PROCEEDINGS SERIES

PHYSICS OF FAST AND INTERMEDIATE REACTORS

I

PROCEEDINGS OF THE SEMINAR
ON THE PHYSICS OF FAST AND INTERMEDIATE REACTORS
SPONSORED BY
THE INTERNATIONAL ATOMIC ENERGY AGENCY
AND HELD IN VIENNA, 3 — 11 AUGUST 1961

INTERNATIONAL ATOMIC ENERGY AGENCY
VIENNA 1962

PHYSICS OF FAST AND INTERMEDIATE REACTORS, IAEA, VIENNA, 1962
STI/PUB/49

FOREWORD

It is generally agreed that the ultimate economic advantage of power produced by nuclear fission over that produced by conventional sources depends on the ability of a certain type of reactor to breed precious nuclear fuel out of the plentiful but not readily fissionable isotope of uranium. This fact is mainly responsible for the importance attached to the development of fast power reactors, but many other interesting properties of unmoderated or weakly moderated reactor systems have also been brought to light by reactor physicists.

In August 1961 the Agency organized in Vienna a Seminar on the Physics of Fast and Intermediate Reactors, at which all the topics relating to this important branch of reactor science were discussed. The main feature of this meeting was extensive discussion of the 66 written contributions, which set the stage for a wide exchange of experience and ideas throughout 13 half-day sessions. The Seminar was attended by 132 scientists from 22 Member States and two international organizations.

It is hoped that these Proceedings of the Seminar, which include both the papers presented and a record of the discussions, will be useful as a reference work both to research workers in the field and to newcomers to it for many years to come. The Agency's thanks are due to all the participating scientists for their written or oral contributions and especially to those among them who, as session chairmen, led the discussions and contributed greatly to the success of the meeting.

March 1962

Scientific Secretary
Seminar on the Physics of Fast
and Intermediate Reactors

EDITORIAL NOTE

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INTRODUCTION

During the Seminar, sixty-five papers were orally presented, and seven more were accepted for publication in the Proceedings. In order that these Proceedings might be in the hands of their users at an early date, the method of presentation of the papers and of the extensive session discussions had to be somewhat different from the one usually followed. The complete record of the sessions will be found at the end of Volume III.

The order in which the papers are presented here is not in all cases that of their presentation during the Seminar. Changes have been made where it was considered that these would enhance the usefulness of these volumes as reference books. The subject grouping adopted is given below.

Volume I

I. *Neutron Physics*

- I.1. Data requirements
- I.2. Cross-section measurements
- I.3. Fission properties
- I.4. Nuclear theory
- I.5. Multi-group cross-sections

II. *Integral Experiments*

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1. DATA REQUIREMENTS

NUCLEAR DATA REQUIREMENTS FOR FAST AND INTERMEDIATE REACTOR CALCULATIONS

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Abstract — Résumé — Аннотация — Resumen

Nuclear data requirements for fast- and intermediate-reactor calculations. During 1960/61, some work has been done at Karlsruhe in the compilation of reliable nuclear data for fast- and intermediate-reactor calculations. Materials included thus far are He, O^{16}, Na^{23}, Cr, Fe, Ni, Mo, U^{235}, U^{238} and Pu^{239}.

For fast- and intermediate-reactor optimization studies, reliable theoretical prediction of space-dependent neutron energy spectra, critical masses, breeding ratios, burn-up, Doppler coefficients and related subjects, the author was interested in much more detailed microscopic cross-section data—concerning mainly neutron absorption and fission—than are needed, for example, for the evaluation of few-group constants in the conventional multi-group diffusion theory.

In spite of much progress in the experimental determination and theoretical interpretation of nuclear data, many inconsistencies in the experimental results of different authors and laboratories and large gaps in experimental nuclear-data work still remain. This paper discusses a great number of these gaps and inconsistencies in respect of the above-mentioned nuclei.

Constantes nucléaires requises pour les calculs de réacteurs à neutrons rapides et à neutrons intermédiaires. En 1960/61, des travaux ont été exécutés à Karlsruhe en vue de réunir des données nucléaires sûres pour les calculs de réacteurs à neutrons rapides et à neutrons intermédiaires. Les matières étudiées jusqu'à présent sont: He, ^{16}O, ^{23}Na, Cr, Fe, Ni, Mo, ^{235}U, ^{238}U et ^{239}Pu.

Pour l'établissement des conditions optima concernant les réacteurs à neutrons rapides et à neutrons intermédiaires, pour calculer avec précision la distribution spatiale des flux de neutrons, les masses critiques, les taux de surgénération, la combustion, les coefficients Doppler et les éléments connexes, l'auteur avait besoin de données beaucoup plus détaillées sur les sections efficaces microscopiques — notamment en ce qui concerne l'absorption et la fission des neutrons — que, par exemple, pour l'évaluation des constantes fondées sur l'emploi de quelques groupes dans la théorie classique de la diffusion à plusieurs groupes.

En dépit de grands progrès dans la détermination expérimentale et l'interprétation théorique des données nucléaires, on constate encore des contradictions dans les résultats des expériences de divers auteurs et laboratoires, et il existe toujours de graves lacunes dans les travaux sur les données nucléaires. L'auteur traite d'un grand nombre de ces lacunes et contradictions en ce qui concerne les éléments mentionnés plus haut.

Требования в отношении ядерных данных для расчетов реакторов на быстрых и промежуточных нейтронах. В течение 1960/61 годов в Карлсруэ была проведена некоторая работа по компиляции надежных ядерных данных для расчетов реакторов на быстрых и промежуточных нейтронах. К материалам, которые пока вошли в компиляцию, относятся гелий, кислород-16, натрий-23, хром, железо, никель, молибден, уран-235, уран-238 и плутоний-239.

Для осуществления успешных исследований реакторов на быстрых и промежуточных нейтронах, для надежного теоретического предсказания пространственной зависимости спектров нейтронной энергии, критических масс, коэффициентов воспроизводства, выгорания, коэффициентов Допплера и относящихся к ним проблем автор данного доклада

заинтересован в получении более подробных данных о микроскопических поперечных сечениях, касающихся главным образом поглощения и деления нейтронов, чем требуется, например, для оценки констант небольшего количества групп в обычной многогрупповой теории диффузии.

Несмотря на значительный прогресс в экспериментальном определении и теоретическом толковании ядерных данных, по-прежнему имеют место многочисленные расхождения в экспериментальных результатах различных авторов и лабораторий и большие пробелы в экспериментальной работе по получению ядерных данных. В настоящем докладе рассматривается целый ряд этих пробелов и расхождений, касающихся ядер вышеупомянутых материалов.

Datos nucleares necesarios para los cálculos de reactores rápidos e intermedios. En los años 1960 y 1961 se ha estado trabajando en Karlsruhe en la compilación de datos nucleares fidedignos que se utilizan en los cálculos de reactores de neutrones rápidos e intermedios. Los elementos estudiados hasta el presente comprenden: He, ^{16}O, ^{23}Na, Cr, Fe, Ni, Mo, ^{235}U, ^{238}U y ^{239}Pu.

Para establecer los parámetros óptimos de los reactores rápidos e intermedios, y para poder predecir teóricamente con suficiente precisión los espectros de las energías neutrónicas en función del espacio, las masas críticas, las razones de reproducción, el grado de combustión, los coeficientes Doppler, etc., se requieren datos mucho más detallados sobre las secciones eficaces microscópicas —especialmente sobre la absorción neutrónica y la fisión— que, por ejemplo, para evaluar las constantes de algunos grupos en la teoría clásica de difusión de grupos múltiples.

A pesar de los notables progresos realizados en la determinación experimental y en la interpretación teórica de los datos nucleares, siguen existiendo todavía muchas discrepancias entre los resultados experimentales dados a conocer por distintos autores y laboratorios; asimismo, subsisten grandes lagunas en los datos nucleares. En la memoria se examina toda una serie de esas lagunas y contradicciones en relación con los núclidos mencionados más arriba.

Introduction

A crucial point in the physical design of fast and intermediate reactors is the exact knowledge of the nuclear data of the fissionable, breeding, structural and cooling materials involved. Design parameters such as space-dependent neutron energy spectra, reactor material composition, critical masses and radii, breeding ratios, burn-up, and Doppler-coefficients are more or less strongly affected by the energy-dependence of the neutron cross-sections of the reactor materials chosen. The fast-reactor theory [1, 2] has been developed sufficiently to allow accurate prediction of these design parameters, provided that the nuclear data involved in the theory are accurately known.

As to the Karlsruhe fast power-breeder reactor project, two principal possibilities of cooling are considered. The first is the almost classical one, i.e. Na-cooling, the other one, He-cooling, being somewhat unusual in connection with fast power-breeder reactors. Principal investigations have shown that He-cooling is sufficient to meet the requirements of a fast power-breeder cooling material. Large-scale experiments for He-cooling are being prepared. For optimization studies regarding these two principal types of oxide power-breeder reactor, we have made a compilation of all neutron nuclear data in respect of He, O^{16}, Na^{23}, Cr, Fe, Ni, Mo, U^{235}, U^{238} and Pu^{239}, in the energy region between 1 eV and 10 MeV. We have taken account of all available pertinent literature. A report about this work, including extensive microscopic cross-section tables, graphs, and a detailed description of the compilation procedures used, is in preparation and will soon be published as an external Karlsruhe report.

Unfortunately, in spite of much progress in the experimental determination and theoretical interpretation of nuclear data since the 1958 Geneva Conference, many inconsistencies in the experimental results of different authors and laboratories and gaps in experimental nuclear data work still remain. In the following sections I should like to discuss some striking discrepancies and gaps in the nuclear data of the above-mentioned nuclei.

The consideration of these and other discrepancies and gaps has led us to the decision to install a pulsed 3-MeV Van de Graaff for nuclear-data measurements at Karlsruhe.

1. Inelastic neutron scattering

In regard to inelastic neutron scattering, which is mainly responsible for the neutron-energy distributions in unmoderated reactors, large gaps still remain to be filled.

To begin with, let me quote briefly the present status as to inelastic scattering in O^{16}, Na^{23}, U^{235}, U^{238} and Pu^{239}. By use of the time-of-flight method, CRANBERG and LEVIN [3—5] at LASL (the Los Alamos Scientific Laboratory) measured inelastic excitation cross-sections of U^{235}, U^{238}, and Pu^{239} at 550, 980 and 2000 keV, and for some U^{238}-levels also at 1500 and 1893 keV. SMITH [6] at ANL (Argonne National Laboratory) measured the inelastic excitation cross-sections of the two lowest levels of U^{238} at 44 and 146 keV from about 0.5 to 1.5 MeV. The gamma-ray detection method has successfully been applied to the lowest levels of many nuclei, e.g. by HALL and BONNER [7] at the Rice Institute to excitation cross-sections of some O^{16}-levels below 9.8 MeV, by FREEMAN and co-workers [8] at Harwell, and LIND and DAY [9] at LASL to the inelastic excitation of Na-levels up to about 3 MeV. In particular, the work of Cranberg, whose results have been confirmed by the measurements of Smith [6], has clearly shown the superiority in resolution of the pulsed time-of-flight method in comparison to the earlier spherical shell transmission measurements with threshold detectors [10—12].

Unfortunately, the range of applicability of the gamma-ray detection method and, as Cranberg discusses in detail in his recent paper [5], the reliability of the pulsed time-of-flight method are restricted to regions of low-lying and well-separated target excitation levels, say, to a few MeV at the most, the upper energy-limit depending on the size of the target nuclei. Above this limit the excited nuclear levels get so close to each other that they cannot be separated any more by the methods just mentioned. As to the time-of-flight technique applied to the heaviest nuclei, it also becomes difficult at higher neutron incident energies to discriminate between elastic scattering and inelastic scattering leading to low-lying excitation levels. Therefore, above a few MeV only measurements of the total inelastic scattering cross-section or of the non-elastic cross-section may be made; experimental information as to non-elastic cross-sections in the higher MeV-region remains rather scarce. This is particularly true of the U- and Pu-isotopes: for example, above 2 MeV nothing is known experimentally about the inelastic scattering of common fissionable isotopes such as U^{235} and Pu^{239}; for U^{238} only four not sufficiently reliable σ_X-measurements at 2.5, 4, 6 and 7 MeV [13, 14] have been made; between about 4 and 10 MeV no $\sigma_{n,n'}$ or σ_X-measurements are available for Na; for Cr and Mo similar gaps in the inelastic scattering data in the higher MeV-region exist.

Some of these gaps and experimental impossibilities have been filled up efficiently by theoretical calculations: for single-level excitation cross-sections by means of the statistical HAUSER-FESHBACH theory [24] and, for example, for the cross-section of compound nucleus formation, by application of the optical model with improved nuclear potentials in adapting theoretical parameters to the few experimental results. To quote only some work concerning the heaviest nuclei, such semi-empirical Hauser-Feshbach calculations as those done by DRESNER [15, 23] for U^{238}; RAE, MARGOLIS and TROUBETZKOY [16] for U^{235}; and MOLDAUER [17] for U^{235}, U^{238} and Pu^{239}, partly in addition to the measurements, furnished useful results with regard to excitation cross-sections for some individual low-lying levels or groups of levels of the above-mentioned nuclei, useful mainly in those energy regions where it is very difficult to do measurements. This is particularly true of the energy-dependence of the excitation cross-sections pertaining to the lowest excited states of U^{235} and Pu^{239} below about 100 keV.

Above about 7 MeV, according to the work of BJORKLUND and FERNBACH [18—20], the optical model of the nucleus with nuclear surface-absorption and inclusion of a spin-orbit coupling term of the Thomas-type in the nuclear potential is able to reproduce reasonably well the few experimentally determined non-elastic cross-sections. Adapting the theoretical parameters to the few measured cross-section values permits fairly good prediction, within the limits of about $\pm 10\%$, of unknown non-elastic cross-sections of the same nucleus or by nuclear systematics of the neighbouring nuclei, which in the case of isotopes without magic neutron number are nearly identical with the compound nucleus formation cross-sections [21, 22].

Therefore, mainly with regard to the already-mentioned nuclei, an intermediate energy range between about 2 and 7 MeV remains, where neither enough measurements nor reliable theoretical predictions exist. According to this lack of data, and because neutron-energy distributions in fast reactors are still rather sensitive to inelastic slowing-down from these energies, and for better insight into the range of applicability and further improvement of the optical model, there is great need for measuring the total inelastic scattering cross-section at more points between 2 and about 10 MeV. Also, with regard to calculations of fast-reactor neutron-spectra, further determination of nuclear excitation levels and

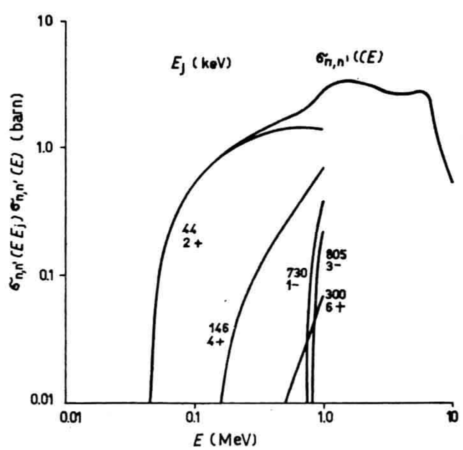

Fig. 1
Inelastic excitation cross-sections and total inelastic scattering cross-section of U^{238} between threshold and 10 MeV.

inelastic excitation cross-sections, and the assignment of spins and parities to these and other already known levels are particularly needed for the heavy nuclei such as U^{235}, U^{238} and Pu^{239} below about 2 MeV, and for the medium-weight and lighter nuclei such as Na, Cr, Ni and Mo below about 3 to 5 MeV.

Fig. 1 shows the inelastic scattering cross-section of U^{238}. Combined are the excitation cross-sections for levels up to 1 MeV as calculated by Dresner [15, 23] and the available experimental and theoretical information about non-elastic cross-sections in the higher MeV region [13, 14, 31].

With regard to neutron-energy spectra in fast reactors it is also important to know the energy distributions of neutrons inelastically scattered in the higher MeV region, where it is impossible to distinguish between different single levels on account of the small level-distance. I should like to mention here only some of the main features. The results of the theory of nuclear evaporation [25—29] show rather good agreement with the experimentally determined spectra of inelastically scattered neutrons. Unfortunately, there are also gaps and inaccuracies in this experimental work. To quote only some of the main gaps: for the fissionable isotopes U^{235} and Pu^{239} no results of measurements of inelastically scattered neutron-spectra have as yet been reported; for U^{238} only three such spectra have been measured and nuclear temperatures assigned: at 2.45 MeV by Cranberg [3], and at 2.5 and 3.5 MeV by Fetisov [30] (see also [31]). Similar gaps exist for Fe (only one preliminary temperature value has been reported [32, 33]), Cr, Ni and Mo. For a proper adjustment of nuclear temperatures so as to apply successfully the evaporation theory to the spectra of inelastically scattered neutrons, it would be interesting to measure inelastically scattered neutron distributions at several energies: for the heavy elements, mainly U^{235} and Pu^{239}, and also U^{238}, above 2 MeV; for medium-weight structural nuclei, such as Cr, Ni, Fe and Mo, above 3—5 MeV; and finally for Na, above about 4 MeV.

2. Neutron absorption

In fast and epithermal breeder-reactor design, an exact knowledge of the energy-dependence of neutron-absorption cross-sections, such as σ_{γ}, σ_{p}, is required for:

(*a*) Liquid-metal coolants, such as Na, in order to determine exactly the cooling-circuit activation for the purpose of shield-design;

(*b*) Breeding and fissionable materials, such as U^{235}, U^{238}, and Pu^{239}, for reliable evaluation of critical reactor dimensions, critical masses of fissionable material, and breeding ratios;

(*c*) Structural materials, such as Fe, Ni, Cr and Mo, for judging suitably the influence of parasitic neutron-absorption on critical masses and on reactor neutron economy; and

(*d*) Fission products, for judging the long-term reactivity behaviour in reactors with high burn-up.

Between 1 and 10 MeV only very few measurements of neutron radiative capture cross-sections have been made. Also, in this energy region, at least for medium-weight structural nuclei, the neutron-absorption by the (n, p) process is considerably higher than by radiative capture: there are still gaps to be filled by further measurements, particularly for Ni^{58} between the threshold at 0.39 MeV [34] and 1 MeV, and also between 3.6 MeV and about 10 MeV. Among the naturally occurring isotopes of the three neighbouring elements Fe, Ni and Cr, Ni^{58}—which, with an isotopic fraction of 67.76%, is the most abundant of the stable Ni-isotopes—has the lowest threshold and the highest probability of occurrence of the (n, p) process. To quote an example for the influence of the Ni^{58} (n, p) process on critical masses, the replacement of stainless steel (only about 8% Ni) by inconel (about 77% Ni) as structural material enlarges the critical masses of highly enriched fast reactors with hard neutron-energy spectra.

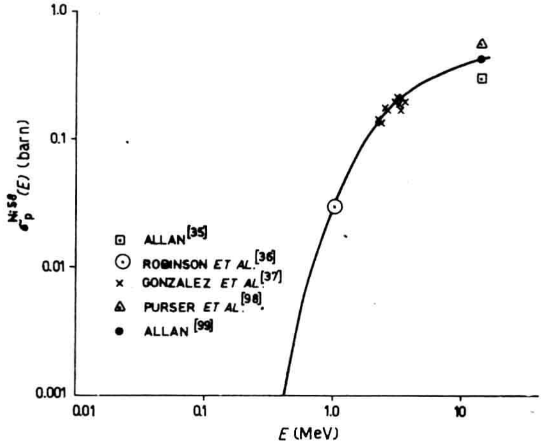

Fig. 2
σ_p (Ni^{58}) from threshold to about 15 MeV.

Fig. 2 shows the energy-dependence of σ_p (Ni^{58}) between threshold and about 15 MeV obtained by extrapolation from available measurements [35—37, 98, 99]. Fortunately, the bulk of the neutrons in a true fast power-breeder reactor have, in general, energies below 1 MeV; furthermore, one can, with regard to the radiative capture cross-sections of many nuclei, theoretically predict with rather good reliability a $1/E$-behaviour above 1 MeV by combining results of the statistical theory of nuclear reactions with the few existing measurements [17, 38]. Finally, above 1 MeV neutron-absorption by any of the mentioned absorption processes, mainly in the fissionable and fertile isotopes, is much less probable than inelastic scattering or fission, and thus does not influence fast-reactor neutron economy very much.

The most important energy region of fast breeder neutrons ranges from about 1 keV to 1 MeV [1, 2]. Between about 25 keV and 1 MeV there are now many $\sigma_{n\gamma}$-measurements [39–50]. Unfortunately, for the same element the results from different measurements occasionally differ greatly, perhaps in some cases due to systematic errors in different experimental methods or, which seems to be more probable, those introduced by the use of different standard values. A typical example pertaining to this latter situation is that for molybdenum: one has two series of $\sigma_{n\gamma}$ (E)-measurements between about 100 keV and 1 MeV, one repeated several times with nearly equal results by DIVEN [41], the other one made by STAVISKII and SHAPAR [47], both differing consistently by about 50 to 100% at all energies in this region. Fig. 3 shows the experimental results for the radiative capture cross-section of molybdenum in the energy range between 1 keV and 1 MeV together with two curves, showing the energy-dependence of the radiative capture cross-section as assumed by OKRENT *et al.* [17]

(dashed curve) and by ourselves (dash-dotted curve). The experimental points shown up to 6 keV are fast-chopper data obtained by BLOCK and SLAUGHTER [54]; at 30 and 65 keV, data by GIBBONS *et al.* [55]. are plotted; between 50 and 1000 keV the Russian data [47] are shown above the most recent Diven

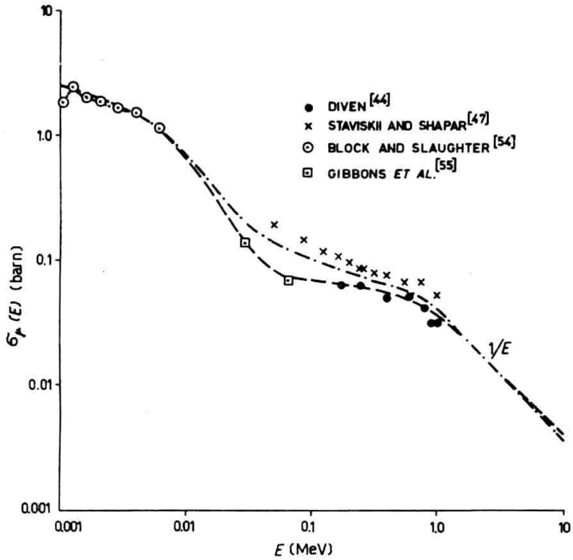

Fig. 3
Radiative capture cross-section of molybdenum between 1 keV and 10 MeV. The dashed curve and the dash-dotted curve refer respectively, to the energy-dependences assumed by Okrent *et al.* [17] and the author.

results [41]. The energy-dependence of the capture cross-section appears to be well established up to about 10 keV. Okrent *et al.* [17] drew a smooth curve through the data by Block, Gibbons, and Diven nearly without consideration of the Russian data, and arrived at an energy-dependence of the capture cross-section, which, from the viewpoint of nuclear reaction theory, can be rather well understood; the curve thus obtained shows the expected appearance of p-wave neutrons above several keV, of d- and f-wave neutrons above some 100 keV. Our own curve differs from that of Okrent *et al.* [17] essentially only in the energy region between 30 and about 1000 keV. There we have chosen a variation midway between the Diven and the Russian data, because we could not clearly decide whether the Russian standard-value of 97 ± 16 mb for σ^{Mo}_{γ} at about 200 keV is really too high or not. Some question remains as to the capture group-constants of molybdenum given by Okrent *et al.* in [17]; we do not understand the discrepancy between these group-constants and the energy-dependence of the Mo-capture cross-section assumed by the same authors and reproduced in Fig. 3 of this paper.

As for U^{238}, the energy-dependence of the capture cross-section has been well established between about 3 and 250 keV by Duke University [51], Los Alamos [39], Harwell [39], and ORNL [52] measurements and by theoretical fitting with reaction theory by, among others, BILPUCH *et al.* [51], with some assumptions as to the p-wave strength-functions for neutron elastic-scattering and absorption. Between 0.25 and 1 MeV the most recent $\sigma_\gamma(E)$-measurements by Diven [41] are much higher than the corresponding curve in [39], which combines many earlier consistent measurements, the largest discrepancy of about 50% occurring at 1 MeV. Fig. 4 shows the capture cross-section of

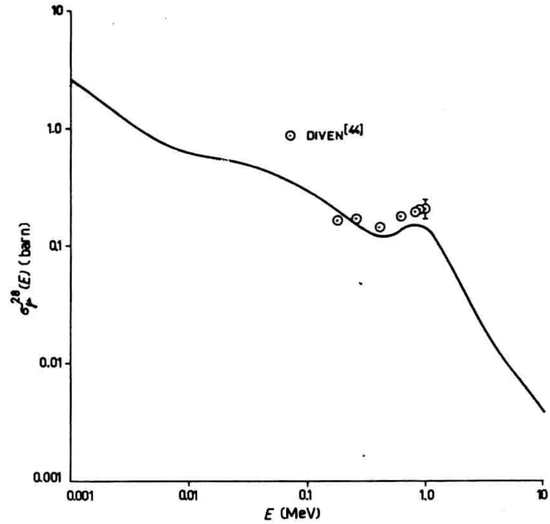

Fig. 4
Radiative capture cross-section of U^{238} between 1 keV and 10 MeV.

U^{238} as a function of the neutron incident energy between 1 keV and 10 MeV, together with the discrepancies mentioned between the former BNL-curve [39] and the new Diven data [41]. Such partially unexplained discrepancies in regard

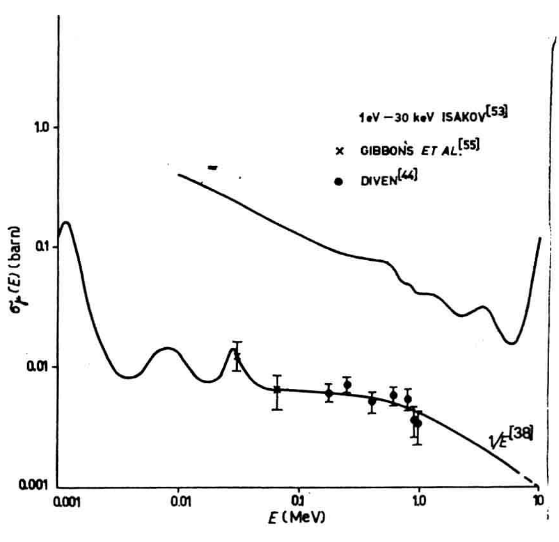

Fig. 5
Radiative capture cross-section of iron between 1 eV and 10 MeV.