\text{Zr} + ^{92}\text{Zr}$, the experimental logarithmic derivative intersects the curve L_{CS} at E_s^{ref} . The energy E_s^{ref} is empirically obtained from the data in these reactions [22];

$$E_s^{\text{ref}} = 0.356 \left[Z_1 Z_2 \sqrt{\frac{A_1 A_2}{A_1 + A_2}} \right]. \quad (5)$$

At this energy the S -factor representation of the same data clearly exhibits a maximum [22]. However, the S -factor representation for $^{19}\text{F} + ^{209}\text{Bi}$ in Fig. 4b steadily increases with decreasing E and does not reach a maximum at $E_s^{\text{ref}} = 76.0$ MeV.

It becomes clear from the results of the systematic analysis that such observed behavior of the logarithmic derivative and the S -factor representation in the present system $^{19}\text{F} + ^{209}\text{Bi}$ is clearly different from that in the medium-heavy mass systems $^{58}\text{Ni} + ^{58}\text{Ni}$ and so on [1, 22], but is similar to that in the medium-light mass system ^{36}S , $^{48}\text{Ca} + ^{48}\text{Ca}$ [7, 8] and in the heavy mass system $^{16}\text{O} + ^{204,206}\text{Pb}$ [5]. Further experimental and theoretical studies are required for understanding different behavior of sub-barrier fusion hindrance between the systems.

4. Summary

In the $^{19}\text{F} + ^{209}\text{Bi}$ reaction, the production cross sections of ^{99}Mo were radiochemically determined to be 26.9–0.00329 mb at the incident beam energies 114.4–82.8 MeV in laboratory system. Fission cross sections deduced from the cross sections of ^{99}Mo were determined down to a limit which is nearly two orders of magnitude smaller than previously reported values.

The fission cross sections show a steep slope below the energy V_{KNS} at the touching point of the projectile and the target nucleus compared with the calculation by the one-dimensional barrier penetration model. The logarithmic derivative L increases and saturates below the L_{CS} value. The S -factor representation steadily increases down to $E_s^{\text{ref}} = 76$ MeV. Sub-barrier fusion hindrance in the heavy mass system $^{19}\text{F} + ^{209}\text{Bi}$ is different from that in the medium-heavy mass systems $^{58}\text{Ni} + ^{58}\text{Ni}$, and so on, but is similar to

that in the medium-light mass system ^{36}S , $^{48}\text{Ca} + ^{48}\text{Ca}$ and in the heavy mass system $^{16}\text{O} + ^{204,206}\text{Pb}$.

This work is the first radiochemical experiment to study sub-barrier fusion hindrance in heavy mass systems.

Acknowledgment. The authors wish to acknowledge the crew of the JAEA Tandem Accelerator for the accelerator operation. We are thankful to A. Toyoshima and Z. Li for their help for γ -ray spectrometry. We would like to thank C. L. Jiang for long term discussion about the fusion hindrance study.

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