第29卷 第7期
2005年7月

高 能 物 理 与 核 物 理
HIGH ENERGY PHYSICS AND NUCLEAR PHYSICS

Vol.29, No.7
Jul., 2005

Measurement of Cross-Section for ^{181}Hf(n,γ)^{182}Hf Reaction *

QIU Jiu-Zi[1,2;1)] YIN Xin-Yi[1] HE Ming[1] DONG Ke-Jun[1] ZHANG Chun-Hua[3] ZHANG Jin-Song[3] DENG Hui[3] CHEN Xi-Lin[1] JIANG Shan[1]

1 (Department of Nuclear Physics, China Institute of Atomic Energy, Beijing 102413, China)

2 (Chinese People's Armed Police Force Academy, Langfang 065000, China)

3 (Nuclear Power Institute of China, Chengdu 610005, China)

Abstract The reactor neutron cross-section for ^{181}Hf(n,γ)^{182}Hf reaction has been measured with the activation method at the heavy water research reactor of China Institute of Atomic Energy. Thermal ionization mass spectrometry was used to determine the ^{182}Hf/^{180}Hf and ^{181}Hf/^{180}Hf atom ratios of the irradiated sample. The reactor neutron cross-section for ^{181}Hf(n,γ)^{182}Hf reaction is 80.0 ± 5.6b, given by 14.80 ± 0.60b for the ^{180}Hf(n,γ)^{181}Hf reaction as a reference.

Key words reactor neutron cross-section, ^{181}Hf(n, γ)^{182}Hf reaction, activation method, thermal ionization mass spectrometry

1 Introduction

Knowledge of the reactor neutron cross-section for hafnium (Hf) is of great importance not only for the design and development of nuclear reactors[1] but also for the activation analysis and other theoretical as well as experimental studies concerning the interaction of neutrons with matter.

Previously, the reactor neutron cross-section for ^{181}Hf(n,γ)^{182}Hf reaction was estimated roughly only by J. Wing et al.[2] in 1961. J. Wing et al. exposed 300 mg HfO_2, enriched in ^{180}Hf to 93%, to high neutron flux of a materials testing reactor in September, 1956, for one-year irradiation. ^{182}Hf was produced by double neutron capture of ^{180}Hf in the reactor. Mass spectrometric analysis of the irradiated hafnium gave a ^{182}Hf/^{180}Hf atom ratio of 0.00147 ± 0.00001. The neutron capture cross-section of ^{181}Hf was estimated to be 40^{+40}_{-20}b, based on the cross-section value of 10 b for reactor neutron induced ^{180}Hf(n,γ)^{181}Hf reaction.

In this experiment, the reactor neutron cross-section for the ^{181}Hf(n,γ)^{182}Hf reaction was measured with the activation method at the heavy water research reactor of China Institute of Atomic Energy. After irradiation, the isotopic ratios of hafnium in sample were measured by a thermal ionization mass spectrometry (TIMS). The reactor neutron cross-section for ^{181}Hf (n,γ)^{182}Hf reaction was obtained from our data of atom ratios.

2 Formula for the cross-section calculation

In the process of sample irradiation, the number of ^{181}Hf atoms produced in sample can be calculated by Eq.(1).

$$N_1 = \frac{N_0 \varphi \sigma_1 (1 - e^{-\lambda t})}{\lambda}, \tag{1}$$

where N_1 is the number of ^{181}Hf atoms, N_0 the number of ^{180}Hf atoms in the sample before the irradiation, φ the neutron flux at the sample irradiation site, σ_1 the cross-section for ^{180}Hf (n,γ)^{181}Hf reaction, t the sample irradiation time, $\lambda = \lambda_1 + \varphi\sigma_2$, where $\lambda_1 = 1.893 \times 10^{-7} s^{-1}$ is the decay constant of ^{181}Hf and σ_2 is the cross-section for ^{181}Hf (n,γ)^{182}Hf reaction.

In the process of sample irradiation, the number of ^{182}Hf atoms produced from ^{181}Hf(n,γ)^{182}Hf reaction can be calculated by Eq.(2).

$$N_2 = \frac{N_0 \varphi^2 \sigma_1 \sigma_2}{\lambda^2} (\lambda t - 1 + e^{-\lambda t}), \tag{2}$$

Received 1 November 2004, Revised 17 January 2005

* Supported by National Natural Science Foundation of China(10490185)

1) E-mail: jiumm@163.com

where N_2 is the number of ^{182}Hf atoms produced from ^{181}Hf(n,γ)^{182}Hf reaction. Because the half-life of ^{182}Hf is about $(8.9 \pm 0.09) \times 10^6$a [3], the number of ^{182}Hf atoms decayed during irradiation is neglected. Here, we also do not take into account the decrease of the number of ^{182}Hf atoms via the ^{182}Hf (n,γ)^{183}Hf reaction.

From Eq.(1), we can get

$$\varphi = \frac{\lambda}{\sigma_1(1 - e^{-\lambda t})}\left(\frac{N_1}{N_0}\right), \quad (3)$$

From Eqs.(2) and (3), we can get

$$\sigma_2 = \frac{(1 - e^{-\lambda t})^2}{\lambda t + e^{-\lambda t} - 1}\left(\frac{N_2}{N_0}\right)\left(\frac{N_0}{N_1}\right)^2 \sigma_1, \quad (4)$$

In this experiment, $\varphi\sigma_2 \ll \lambda_1$, so $\lambda \approx \lambda_1$. If the ratios of N_2/N_0 and N_1/N_0 in sample are known at the moment of the sample irradiation completion, the reactor neutron cross-section of ^{181}Hf (n,γ)^{182}Hf reaction can be calculated using Eqs.(3) and (4) and $\lambda = \lambda_1 + \varphi\sigma_2$ by iteration method.

3 Experiment

3.1 Sample irradiation

In this experiment, 50 mg HfO_2, enriched in ^{180}Hf to 98.3%, was exposed to the high neutron flux of the heavy water research reactor of China Institute of Atomic Energy, in December 2002, for eighteen-day irradiation. The reactor neutron flux was about $4.54 \times 10^{13} n \cdot cm^{-2} \cdot s^{-1}$ at the sample irradiation site. In the reactor, ^{180}Hf may capture a neutron to produce ^{181}Hf, and the produced ^{181}Hf may capture another neutron to produce ^{182}Hf. Meanwhile, the produced ^{181}Hf decays with a half-life of 42.39 ± 0.06d [4].

3.2 Isotope ratios measurement

After a cooling time of 232 days, the sample was dissolved in 10 ml 40% HF and 10 ml 68% HNO_3 mixed solution. The solution was heated on a hot plate, and evaporated to near dryness, then 10 ml 40% HF solution was added and evaporated to dryness to obtain a HfF_4 sample. The sample was re-dissolved in 50 ml 1M HF solution for column separation. A 1ml sample solution was loaded onto an anion exchange column. The column was rinsed with 10 ml of 1 M HF. Hf was then eluted by 30 ml of 0.01 M HF-9 M HCl, while W and Ta retained on the column. Tracer experiments showed that the average chemical yield of Hf was greater than 95%, and the decontamination factors for W and Ta were larger than 1000. The Hf sample purified with this procedure was satisfactory for keeping the isobaric interferences from W and Ta minimized in TIMS determination of isotopic ratios of Hf.

Following the chemical purification of hafnium, the sample was isotopically analyzed for eight times by a Finnigan MAT 262 thermal ionization mass spectrometry[5] at Nuclear Power Institute of China. Mass 182 was corrected for isobaric interference from ^{182}W, using isotope ratio $^{182}W/^{183}W = 1.77$ in original HfO_2 sample. The average ^{182}Hf/^{180}Hf and ^{181}Hf/^{180}Hf atom ratios measured by TIMS were $(2.33 \pm 0.07) \times 10^{-6}$ and $(1.92 \pm 0.05) \times 10^{-5}$, respectively, on August 6th, 2003, the day of measurement. From the data we obtained the atom ratios of ^{182}Hf/^{180}Hf and ^{181}Hf/^{180}Hf at the moment of the sample irradiation completion were calculated to be $(2.33 \pm 0.07) \times 10^{-6}$ and $(8.53 \pm 0.23) \times 10^{-4}$, respectively.

4 Result

The reactor neutron cross-section for ^{181}Hf(n,γ)^{182}Hf reaction has been calculated to be 80.0 ± 5.6b using twice iteration from our data, in which the cross-section value of 14.80 ± 0.60b for the ^{180}Hf (n,γ)^{181}Hf reaction was used as a reference. The main sources of the uncertainties are due to the atom ratios of hafnium isotopes (5.0%), the cross-section for ^{180}Hf (n,γ)^{181}Hf reaction (4.0%), the neutron flux (2.0%) and other errors (2.0%). The total standard uncertainty of the result is 7.0%.

The reference value of 14.80 ± 0.60b for reactor neutron induced ^{180}Hf (n,γ)^{181}Hf reaction was calculated based on the recent literature values of 13.04 ± 0.47b and 32.4 ± 1.2b[4] for the thermal neutron cross-section and the resonance integral of this reaction, respectively, and the thermal over epithermal neutron flux ratio of 10 at the sample irradiation site of the reactor used in this work.

5 Discussion

The reactor neutron cross-section for ^{181}Hf(n,γ)^{182}Hf reaction has been measured at the heavy water research reactor of China Institute of Atomic Energy relative to the reference cross-section value of 14.80 ± 0.60b for the ^{180}Hf(n,γ)^{181}Hf reaction. Thermal ionization mass spectrometry is the most accurate technique available for quantitative analysis of isotope ratio. The previous value of 40^{+40}_{-20} b given by J. Wing et al.[2] was obtained relative to the reference cross-section value of 10 b for ^{180}Hf(n, γ)^{181}Hf reaction. If the reference cross-section value of 14.80 b for ^{180}Hf (n, γ)^{181}Hf reaction had been used by J. Wing et al, the value would be $59.2^{+59.2}_{-29.6}$ b, 26% lower than the present measurement 80.0±5.6b, but it is still well within the error limits.

The authors would like to express their thanks to Prof. TIAN Wei-Zhi for his direction in the present work.

References

1 Klochkov E P, Risovany V D. At. Energ. Rubezh., 1987, **10**: 12—15

2 Wing J, Swartz B A, Uizenga J R H. Phys. Rev., 1961, **123**: 1354—1355

3 Vockenhuber C, Oberli F, Bichler M et al. Phys. Rev. Lett., 2004, **93**: 172501-1—4

4 Cho H-J, Kobayashi K, Yamamoto S et al. Nucl. Instrum. Methods in Phys. Res., 2001, **A462**: 442—450

5 Ayoub A S, McGaww B A, Midwood A J. Talanta, 2002, **57**: 405—413

^{181}Hf(n,γ)^{182}Hf反应截面测量*

仇九子[1,2;1)]　寅新艺[1]　何明[1]　董克君[1]　张春华[3]
张劲松[3]　邓辉[3]　陈细林[1]　姜山[1]

1（中国原子能科学研究院核物理研究所　北京　102413）
2（中国人民武装警察部队学院　廊坊　065000）
3（中国核动力研究院　成都　610005）

摘要　在中国原子能科学研究院重水研究堆上用活化法对反应堆中子引起的^{181}Hf(n,γ)^{182}Hf反应截面进行了测量.反应堆中样品照射位置的热中子与超热中子的通量比值约为10.照射后样品中的^{182}Hf/^{180}Hf和^{181}Hf/^{180}Hf的原子数比值是用热电离质谱测量的.本工作以^{180}Hf(n,γ)^{181}Hf反应截面(14.80±0.60b)为标准,测得的反应堆中子引起的^{181}Hf (n,γ)^{182}Hf反应截面值为80.0±5.6b.

关键词　反应堆中子截面　^{181}Hf(n,γ)^{182}Hf反应　活化法　热电离质谱

2004－11－01 收稿,2005－01－17 收修改稿
* 国家自然科学基金(10490185)资助
1) E-mail: jiumm@163.com