Activities of the ESARDA Working Group for techniques and standards for Non-Destructive Analysis

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Abstract

ESARDA (European Safeguards Research and Development Association) is an association of European organizations involved in the R&D of Nuclear Safeguards mainly appointed to assist the Safeguard community with advanced progress in safeguards, enhancing the efficiency of systems and measures, as well as investigating how new techniques can be developed and implemented. Among the various activities of the association, Working Groups are established to promote and undertake collaborative R&D and information exchange activities in various fields.

This paper will describe the objectives, composition and activities of the Working Group for techniques and standards for Non-Destructive Analysis (ESARDA NDA-WG) and will give an overview of the major (ongoing or recently accomplished) projects. These projects can be categorised in four main technical fields: general function of NDA instrumentation, NDA techniques for Safeguards (γ spectrometry and neutron counting), NDA techniques for waste sentencing and modelling of NDA instruments.

1. Introduction

ESARDA is the European Safeguards Research and Development Association, gathering Safeguards Authorities, Operators of Nuclear Facilities, Research Centres and Universities to exchange information, co-ordinate and execute joint R&D programmes in the field of Nuclear Safeguards. Currently the members of the association are: AREVA (France), ATI (Austria), BNFL (UK), CEA (France), DTI (UK), EDF (France), ENEA (Italy), European Commission (representing the European Atomic Energy Community EURATOM), FZJ (Germany), HAEA (Hungary), IRSN (France), IKI (Hungary), SCK-CEN (Belgium), SKI (Sweden), STUK (Finland), UKAEA (UK), VATESI (Lithuania) and WKK (Germany). Representatives from other organisations that are not member parties (amongst them the IAEA, ABACC, and INMM) regularly take part in the activities of the association. Among the various activities of the association, Working Groups are established to promote and undertake collaborative R&D and information exchange activities in various technical fields.

The Working Group for techniques and standards for Non-Destructive Analysis (ESARDA NDA-WG) was established with the mission to provide the Safeguards Community with expert advice on Non Destructive Analysis methods, procedures, reference materials and on the performance of NDA methods, as stated in its Terms of Reference. It acts as a forum for the exchange of information on Non Destructive

Analysis methods, gathering national, regional and international control authorities, together with plant operators and R&D laboratories. This paper will describe the function, the composition and the activities of the NDA Working Group, by giving an overview of the major ongoing or recently completed projects.

2. Terms of reference

The NDA WG gives its support to the Safeguard community on Non Destructive Analysis methods by carrying out a series of tasks :

1. Maintain a list of NDA methods and instruments currently used or under development for accountancy and verification purposes.
2. Determine the reliability of NDA methods where possible with inter-comparison exercises of safeguards measurements.
3. Advise EURATOM and IAEA on the implementation of new and improved methods and advise on areas where R and D is needed.
4. Promote and coordinate R and D programs to fulfill safeguards and nuclear material management needs.
5. Promote the systematic and correct use of Reference Materials.
6. Assess and disseminate Performance Values for Uncertainties in NDA methods of nuclear material measurement.
7. Participate in the review, and promote the use of, International Target Values for uncertainties in measurements of nuclear material.
8. Consider sampling errors and sampling problems and their significance for NDA results.
9. Promote the use of internationally agreed definitions and terminology in the reporting of measurement results.
10. Assist in the development of new NDA methods in support of new safeguards requirements.
11. Promote cooperation with other working groups and the inspection authorities.
12. Collaborate with other working groups to develop comprehensive and integrated tools to fulfill new safeguards requirements.

The above tasks are undertaken by the NDA Working Group generally meeting twice per year in Europe: in spring around the ESARDA annual event and in autumn at the working place of one of the members.

3. Participants of the ESARDA NDA-WG

The NDA WG is composed of specialists from nuclear plant operators, NDA equipments suppliers, R&D laboratories and regulatory authorities. Some of the ESARDA members have representatives participating regularly to the WG activities: AREVA (through

CANBERRA), BNFL (through BIL Solutions ltd), CEA, European Commission (JRC and TREN), IRSN, IKI, SCK-CEN, SKI and UKAEA. The group receives as well an active contribution also from observers (participants from non-member organisations): IAEA, ABACC and US national laboratories (Los Alamos and Lawrence Livermore).

4. Projects and achievements of the ESARDA NDA-WG

The WG activities undertaken these last years can be categorised according to four main technical fields:

- General functions of NDA instrumentation
- NDA techniques for safeguards (gamma spectrometry and neutron counting)
- NDA techniques for waste sentencing
- Modelling of NDA instruments (mainly Monte Carlo techniques)

4.1. NDA instrumentation

In the field of general NDA instrumentation, the NDA WG has been active for many years in the assessment and the updating of a comprehensive list of performances values for NDA techniques currently used for the assay of nuclear materials encountered in the fuel cycle for Safeguards purposes, according to the tasks 6 and 7 of the terms of reference. The successive evaluations are the result of specific international round robin exercises (carried out with specifically designed reference materials with the aim of assessing NDA performances), field measurements (field inspections activities) and tailored laboratory measurements, evaluated and discussed in the Esarda NDA WG.

The WG has participated to the redaction of the document on “International Target Values 2000 for Measurement Uncertainties in Safeguarding Nuclear Materials” [1]. This document lists for all the available analytical techniques (including DA and NDA) the desired measurement uncertainties that should be achieved in order to fulfil the Safeguards objectives.

Then the real performances of NDA instruments have been reviewed in the document “Performance Values for NDA techniques applied to safeguards” [2]. This document catalogues all the NDA techniques applied to Safeguards of nuclear materials, provides an extensive description of the principles, of the equipment and of the procedures and finally for each technique/application combination performs an accurate assessment of all the uncertainty components compiling tables of the real performances achievable with the NDA techniques.

Another performance value document for NDA techniques applied to waste sentencing is in progress. The document will contain a categorisation of the typical waste and waste packaging methods used in the nuclear plants, review the NDA techniques available for waste sentencing and finally compile the performance value with main focus on the fissile material determination. The performance values will include the detection limit and accuracy (total measurement uncertainty), to allow intercomparisons to be made, and the most appropriate measurement technique to be selected.

The use of unattended and remotely-operated instrumentation is becoming more and more used in nuclear safeguards in order to reduce the on-site inspection effort. Following a request of the IAEA and in collaboration with the Containment and Surveillance Working Group (ESARDA C/S-WG), a joint document on "Guidelines for developing Unattended Remote Monitoring and Measurement Systems" has been issued [3]. The scope of this document was provide a list of technical specifications and requirements that unattended equipment must (or in some cases simply should) fulfil in order to be acceptable for field deployment.

Finally a collection of NDA instrument characteristics is going to be compiled in a data-base, according to the 1st task of the terms of reference. As a first step all the NDA instruments applied for spent fuel have been catalogued together with their main technical characteristics. The data have been filled within an MSAccess database. Currently the structure of the database must be optimised in function of the interrogation needs and appropriate query tools must be developed. When this will be completed the database will be extended to all types of NDA instruments.

4.2 Gamma spectrometry

In the field of gamma spectrometry several inter-comparisons have been organised in order to assess the capabilities of this technique. The last of these was the Pu-2000 exercise dedicated to the determination of the plutonium isotopic composition. The main purpose was to test the performances of recent X and γ spectrometry methods developed for determining Pu isotopic composition over a wide range of abundances and to investigate possible sources of error. 20 plutonium-bearing reference samples have been prepared by the IRMM in Geel and measured by 8 laboratories using 18 different techniques (detector, acquisition chain and analysis software as MGA, MGA++ and FRAM). The results have been analysed and published [4].

The analysis work has led to a range of uncertainties that can be expected using those methods: from 3% to 20% for ^{238}Pu/Pu mass ratio when the ratio varies from 1.5% to 0.005%, 0.2 to 2% for the ^{239}Pu/Pu mass ratio (90 to 60%), about 4% for ^{240}Pu/Pu, about 3% for ^{241}Pu/Pu and about 3.5% for ^{241}Am/Am. As 10^4 results were to consider, only general conclusions have been drawn. A more elaborated statistical analysis of the leading parameters was then performed grouping data by plutonium type (low, medium and high burnup), revealing some bias for medium and high burnup plutonium and also some differences in the results obtained by different versions of the codes used. At that time, the NDA WG has decided to launch a complete performance evaluation of the plutonium isotopic codes, in cooperation with the developers, using the last versions of the codes, based on a set of ideal spectra firstly (taken in lab with good statistics on well-characterized reference samples), then on a selected number of spectra from the Pu2000 library. The measurement of real high burn-up plutonium, being more and more common in the European facilities, will be investigated by JRC/ITU, and the results will be reported within the NDA WG.

A compilation of uranium and plutonium spectra acquired during the Pu-2000 exercise and a previous one dedicated to uranium enrichment has been collected in the "ESARDA U/Pu Spectra Library" available on the web [5] for anyone who wants to use them to assess the performance of spectra analysis codes. The U and Pu reference spectra were provided by LNHB using two GeHp detectors (planar + coaxial) and a CZT detector. Other U ref spectra from LLNL were added with $^{235}U/U$ mass ratio from 0.018 to 99.1%, measured with a planar detector.

The benchmarking activity has recently found an important milestone in the organisation of the Workshop on "Gamma Evaluation Codes for Plutonium and Uranium Isotope Abundance Measurements by High-Resolution Gamma Spectrometry: Current Status and Future Challenges" held in Karlsruhe in November 2005. The workshop gathered 44 specialists from 12 countries including software developers, detector manufacturers, users from national laboratories and safeguards inspectorates (IAEA and Euratom). The current state-of-the-art of gamma spectrometry has been extensively evaluated and recommendations have been issued on harmonisation and version control, nuclear data standardisation, future requirement for unattended measurement and new materials from future fuel cycles, procedure optimisation. All the presentations and the minutes of the round table will be published in the proceedings of the Workshop [6].

4.3 Neutron counting and Monte Carlo modelling

A large interest is devoted to the application of Monte Carlo techniques to the numerical simulation of NDA instruments in general and neutron counters in particular, widely used in safeguards for the verification of fuel pins and assemblies, the measurement of the fissile content of scrap residues from reprocessing activities and the assay of individual fuel pellets for process control. The use of MC modelling is becoming increasingly widespread as a tool for reducing the reliance upon experiment for calibration of neutron coincidence counting systems.

Three benchmark exercises have been carried out in the last years in order to assess the capabilities of Monte Carlo to reproduce the experimental data:

- The first one dealt with the comparison of interpretational models used for the prediction of the real coincidence rates from a reference PWR fuel assembly measured with an active neutron collar when using the MCNP code [8] The participants used the same nuclear data for MCNP runs, with a fixed, predefined geometry model, but different interpretational models. The results of comparisons with experiment demonstrated that predictions could generally be made to an accuracy of 5 – 10%. However, due to uncertainties in the accuracy of the nuclear data constants used, it was difficult to evaluate the factors which determine the fundamental limitations to the level of absolute agreement that can be expected between measurement and calculations.
- The second one was then launched to analyse the influence of the main basic physical parameters (influence of fission spectrum, thermal treatment, cross section dataset, geometry model approximations) for Monte Carlo codes in common use on a simple case [7]: a point californium source placed at a fixed distance from a slab detector with interposed layers of moderator (polyethylene) and absorber (cadmium). The exercise has demonstrated that Monte Carlo modelling can be used to predict the Totals counting rate for a simple neutron counting geometry to a typical level of agreement of about 5% for typical lightly moderated geometries. The spread of results according to different nuclear data

and modelling styles is also of the order of a few % for lightly moderated geometries, but increases up to 10% for heavily moderated geometries. The experiments and comprehensive uncertainty analysis have provided a useful insight into the fundamental limitations to the level of agreement that can be accepted between measurement and calculation. The physics uncertainties concerning largely the physics and design of the 3He detectors lead to a minimum uncertainty of about 2% in the efficiency for this geometry. Furthermore, if one considers the typical uncertainties in the bulk density of polyethylene, an additional component of between 1 and 2.5% can be expected, increasing the overall uncertainty to up to 3%.

- The most recent one intended to model a passive multiplicity counter. This last benchmark was split in two parts. A full simulation of neutron generation, transport and detection, coupled with the simulation of electronics had the purpose to compare coincidence counting simulation tools, such as MCNPX and MCNP-PTA. A second phase was aimed to compare only the pulse train analysis models, also studying dead-time effects, and all the participants analysed the same set of pulse trains. An AWCC in fast mode was chosen, considering a set of 13 sources: random sources, pure spontaneous (^{252}Cf and Pu metal) sources, Pu oxide samples and mixed random source/PuO_2 samples. The results of this exercise have been described in a final report [9]. The exercise has confirmed that the point model works well for a typical well counter with reasonable samples. A special trick had to be used to make the point model work with the case of (α,n) neutrons having a significantly different detection efficiency from spontaneous fission neutrons. Both MCNPX and MCNP-PTA are reliable tools for the calculation of coincidence counter performance. The exercise also showed a consensus among the main labs concerning the method of calculating singles, doubles and triples from pulse trains. The results for the high counting rate and high (α,n) cases have large statistical uncertainties, preventing very precise conclusions being drawn about dead-time effects and correction methods.

  A follow-up of this benchmark is currently considered. The idea is to repeat the exercise with an experimental pulse train acquired in LIST mode, instead of a simulated one. The goal is to compare the available software for LIST mode data analysis in view of possible future developments of neutron counting towards the abolition of shift register analysers and direct acquisition and processing of pulse trains by a PC.

Under specific request of the IAEA, the NDA-WG is also redacting a “Good Practice Guide in the use of Numerical Simulation in NDA”. The objective is to set up a system of behaviour rules to be followed by anybody who is using computational modelling applied to NDA techniques, comprising both technical and non-technical considerations. Technical considerations will include the nuclear data used, the validity of the physics treatments and interpretational models, benchmarking the code under representative conditions, and the use of specific codes according to recognised procedures. Non-technical factors will include Quality Assurance, training and competency of the modelling practitioner. The guide will cover generic families of codes and application areas: Monte Carlo, gamma-attenuation modelling codes and burnup / depletion codes), but be limited to applications having a direct impact on an NDA system result, through calibration or data interpretation. The correct implementation of the good practises will constitute a sort of quality certification that will allow to help in the acceptance of modelling results in measurement techniques and evaluation procedures.

4.4 Waste sentencing

Apart from the above mentioned document on performance values, the working group has sponsored the preparation of reference standard for measurements of nuclear material in waste drums. 16 waste drums have been produced having different size (100 and 200 litre drums) and different matrix (homogeneous/heterogeneous with plastic, metal or mixtures). These drums are provided with insertion tubes where different reference sources can be located. A set of 37 pins containing plutonium can be arranged in order to load the drums with masses ranging from 15 mg up to 10 g and simulate concentrated and distributed sources. Waste drums and sources are currently stored at SCK in Mol.

The working group is currently organising an inter-comparison exercise of measurement techniques applied to the characterisation of plutonium in waste drums. The main goal of the proficiency test is the comparison of neutron counting systems especially passive neutron coincidence counting (determination of coincident pulse pairs) and multiplicity counting (determination of coincident pulse pairs and pulse triplets). The outcome of these systems is a ^{240}Pu equivalent mass, from which isotope mass can be computed if the isotopics are known. Hence the determination of the isotope vector (using gamma spectrometry) on the waste drums could be considered as well as part of the exercise.

5. Conclusions

The Esarda NDA WG will continue assessing the performances of NDA techniques in use for safeguard purposes as well as of new emerging methods, organising intercomparison exercises as round robin exercises where labs make comparative NDA measurements on a set of samples, or intercomparisons for data analysis codes.

The implementation of the additional protocol and the new Euratom approach could require developments such as newly designed instruments, integrating advanced technologies, more automated and remotely controlled. These aspects should be covered within the WG. A special attention need to be paid to spent fuel verifications. Regarding the detection of undeclared materials, a need has been expressed by the IAEA for developing rapid and sensitive screening techniques for environmental sampling. A joint meeting between both DA and NDA WGs was organised in May 2006 in order to initiate a common work.

The strengthening of policy towards the nuclear threat is requiring new concerns for the NDA WG in the area of fighting the illicit trafficking. The NDA WG is currently monitoring the international scenario and technical developments, as a few members are involved.

Future developments for the generation 4 reactors should also be of interest for the NDA WG, with the need to adapt current methods to the new fuel. Safeguards need to be developed with the design, and one of the members is involved in the generation 4 project.

References

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