

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 Objectives

The need for evaluated fission yields

- Review the transmutation scenarios and associated data needs on safety related parameters and determine the requirements for the development of a fission yield data evaluation.

Collection of data

- Create a computer file of literature references containing experimental fission yield data.
- Create an experimental data-base required for the development of models and systematics.

Yield data measurements and evaluation

- Perform experimental studies and analysis of neutron and proton induced fission yields at intermediate energies.
- Further develop the methodology and tools for fission yield data evaluation, with emphasis on covariance matrix construction, correlation transmission and production, and evaluation systems.
- Define the requirements of evaluators for the systematics and models to be used in the evaluation process.
- Study systematic trends (from measured data) for mass and charge distribution in fission, and develop a system of equations describing these systematics for application in the evaluation of neutron induced fission yields from 10 to 150 MeV for all actinides.

Theoretical studies and model development

- Study fission modes and theoretical models for the fission cross section in the energy range 1-200 MeV, after compound nucleus formation or direct reactions, particularly the effect of multiple-chance fission.
- Study the role in the formation (projectile) of the fissioning nucleus on the fission product mass and charge distributions.
- Develop phenomenological models for the description of charge and mass distributions as functions of incident particle energy and nucleonic composition of fissioning nuclei. Utilize these models for the analysis of experimental yield distributions to determine the model parameters and to obtain the systematics of their dependencies.
- Determine from theoretical and phenomenological studies the energy dependence of pre- and post-fission particles' multiplicities as basic characteristics influencing the prediction power of the evaluation of yields from intermediate energy neutron-induced fission.
- Predict fission product yield data resulting from nucleon-induced reactions at intermediate energies, based on dedicated nuclear reaction models.

Benchmark exercise to test the quality of predictions

The prediction capabilities of the models and systematics developed during this CRP need to be tested and fully defined so that improvements and recommendations regarding their use in the

development of fission yield evaluation methods and evaluated data files can be formulated. This objective can be fulfilled through a benchmark exercise designed to produce the desired results within one year. This exercise will generate uncertainty estimates for the fission yields provided by the CRP for applications.

3. Continued Relevance to Agency Projects and to Target Countries

Waste management and studies of the transmutation of nuclear waste play an important role in the Agency's programme. Fission product yields of the minor actinides and their uncertainties are required to undertake valid and credible studies of transmutation concepts. A computer program has been developed during this CRP that can calculate fission yields for application in transmutation studies. Fission yield uncertainties will be produced during the proposed benchmark exercise.

4. Outputs

Expected	Present Status
1. <u>Bibliographic data base</u> 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 completion of the CRP.
2. <u>Compilation</u> 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 be made available to the scientific community in some suitable form.
3. Sets of <u>reference fission yields</u> for use as standards in yield measurements and as monitors in applications	Higher accuracy reference fission product yields are available for thermal, fast and 14-15 MeV neutron fission of ^{235}U and for fast and 14-15 MeV neutron fission of ^{238}U , including fission products used as standards in fission yield measurements and requested by users as monitors in applied measurements. Reference yields for ^{252}Cf spontaneous fission are in preparation.
4. Treatment of <u>correlations</u> 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. An extension of the project is in progress to include fission yields estimated from systematics.
5. <u>Systematics and phenomenological models</u> for the description of fission yield distributions derived from <u>experimental</u> data.	<ul style="list-style-type: none"> • The systematic behaviour of the energy dependence of experimental fission yields has been studied for several fissioning nuclides. • Global systematics of the dependences of fission product mass and charge distributions on the fissioning nuclides and incident particles have been derived from experimental data for neutron, photon and charged particle induced fission. • Phenomenological models have been utilized to analyse experimental fission yield distributions in order to derive their dependences on model parameters. The goal, to obtain a description of the systematic behaviour of yield distributions by phenomenological models that will allow the prediction of unmeasured fission yields, has not yet been reached.
6. Study of the <u>emissive fission</u> influence on the energy dependence of fission yields.	<ul style="list-style-type: none"> • Fission cross sections have been studied for neutron and proton (for comparison) induced fission reactions using the statistical model as the most adequate one for this purpose. The method of partitioning the observed neutron-induced fission cross-sections into emissive and non-emissive fission has been validated for the whole energy range and Th, U, Np and Pu target nuclides. An extension of the study to include minor actinides is required. • Differences of fission mechanisms and contributions of actually fissioning nuclides in neutron, photon and charged particle induced fission reactions have been studied in view of their application for deriving systematics for neutron induced fission yield distributions.

7. <u>Theoretical prediction</u> of fission product yields at intermediate neutron energies (10-150 MeV)	The treatment of intermediate energy fission in theoretical models is complicated by two factors: multi-chance (emissive) fission and the changing fission characteristics with excitation energy. A new theoretical approach has been developed for the prediction of fission product yields in this energy range, which tackles both problems simultaneously. This approach is based on an extended version of the Brosa model coupled to either the nuclear reaction code ALICE91 or TALYS.
8. Evaluated fission yields and their uncertainties up to 150 MeV incident neutron energy: goal is to develop <u>tools for an evaluation</u> and propose an evaluation method.	<ul style="list-style-type: none"> • Several models for the description of fission yield distributions have been developed, but need further improvements and modification of codes to enable reliable predictions of fission yields. • Empirical equations representing systematics of fission-product yields have been derived that allow estimation of yields from fission of any actinide with atomic number $Z=90-98$, mass number $A=230-252$ and excitation energy 0-200 MeV. A set of equations has been developed to estimate the uncertainties of these yields. • A computer program has been written to perform the calculations of fission yields and their uncertainties. It will be made available to the scientific community. • At present there is no information on the reliability of the predicted fission yields and the validity of the uncertainty estimates. Therefore a benchmark exercise has been proposed and designed to derive this information.

5. Activities

Expected	Present Status
1. Collection of references for fission yield measurements	The literature search and collection of pertinent references has been continued and the bibliographic data-base updated (see 4. Output, item 1.)
2. Compilation of experimental data	As described in the previous section (4. Output, item 2.)
3. Measurements performed to support the investigations of the CRP	Since the previous RCM and progress report, further measurements have been completed. The results have been used for the development of systematics and to improve models studied during the CRP. A few more measurements are still in progress and results are expected in 2002.
4. Evaluation of reference fission yields	Reference fission yields have been obtained with higher accuracy than complete yield sets in evaluated files, making full use of correlations and a careful analysis, correction of experimental data and assessment of uncertainties. The resulting yield data were compared to evaluated data files and discrepancies investigated.
5. Use of correlations in fission yield evaluations	A method and computer code have been developed for the simultaneous evaluation of measured relative and absolute fission yields and ratios, using correlations for constructing the covariance matrix. The method and code have been tested, and the effect of correlations introduced in the evaluation process on the evaluated yield and their uncertainties have been studied.
6. Investigations whether models and experimental data for yields from fission induced by projectiles other than neutrons can be used for descriptions of yield distributions from neutron-induced fission.	These investigations have been completed with the following results: General conclusions drawn from model analysis of yields from fission with different projectiles are also valid for neutron-induced fission. General trends observed in systematic studies of yield distributions for non-neutron fission are also valid for neutron fission. However, not all functional dependencies, values of model parameters and numerical results can be used.
7. Study of the energy dependence of neutron induced fission yields.	Available higher-accuracy experimental fission yields from mono-energetic neutrons have been analysed for different regions of the mass distributions and systematic trends derived as functional dependences.
8. Global systematics of fission yields as functions of fissioning nuclides and excitation energy.	<p>All available data of experimental mass and charge distributions from neutron and proton induced fission have been investigated.</p> <ul style="list-style-type: none"> • Systematics of mass distributions: parameters for the multi-Gaussian model were determined by the method of least squares from experimental chain-yield data for 63 fission reactions. The derived functions allow calculation of mass distributions for any fissioning nuclide in the energy range investigated. • Parameters for Wahl's charge distribution model were determined by the method of least squares from fractional independent and cumulative yield

	<p>values derived from experimental data. Parameter values for 25 fission reactions with excitation energies of 0-170 MeV were used.</p> <ul style="list-style-type: none"> • Empirical functions for calculating uncertainties of estimated yields have been derived from numerous systematic studies of mass and charge yield distributions. • A computer program has been written that allows the calculation of any desired fission yield at intermediate neutron energies together with an uncertainty estimate. <p>The investigation has been completed. However, it would be desirable to test the validity of calculated fission yields and their uncertainties.</p>
9. Moriyama-Ohnishi systematics for mass distributions	Moriyama-Ohnishi systematics have been used to analyse and reproduce measured fission product mass distributions. The model has been improved but is still unsuitable for certain fission reactions. Further modifications are planned to remove the last discrepancies, and to enable the prediction of unmeasured yields for participating in the benchmark calculations.
10. Development of a multi-modal approach to the description of fission product yield data for actinides	Fragment mass and energy distributions for the proton induced fission of several actinide nuclei have been measured and analysed in terms of the multi-modal approach: detailed studies of the independent mode parameters have revealed regularities in their behaviour that can be used for the development of predictive systematics. The model and computer code need further modifications to enable the prediction of unmeasured fission yields for participation in the planned benchmark calculations.
11. Theoretical study of the fission mechanisms and emissive fission contributions to the total fission cross-section, in order to obtain the contributions of actually fissioning nuclides to the total fission yield distributions.	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 show that the emissive contribution to the observed fission cross section is dependent on target fissility and fission probability for high excitation energies. The conclusions have been validated for neutron-induced fission of Th, U, Np and Pu target nuclides and proton-induced fission of ^{238}U . An extension to minor actinides is planned.
12. Analysis and evaluation of chain yield data for high-excitation fission of actinides.	The results of study 11 above are to be used for a theoretical analysis of fission yields to determine the main parameters of Brosa-channels and their energy dependence. Results of that study are still pending.
13. A phenomenological model for the study of the dependence of fission product mass distributions on incident particles and their energies, and on theoretical model parameters, and the prediction of fission yields.	A new approach to describe fragment mass distributions of actinide nuclei fission accounting for the influence of total nuclear angular momentum has been used to analyse experimental fragment mass distributions from fission with neutrons, photons and α -particles. The dependence of fragment mass distribution parameters on excitation energy and transferred angular momentum of the fissioning nucleus is derived. Planned further work: extension of studies to higher energies, and modification of the model code to improve the fitting procedure and enable the prediction of fission yields.
14. Theoretical prediction of fission product yields at intermediate neutron energies (10-150 MeV)	Brosa model has been adapted for application at intermediate energies, where the contributions of different fission modes change, thus leading to an enhancement of symmetric fission. First, the temperature dependence has been added to the calculation of the deformation energy. In this manner the vanishing shell effects are taken into account in the calculation of fission modes contributing at different energies. Second, the relative contribution of each fission mode is now calculated using the Hill-Wheeler expression for the transmission through a parabolic fission barrier. Third, the original random-neck rupture model by Brosa is employed in the determination of the fission fragment mass yields, but with pre-scission shape parameters depending on temperature. The coupling of the random-neck rupture model to a nuclear reaction code like ALICE91 or TALYS enables the prediction of fission product yields, incorporating the competition between fission and all other reaction channels.

6. Overall Assessment of Progress towards Achieving Objectives

The objectives to develop models and systematics for fission yields at intermediate incident neutron energies, where they did not exist before, are very ambitious. At the beginning of the CRP, it was clear that completion of the proposed research work within the scheduled time could be uncertain. Therefore, the goal of the CRP was carefully formulated “to develop fission yield systematics and nuclear models as **tools** for evaluations

. This initial goal has been achieved: global systematics and nuclear models are available along with a computer program to calculate any desired fission yield. However, participants see the need for further improvements of the developed models and associated codes

(1) to achieve agreement with experimental data for all fission reactions considered in this CRP;

(2) to enable the prediction of fission yields.

Furthermore, it is considered of high importance, particularly for application fields, that the reliability and uncertainty of the fission yields calculated with the developed models and systematics be assessed. Thus, benchmark calculations are proposed, the results of which should be available within one year.

With respect to the development of a complete fission yield evaluation from the products of this CRP, such a task would be better achieved by a smaller group or an individual.

7. Adjustment to Proposed Workplan until next RCM

Assuming a one-year extension of the CRP is approved, the following work is proposed:

Where necessary, nuclear models should be improved and model codes adapted to enable the prediction of fission yields.

A benchmark exercise for the prediction of fission yields should be performed, consisting of 2 parts:

Part A: Perform calculations for defined sets of fission reactions where many experimental data are available, and compare the results with the evaluated experimental data.

Part B: Perform calculations for defined sets of fission reactions where no experimental data are available, and compare results.

From both parts: identify discrepancies and deficiencies of models, assess the reliability and uncertainties of predictions.

The proposed **new workplan** would have the following timetable:

Until end of April 2002 - complete modifications for most nuclear models and codes. Furthermore, for exercise A, all available experimental data should be collected and evaluated for comparison with calculations.

Until end of June 2002 - part A of the benchmark calculations should be completed; evaluation of results should commence.

Until end of September 2002 - part B of the benchmark calculations should be completed and results distributed to all participants; evaluation of results should commence.

November 2002: a fourth and final RCM should take place at the end of the additional year.

8. Expenditure to date (\$)

RCM1	1997	16,978
RCM2	1999	18,400
RCM3	2001	18,080
Contracts	1997/1998	17,000
	1998/1999	13,000
	1999/2000	18,000
	2000/2001	18,000

Total		119,458

9. Proposed Future Expenditure (\$)

RCM4	2002	20,000
Contracts	2002	18,000

Total		38,000

PART B.

Summary of Third 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 2000/2001: G.1.03, task 5)

Dates: 29 October – 2 November 2001

Location: VIC, Vienna, Austria

Scientific Secretary: Meinhart Lammer

List of Participants:

Name	Institute/Country	Contract/ Agreement No.	Title of Project
I. Tsekhanovich representing 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
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*) Could not participate for health reasons, but his paper was presented at the RCM.

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	No	No	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:

Fairly complete collections of bibliographic and experimental data for neutron-, photon- and light charged particle-induced fission yields at intermediate (excitation) energies have been established for the first time, along with data bases. These data should be maintained after the CRP and be made available to the scientific community. Some of the CRP results like the sets of evaluated reference fission yields and the introduction of correlations in fission yield evaluation, although obtained as tools for intermediate energy neutron-induced fission yields, could also be used to improve current evaluations of thermal and fast neutron-induced fission yields.

The empirical systematics of fission yields are complete since they are derived from all available experimental data for the energy range and fission reactions considered, and a computer program has been developed for the calculation of any desired fission yield. Nuclear models have been developed to successfully reproduce most experimental fission yield distributions, but still fail in some specific cases. The reasons have to be investigated and the model corrected. At present, only the model and code by Duijvestijn and Koning is capable of predicting fission yields. All models and associated codes should be modified to enable the prediction of fission yields and participation in the benchmark exercise.

A benchmark exercise (for details see “workplan”, above) should be conducted involving all models and computer codes developed during this CRP to test their predictive capabilities. The analysis of the results would provide information on the reliability and uncertainties of the calculated yields and a check of Wahl’s estimated model uncertainties.

The implementation of the results of the CRP in the development of a new complete evaluation method and an actual evaluation for intermediate energy fission yields could be realised later by a few or one individual scientist, possibly guided by IAEA consultants meetings.

Recommendations:

(a) *To participants:*

implement the modifications of models and computer codes decided at the last RCM in order to improve the agreement with experimental data and allow the prediction of fission yields; participate in the benchmark exercise which is designed to test nuclear models and systematics for the prediction of fission yields; prepare drafts of their contributions to the final publication for presentation at the next RCM.

(b) *To Agency:*

approve a continuation of the CRP for one year.