

PROGRESS REPORT

PART A.

1. Title of Co-ordinated Research Project

Fission product yield data required for transmutation of minor actinide nuclear waste.

2. Specific Research Objective

(a) Overall objective and goal of the CRP

The objective of the CRP is to develop fission yield systematics and nuclear models as a tool for an evaluation of energy dependent neutron induced fission yields up to 150 MeV. A computer code will (possibly) be developed/adapted that will allow the calculation of fission yields for any given actinide at any desired neutron energy, although with varying accuracy. The final goal of this effort is to provide the users with the desired evaluated fission yields.

(b) Specific research objectives

- To create an experimental data base required for development of models and systematics.
- To study fission modes and theoretical models in the energy range from 10 to 150 MeV: fission after compound nucleus formation or direct reactions, particularly the effect of multiple-chance fission.
- To study the role of the way of formation (projectile) of the fissioning nucleus on the fission product mass and charge distributions.
- To study systematic trends (from measured data) for mass and charge distributions as functions of neutron energy and fissioning nuclide.
- To develop theoretical and phenomenological models and systematics for the description of mass and charge distributions as functions of neutron energy and fissioning nuclide.
- To work on the development of computer programs for calculating fission yields for any desired neutron energy and/or target nuclide, and for calculation of appropriate uncertainties, correlations and covariances.

(c) Specific problems encountered in the present project and proposed approaches to solve them

Unlike in the energy range of thermal and fast reactors, there are by far not sufficient experimental data available for producing evaluated fission yields in the neutron energy range of 10-150 MeV (intermediate energy range). A new approach is required where fission yields are derived from theoretical descriptions and empirical systematics. However, fission of actinides is not yet fully understood for incoming energies above a few MeV, and no satisfactory theoretical description exists. The description of the fission process is complicated by multiple-chance fission: fission is preceded by prompt emission of one or more particles, leaving different nuclides that undergo fission with different product yield distributions. Hence, due to the emission of nucleons prior to fission ('emissive fission'), there is, at each incident energy and for a given target nuclide, a variety of actually fissioning nuclides, all of which contribute to the experimental fission observables.

It is the task of this CRP to thoroughly study existing models for their applicability to fission yield evaluation and adequacy of their prediction capabilities, and, if necessary, to modify these models or develop new ones to obtain an adequate description of the fission process including all

fissioning nuclides and the corresponding yield distributions. Fission yield systematics are required to define ranges of model parameter values for describing the dependence on fissioning respectively target nuclide or neutron energy. To get a complete picture of fission yield distributions for the development of models and systematics, all actinides had to be included in the studies.

In the first RCM the following approach to proceed with the studies was adopted:

- To study the contributions of different fission mechanisms (compound nucleus and direct, including multiple-chance fission) at intermediate energies and define the nuclei formed that actually undergo fission.
- To study individual mass and charge distributions from these fissioning nuclei and develop models to compose the overall mass and charge distributions.
- Since experimental data for neutron induced fission are very scarce in this energy region, it was decided to investigate also other fission reactions (like photo-fission and charged particle induced fission) that lead to the same ‘composite nuclei’ (= target + projectile) as formed in neutron induced fission, and where many more experimental data exist. The question has to be answered whether and in-how-far the results from these other fission reactions can be used for neutron induced fission studies.

With this approach it was decided to compile bibliographic information and experimental yield data also from photo-fission and charged particle induced fission. Parallel to the above mentioned investigations also studies of pertinent theories and models and their modifications to serve the purpose of this CRP are to be pursued.

3. Continued Relevance to Agency Project and to Target Countries

Waste management and specifically the transmutation of nuclear waste plays an important role in the Agency’s programme. Nuclear data including fission product yields of minor actinides are required for feasibility studies of transmutation concepts. A task force of the OECD Nuclear Energy Agency (NEA) is developing a data base of nuclear data for transmutation studies. The first stage includes neutron reaction data up to 150 MeV. The current CRP is co-ordinated with the NEA effort, and a final evaluation is envisaged to be part of that data base.

4. Outputs

Expected	Present Status
1. Bibliographic data base of experimental yield data from neutron, photon, and light charged particle induced fission.	A data base containing references on pertinent experimental data has been established and is updated continuously. It is currently available to CRP participants and will be published in the final report. It may be made available to the scientific community after the completion of the CRP.
2. Compilation of experimental data from neutron, photon, and light charged particle induced fission and initiation of a data base.	Experimental yield data from these fission reactions have been collected and compiled, some into the well established EXFOR format. A data base has been created which is being updated continuously. It will in some form be made available to the scientific community.
3. Measurements performed to support the investigations of the CRP	Systematic measurements, proposed at the beginning of the CRP, have been completed. The results have been used for the development of systematics and to improve models studied during the CRP. A few further measurements have been proposed at the 2 nd RCM. They are still in progress.
4. Sets of reference fission yields for use as standards in yield measurements and as monitors in applications	Higher accuracy for reference fission product yields can be obtained when evaluated individually rather than in a global evaluation of all yields with imposed constraints. Reference fission yield evaluations have been

	completed for thermal, fast and 14-15 MeV neutron fission of ^{235}U and for fast and 14-15 MeV neutron fission of ^{238}U .
5. Treatment of correlations in fission yield evaluation and construction of a covariance matrix	A method has been developed for the treatment of correlated data in fission yield evaluations and for the construction of a covariance matrix for various measurement methods. A computer program has been developed for use in fission yield evaluations.
6. Theoretical models {for description of fission mechanisms (6.1) and for calculation of yield distributions (6.2)}.	Theoretical studies are undertaken to obtain adequate descriptions of the fission process and enable investigations of <u>individual</u> contributions of fission mechanisms and fissioning nuclides to the total observable fission yield distributions for given target respectively composite nuclides (which cannot be obtained from analyses of experimental data only). The best suitable models and their improvements are obtained via comparison with experimental results and testing of model modifications, but also via fitting models to experimental results by adjusting parameters.
6.1. Adequate theoretical model for the description of fission mechanisms and fission cross sections.	Existing models for descriptions of fission mechanisms and fission cross sections have been studied for neutron, photon and charged particle induced fission reactions. The statistical theory approach was adopted to be most adequate for the description of neutron induced fission cross sections. Dependencies of compound nucleus formation as well as contributions of first chance and the higher chances of emissive fission on incident energy have been obtained for the whole energy range. This way, the individual contributions of actually fissioning nuclides have been established.
6.2. Theoretical models for calculating fission yield distributions: Existing models give only qualitative correct descriptions of fission yield distributions. They have to be modified or new approaches developed to achieve predictions with sufficient accuracy for applied transmutation calculations.	<ul style="list-style-type: none"> • Existing models for the calculation of fission yield distributions have been studied and the so-called Brosa model selected as the most suitable one. Several modifications of the original model as well as of computer codes for calculating fission yields have been examined and parameters adjusted by fitting to experimental data. Considerable improvements have been achieved but the predictions are still not accurate enough for applied purposes. Further modifications to achieve improvements have been suggested and will be implemented and tested. • In another study a new approach has been developed that employs phenomenological models for the description of yield distributions, and which was successfully applied for the systematic analysis of experimental data. It could be shown that the process of fission fragment formation (and hence the observable yield distributions) depends not only on the excitation energy but also on the total angular momentum of the fissioning nucleus. Quantitative descriptions of the dependencies and parameter sets could be obtained for several target-projectile combinations up to 80 MeV excitation energy. To get a complete description and possibly a unified parameter set for the whole range of reactions covered in this CRP, the studies have to be extended to further fission reactions and excitation energies up to 150 MeV. • Further studies are devoted to models and computer codes specifically designed for higher energies where shell effects disappear and descriptions are simpler. Some of these studies are still continuing.
7. Modelling of fission yields in evaluations: It was clear before the start of the CRP that evaluated fission yields for intermediate energies up to 150 MeV cannot be obtained and presented in the traditional way used for low energy fission. A new method has to be developed, based on model and sets of equations by which desired yields are calculated.	This objective implies that models have to be developed which allow the derivation of evaluated independent and cumulative yields from neutron induced fission of given target nuclides at given neutron energies. This means that the models have to be derived from systematics of <u>observable</u> yield distributions for 'composite' nuclides. Global systematics employ generally empirical models, derived from experimental results, that are based on assumptions regarding shapes of distributions and functions describing dependencies on 'composite' nuclides and excitation energy. Theoretical studies help to check the validity of assumptions and functions, and to improve models and parameters.
7.1. Models for mass distribution	Models for mass distribution are based on the description of fission modes in the theoretical Brosa model (see 6.2). Systematic studies of the dependencies on target nuclide, projectile and excitation energy have been performed. Existing models for low energy fission have been modified, new parameterizations introduced and sets of equations developed to describe the dependencies. However, presently the model parameters and systematics are still preliminary, as calculated mass distributions are not yet satisfactory for

	all reactions studied and could not be checked where no measurements are available. Further analysis of new experimental data and comparison with theoretical model investigations is necessary to improve empirical models.
7.2. Models for charge distribution	Here also models developed for low energy fission were used with modified parameters, as there is neither a theoretical reason nor an indication from experimental results for changing the distribution functions for intermediate energies. Systematics have been developed including analyses of non-neutron induced fission yield measurements, but even there data are scarce and in some cases doubtful. Therefore an improvement of the description achieved so far can only result from further measurements and is not very likely within this CRP.
7.3. Evaluation method and actual evaluation: The goal was to develop tools for an evaluation and propose a method, as the time given in this CRP is not sufficient to produce a complete evaluation and associated computer programs.	The achievement of this goal of the CRP has to await the completion of the other investigations, particularly the models for mass and charge distribution (see 7.1 and 7.2).

5. Activities

Expected	Present Status
1. Collection of references for fission yield measurements	References have been collected and a bibliographic data base has been established (see 4. Output, item 1.)
2. Compilation of experimental data	As described in the previous section (4. Output, item 2.)
3. Measurements	<u>Petten, The Netherlands</u> : product yields from 190 MeV proton induced fission of ^{nat}W , ^{197}Au , ^{nat}Pb , ^{208}Pb and ^{232}Th : measurements are completed, nothing planned for the coming year. <u>INP, Almaty, Kazakhstan</u> : Mass and energy distributions from proton induced fission of: $^{234,7,8}Np$, $^{239,40,1,2}Am$ and ^{245}Bk at 10 MeV, of ^{236}Np at 10-18 MeV and of ^{233}Pa at 7.4-30 MeV: measurements have been completed; new measurements of 60 MeV 3He ion induced fission yields are planned. <u>IPPE, Obninsk, Russia</u> : yields from 1,5,18,20 MeV neutron induced fission of ^{237}Np have been completed. Measurements of neutron multiplicities and fragment mass distributions for light plutonium isotopes up to 50 MeV excitation energy are in progress, studies of yields from fission induced by 100,200,300 MeV protons and 20-150 MeV neutrons were initiated.
4. Evaluation of reference fission yields	As described in the previous section (4. Output, item 4.)
5. Use of correlations in fission yield evaluations	As described in the previous section (4. Output, item 5.)
6. Investigations whether yields from fission induced by projectiles other than neutrons can be used for descriptions of yield distributions from neutron induced fission.	<ul style="list-style-type: none"> • Generally the same models are used with different parameters for all projectiles. Therefore the validity of models and assumptions made can be tested for projectiles where ample experimental data exist and general conclusions are also valid for neutron induced fission, but not necessarily numerical results (like quantitative contributions, thresholds, critical values). • Improvements of model parameters by fitting to experimental results for non-neutron induced fission were also successfully used for neutron induced fission. • General trends observed in systematic studies of yield distributions for non-neutron fission are also valid for neutron fission, and even numerical values for model parameters and functional dependencies can be used.
7. Analysis of experimental data	Purpose: to check the validity of models, improve model parameters or obtain parameters and dependencies for derivation of systematics.
7.1. Fitting of model to measured yield distributions for adjustments of model parameters and identification of deficiencies and possible improvements of the models	All measurement results described under item 3. above were analysed by fitting of models to the experimental data. Similar studies were done for measurements performed by other scientists. This way parameters values for the generally used Gaussian (mass and charge) distributions were obtained but could still be improved further by continuing these studies. Modifications of the descriptions of yield distributions, e.g. in the number of Gaussians

	fitted or alternative distribution functions were tested but no decisive conclusion has been reached. Further studies have been proposed.
7.2. Comparison with theoretical model predictions to identify possible and necessary improvements of the models	The Brosa model has been improved in several studies: new features have been added like the temperature dependence; the modelling of processes in computer codes has been modified (e.g. for the description of fission barriers, of excited states above the fission barrier, of the splitting into fragments and division of mass and charge, etc.). After the testing of these modifications, further improvements have been suggested which still need to be implemented and tested.
7.3. Study of individual fission yield dependencies on target nucleus, excitation energy, etc.	Systematic studies have been performed of the changes of yield distributions with the variation of one parameter only such as: <ul style="list-style-type: none"> • the excitation energy for given targets; • the projectile and target for the same 'composite' nuclide; • the target nuclide for given excitation energies and projectiles, etc. The obtained descriptions of dependencies can be used for the development of global empirical fission yield systematics.
8. Theoretical description of the fission mechanism	Fission cross sections for neutron-, photon and light charged particle induced reactions have been analysed and compared with different theoretical models to obtain the best descriptions. The selected models have been used to calculate individual contributions of: compound nucleus fission, first chance fission, multiple-chance fission (thus establishing the resulting fissioning nuclides) This research has been completed.
9. Theoretical calculations of fission yield distributions	Such calculations have been performed for improving of the models as described under 7.2 above, but also for theoretical studies of the dependencies of fission yields on excitation energy and angular momentum of the fissioning nucleus (see 4. Output, item 6.2). These studies have not been completed.
10. Empirical models and systematics for mass distributions	Models and systematics by Wahl and by Moryama-Ohnishi have been investigated. Comparisons with measured data revealed varying agreement, and improvements have been envisaged. Wahl's model has been used as the basis for the development of global systematics for mass yields.
11. Empirical models and systematics for charge distributions	Only Wahl's Z_p model has been considered for use in evaluations. Hence it was also used as the basis for the development of global systematics for charge distributions (independent yields).

6. Overall Assessment of Progress towards Achieving Objectives

In general the progress in the research work assigned in order to meet the objectives is within schedule, considering that the CRP was originally planned and approved for a duration of 4 years. There may possibly be some delays in theoretical investigations, as problems with the descriptions of certain reactions have been indicated where solutions still have to be found. Also, the completion of investigations to cover the whole range of target nuclides and excitation energies appears to be more time consuming than envisaged. But in general has the completion of studies, where further investigations are indicated in sections 4. and 5. above, been envisaged for the next planned RCM. Important tasks that have still to be done is the co-ordination of individual efforts and results to obtain improved global systematics and adequate descriptions of fission yield distributions for use in evaluations, and the development of a concept for an appropriate evaluation method.

7. Adjustment to Proposed Workplan until next RCM

Up to now no adjustments are necessary for individual projects since the workplan was already scheduled for 4 years. However, within the frame of this CRP, one of its (possible) goals, the development of a computer program, will probably not reach a state suitable for producing evaluated fission yields.

8. Expenditure to date (\$)

RCM1	1997	16,978
RCM2	1999	18,400
Contracts	1997/1998	17,000
	1998/1999	12,000
	1999/2000	18,000

Total		82,378

9. Proposed Future Expenditure (\$)

RCM3	2001	33,000
Contracts	2000/2001	18,000

Total		51,000

PART B.

Summary of Second Research Co-ordination Meeting of Co-ordinated Research Project on Fission product yield data required for transmutation of minor actinide nuclear waste (Project code: F4.10.16, budget code 1999/2000: G.1.03, task 7)

Dates: 11-15 October 1999

Location: VIC, Vienna, Austria

Scientific Secretary: Meinhart Lammer

List of Participants:

Name	Institute/Country	Contract/ Agreement No.	Title of Project
J.O. Denschlag	Univ. Mainz/Germany	A 9839	Study of exotic fission reactions for the development of fission product yield systematics
A.A. Goverdovski	IPPE, Obninsk/Russia	A 10894	Evaluation of chain yields data for high-excitation of minor actinides
J.-i. Katakura	JAERI, Tokai/Japan	A 10303	Development of systematics for the dependence on energy and fissioning nuclide
Y. Kibkalo	INR, Kiev/Ukraine	C 9843	Phenomenological model for fragment mass and charge distribution in actinide nuclei fission
A.J. Koning	ECN Petten/Netherlands	A 9840	High energy fission yields: experiments and systematics
Liu Tingjin	CIAE Beijing/China	C 9838	Evaluation of fission yield data for reference and minor actinide nuclides
V. Maslov	IRPCP, Minsk/Belarus	C 9837	Emissive fission influence on energy dependence of fission yield
R. Mills	BNF, Seascale/England	A 9842	Advanced fission product yield evaluation
A.C. Wahl	Washington Univ./U.S.A.	C 9844	Nuclear charge and mass distribution from fission
S. Zhdanov	INP, Almaty/Kazakhstan	C 10893	Development of multi-modal approach to the description of fission product yield data for actinides
T. Ethvignot	CEN Buyer-le-chatel/ France	observer	High-energy neutron induced fission: fragment yields and new isomers

F. Storrer	CEN Cadarache/France	observer	Analysis of fission yield data for study of nuclear systems dedicated to the transmutation of minor actinides nuclear waste using the thorium cycle
------------	----------------------	----------	---

Summary of Work Done/Results Achieved:

Participant	Report received Yes/No	Satisfactory Yes/No	Continued participation Yes/No
J.O. Denschlag	Yes	Yes	Yes
A.A. Goverdovski	Yes	Yes	Yes
J.-i. Katakura	Yes	Yes	Yes
Y. Kibkalo	Yes	Yes	Yes
A.J. Koning	Yes	Yes	Yes
Liu Tingjin	Yes	Yes	Yes
V. Maslov	Yes	Yes	Yes
R. Mills	Yes	Yes	Yes
A.C. Wahl	Yes	Yes	Yes
S. Zhdanov	Yes	Yes	Yes

Main Scientific/Technical Conclusions from Meeting:

For the first time a fairly complete collection and data base of neutron-, photon- and light charged particle induced fission yields for intermediate (excitation) energies has been established. Very good progress has been made in theoretical studies towards the understanding of the fission process and the adequate description of fission mechanisms, cross sections and the distribution of mass and charge in fission. Systematics of the dependence of fission yields on excitation energy and fissioning nuclide has been developed for the first time. After completion of the studies as proposed in Part A, the development of a new evaluation method appropriate for intermediate energy fission yields should be possible.

Recommendations:

(a) To participants:

To continue with their projects and tasks as originally planned and with modifications decided at the RCM, as outlined in Part A. To start with the preparation of drafts of their contributions to the final publication for presentation at the next RCM. To carefully consider whether their projects can be completed until the next RCM or whether a continuation of the work beyond that time and a further extension would be necessary, and inform the Project Officer accordingly in due time.

(a) To Agency:

To approve a continuation of the CRP for a fourth year.