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論文内容要旨

This paper presents experimental studies on fast neutron-induced fission spectra for ^{237}Np and ^{233}U. The representations of the present studies are organized in seven chapters. These are

Chapter 1 Introduction,
Chapter 2 Status of the Research on Prompt Fission Neutron Spectrum,
Chapter 3 General Consideration on Experiment of Fission Spectrum Measurement,
Chapter 4 Experimental Procedure,
Chapter 5 Data Analysis,
Chapter 6 Results and Discussion and
Chapter 7 Summary and Conclusion.

The summaries for each chapter are as follows:

Chapter 1 Introduction

In this chapter, the significance of nuclear energy and nuclear fission, the role of the prompt fission neutron spectrum (PFNS), the importance of ^{237}Np and ^{233}U and the outline of the present work are described.

Nuclear fission is the fundamental physical process underlying the nuclear energy industry. The production of power from controlled nuclear fission is the most important technical application of nuclear reactions at the present time.

The fission spectrum is the energy distribution of prompt fission neutrons. Since it is the source term in fission reactors, it has strong influence on the neutron spectra in reactors. Therefore, it is one of the important reactor parameters for neutronics calculations of nuclear fission reactors, and dominates the reactor performance, criticality (k), and reaction rate R (E). It is also important for deduction of the cross section and neutron spectra of inelastic scattering from the neutron emission spectra data. High-energy fission neutrons are important for the shielding of nuclear reactors.

^{237}Np is very significant in regard to management and transmutation of nuclear wastes, and the performance of high-burn up reactors. The importance of data related to ^{237}Np has increased considerably of late in connection with the transmutation problem of highly active and toxic transuranic isotopes, and for the modeling of the fission process. However, only a few experimental data with inadequate quality are available for the fission spectrum of ^{237}Np because of the experimental difficulties and almost non-existence of pure ^{237}Np samples.

^{233}U is expected as nuclear fuel for nuclear reactors utilizing ^{233}U-^{232}Th fuel cycle which have been acknowledged as an alternative to traditional U-Pu one, and will be useful in future owing to much higher contents of natural resource of thorium than uranium. Besides, it is the fuel for the light-water breeder reactor (LWBR). There are also few experimental data on fission spectra of ^{233}U induced by fast neutrons.

For the reasons mentioned above, accurate knowledge is required on the fission spectrum for ^{237}Np and ^{233}U. As noted in Chap.2, theoretical calculations can not provide definitive prediction of fission spectrum because of the uncertainties in the model parameters. For the reason, reliable experimental data on fission spectrum are required not only for establishments of practical database for neutronic calculations but also for the development of fission spectrum model.

In the present work, therefore,

1) the fission spectra of ^{237}Np and ^{233}U have been measured for 0.62 and 0.55 MeV neutrons, respectively, over wide energy range of fission neutrons with high statistical accuracy using the time-of-flight(TOF) method at the 4.5 MV Dynamitron accelerator facility of Tohoku University, and then
2) the results were compared with those of evaluated data for reactor calculations, theoretical models, semi-empirical systematics used for the data evaluation to assess the validity. Prior to the experiment,
3) experimental method was optimized to obtain reliable results with limited sample atoms contained in the sample.
4) The samples used for the measurements were a NpO_2 powder (19.9 g in ^{237}Np) and U-Al plates (2.28 g in ^{233}U), which will provide better results than fission chamber samples as discussed in Chapter 3.

Chapter 2 Status of the Research on Prompt Fission Neutron Spectrum

In this chapter, the present status of the research on fission neutron spectrum is reviewed from the experimental point of view. Moreover, some theoretical approaches viz. the Maxwellian spectrum, the Watt spectrum, Howerton-Doyas formula, Madland-Nix Model and Modified Madland-Nix Model are described. Data status of ^{237}Np and ^{233}U are also presented.

As described in sect. 2.3, the spectrum of prompt fission neutrons is very difficult to predict exactly by theoretical calculations alone, although various models have been proposed with considerable progresses. For this reason, numerous experiments have also been performed using various methods to investigate the prompt fission neutron spectrum. The data qualities of the previous experiments are not high enough to provide required accuracy as the practical data and the reference data for theoretical models. Furthermore, there are, still marked disagreement among experimental data and theoretical calculations beyond quoted uncertainty that demands further experimental studies with higher precision.

Chapter 3 General Consideration on Experiment of Fission Spectrum Measurement

This chapter deals with measurement techniques of neutron spectrum and types of samples for fission spectrum measurement. The experimental method to obtain high-quality fission spectrum data is discussed.

The fission spectrum is obtained by measuring the spectrum of neutrons emitted from a fission sample bombarded by neutrons. The method of fission spectrum measurement can be categorized into the TOF method and a spectrometer method. As a fission sample, a fission chamber is one possibility, and solid or powder samples can also be used. In the case of fission chamber sample, there are many perturbation sources because fission chamber is rather massive to include multiple fission foils to provide acceptable count rates. The combination of the pulsed-beam TOF method and solid fission samples is preferred to meet the requirement while each method has each own advantage and disadvantage.

Chapter 4 Experimental Procedure

This chapter presents experimental arrangements on the measurements for PFNS of ^{237}Np and ^{233}U.

Referring to the consideration in Chap. 3, the present measurements for ^{237}Np and ^{233}U were undertaken using samples of NpO_2 powder and ^{233}U-Al metal, respectively, and TOF method employing a pulsed neutron beam. The measurements were performed with the 4.5 MV Dynamitron accelerator of the Fast Neutron Laboratory (FNL) at Tohoku University as a pulsed neutron source (Section 4.2).

In the present experiment, it is required to measure fission neutron spectrum over a wide energy range from several hundred keV to 10 MeV or higher using strongly radioactive samples with a small amount. The experimental arrangement, therefore, was designed considering a compromise among a yield of fission neutrons, energy resolution and a signal-to-noise ratio (S/N). Tightly collimated neutron detector of NE213 was adopted for fission neutron detection with three- or four-parameter data acquisition system. The set of three- or four parameter data for TOF, pulse-height and n-γ pulse shape spectrum was accumulated event-by-event as a list data on a magnetic optical disk using data acquisition system (MPC-1600, Laboratory Equipment Inc.), This data acquisition enabled flexible data treatment after the experiment. The present experimental arrangement eliminated uncertainties in the data correction.

Chapter 5 Data Analysis

This chapter deals with deduction of energy spectrum from the multi-parameter data and data correction process. Care was taken for the data correction against background, detector efficiency, energy scale and for neutron transmission of filter materials.

The TOF spectra for the ^{237}Np sample with low bias and high bias [2 MeV (proton) bias] are shown in Fig. 1(a) and 1(b), respectively. Similarly, those for the ^{233}U sample with low bias and high bias [1.6 MeV (proton) bias] are depicted in Fig. 2(a) and 2(b).

Chapter 6 Results and Discussion

In this chapter, the results of prompt fission neutron spectra of ^{237}Np and ^{233}U are presented in comparison with other experimental data, evaluations and semi-empirical systematics. Data comparisons are made using best-fit parameters for Maxwell or Watt functions and by the ratio to these functions.

^{237}Np: The Maxwellian temperature T_m derived is 1.28 ± 0.04 MeV. The result of Watt spectrum fit with the present study is shown in Fig. 3(a). The best fit description for the Watt distribution is $C_w\, sinh\,(\sqrt{1.95E}\,)\; exp\,(-1.01E)$.

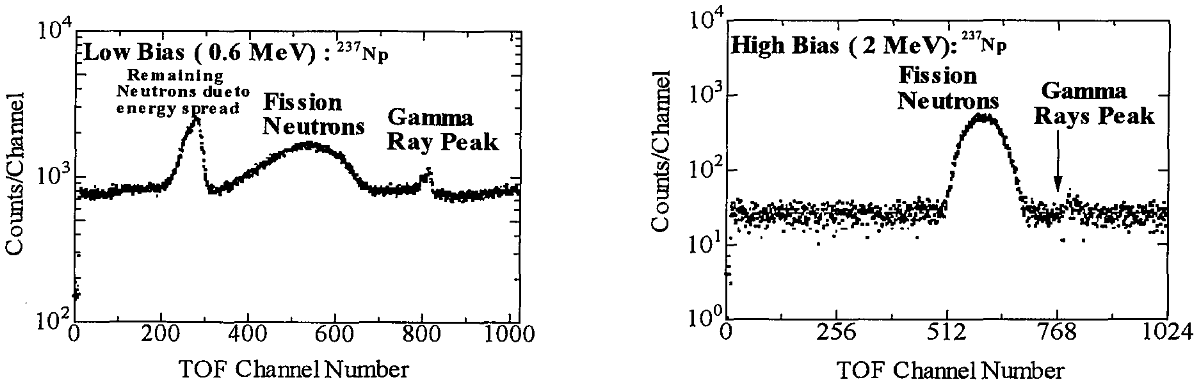
^{233}U: The Maxwellian temperature T_m derived for ^{233}U is 1.29 ± 0.04 MeV. The result of Watt spectrum fit is shown in Fig. 3(b). The best fit description for the Watt distribution is $C_w\, sinh\,(\sqrt{0.7E}\,)\; exp\,(-0.88E)$. Watt spectrum reproduces well the experimental values up to 10 MeV or so.

The present study provided neutron fission spectrum data up to12 MeV with high statistical accuracy. It should be noted that the present data for both ^{237}Np and ^{233}U are very close to Maxwell and Watt functions up to ~ 12 MeV, in contrast to previous data. The present data indicated that evaluated nuclear data showed markedly deviations from the experimental data even for the cases that employed most updated models. On the other hand, it was observed that the Howerton-Doyas formula predicts spectrum parameters very close to the experimental data of both nuclei studied by the present studies as well as major actinides.

The impact of the difference between the experiment and the evaluation on the reactor performance is also discussed.

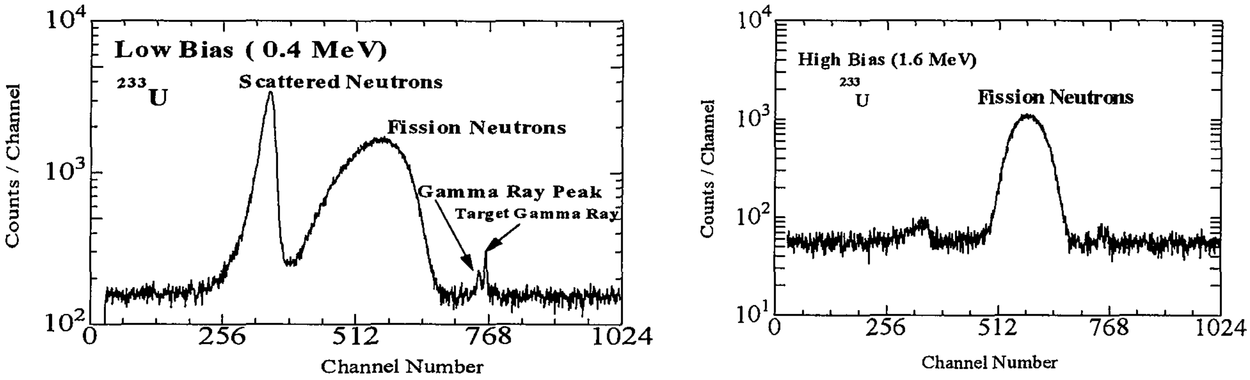
Chapter 7 Summary and Conclusion

In this chapter, the main conclusions of this study are summarized.



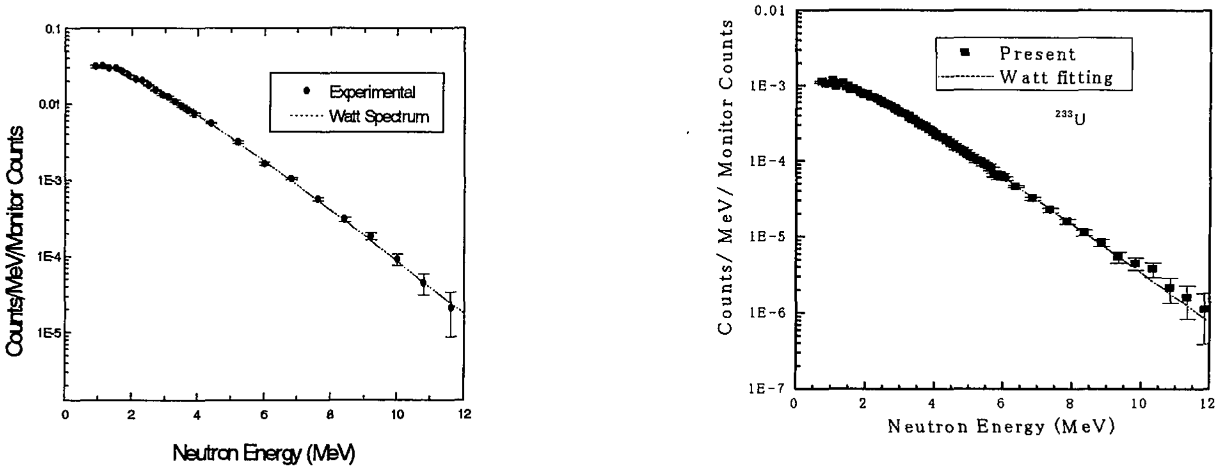
1(a) 1(b)

Fig.1 TOF spectra of the ^{237}Np sample (a) for low bias, and (b) for 2MeV (proton) bias.



2(a) 2(b)

Fig.2 TOF spectra of the ^{233}U sample (a) for low bias, and (b) for 1.6MeV (proton) bias.



3(a) 3(b)

Fig.3 Watt spectrum fit to the experimental fission spectrum (a) of ^{237}Np, and (b) of ^{233}U.

審査の結果の要旨

原子核分裂反応の際に生成する核分裂即発中性子のエネルギースペクトルは原子炉システム、特に高速炉システムの中性子スペクトルを定める上で極めて重要であるが、^{235}U、^{239}Pu 等の主要な核を除けば、試料入手の困難さ等の理由で測定データは非常に少なく、マイナーアクチニド核種の消滅処理に対して最も重要な ^{237}Np に対しては、1990 年代に入りロシアより２、３の測定例が報告されたのみであり、またトリウム核燃料サイクルの主要核種である ^{233}U については高速中性子核分裂によるものは 1980 年以前の１、２の例が報告されているのみである。本論文は東北大学工学研究科ダイナミトロン加速器を高速中性子源として ^{233}Np と ^{233}U について飛行時間法（以下 TOF 法）を用いて精度の良い実験を行い、その実験結果をこれまでに提案されている理論的モデルと比較してモデルの適用性を論じたもので、全編７章より成る。

第１章は序論であり、核分裂即発中性子スペクトルの重要性について述べている。

第２章では、核分裂即発中性子スペクトル研究の現状を、実験的研究、理論的研究の両面から調査し、問題点を述べている。

第３章では核分裂即発中性子スペクトルの測定手法を検討し、固体試料を用い、TOF 法によることが適切であるとしている。

第４章では採用した実験の手法について詳細に述べている ^{237}Np。については円柱形状の二重ステンレス管に封入された NpO_2(Np 19.99g を含む）を用い、0.62MeV の入射中性子に対し、0.8MeV から 12MeV の間の即発中性子スペクトルを、また ^{233}U については U_3O_8 粉末をアルミニウムに分散させ 12mmx12mm の正方形状の平板とした試料を重ねあわせた角柱状試料（^{233}U 量 2.28g）を用いて、入射エネルギー 0.55 MeV に対して同じく 0.8 から 12MeV の間の即発中性子スペクトルを取得している。これらは外国の例に比し少ない量の試料しか使用できないという不利を周到な実験配置により克服して高い精度のデータを得た点で重要な成果である。

第５章ではデータ解析において用いた種々の補正法につき詳細に記述している。

第６章では得られた結果を理論的研究より与えられたスペクトルと比較し、Maxwell スペクトルは 10 MeV 以上の領域で実験値との一致が悪くなるが、Watt スペクトルでは全エネルギー領域にわたって良く記述できることを述べ、また Maxwell 温度（T_m）として ^{237}Np については 1.28±0.04MeV、^{233}U については 1.29±0.04MeV という値を得ている。これらは概ねこれまでに報告されている実験値に近く、^{237}Np については、我が国の標準的な核データライブラリーである JENDL3.2 に採用されている T_m=1.38MeV という値は高すぎること、^{233}U についても実験値と形状が異なることを指摘している。これらは核データ評価上重要な指摘である。また両核種に対する結果が、核分裂即発中性子数と T_m とを関係つける Howerton-Doyas の公式から得られる値とほぼ一致することを示している。

以上要するに本論文は、実験データの乏しい ^{237}Np と ^{233}U の核分裂中性子データを、少ない試料しか利用できないという悪条件を克服して高い精度で測定することに成功したものであって、量子エネルギー工学、特に原子炉物理学の進展に寄与するところが少なくない。

よって、本論文は博士（工学）の学位論文として合格と認める。