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Summary Report of Consultants' Meeting

Assessment of Nuclear Data Needs for Particle Induced Gamma Ray Emission (PIGE)

IAEA Headquarters, Vienna, Austria
1 – 2 March 2010

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March 2010

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Abstract

A summary is given of a Consultants' Meeting assembled to assess the viability of a new IAEA Co-ordinated Research Project (CRP) on *Development of nuclear data for PIGE for analytical applications*. The need for such a CRP was strongly agreed. Both the technical discussions and the expected outcomes of such a project are described, along with detailed recommendations for implementation. The participants discussed the past and present status of the field and the experiments performed in their home institutes. Subsequently, they addressed the nuclear data needs for PIGE analysis and produced recommendations concerning the compilation, assessment, measurements, evaluation and benchmarking of cross-section data for PIGE as well as their inclusion in the existent IBANDL database.

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1. Introduction

Several ion beam analysis (IBA) techniques are important for analytical purposes. Among these, EBS (Elastic Backscattering Spectroscopy) and NRA (Nuclear Reaction Analysis) with charged particles were recently explored by the IAEA in a Coordinated Research Project about IBANDL (Ion Beam Analysis Nuclear Data Library). Particle Induced Gamma-ray Emission (PIGE) is one of the most powerful analytical techniques that exploit the interactions of rapid (~ MeV) charged particles with matter to determine the composition and structure of the surface regions of solids (from ~ 0 to 500 μm) by measurement of characteristic prompt γ rays. This technique has been used since the early 1960's for different applications ranging from analysis of fission reactor materials to biomedicine, environment, cultural heritage and, more recently, fusion reactor materials. The potential for depth profiling of this technique, with better resolution than the other IBA techniques, was soon recognised. The basic physical processes underlying PIGE are now well understood, but the reliability of analytical results is limited by knowledge of the physical data.

Compositions and structures are inferred from measured quantities as γ-ray spectra or excitation curves, via physical models incorporating the sample structure and the basic physical processes and quantities giving rise to the observed spectra or excitation curves. The primary quantities required are the stopping power and the cross sections of the interactions involved. Whilst work remains to be done on accurate stopping powers, the field is largely catered for by the considerable body of work of Ziegler and co-workers, embodied in the recently released SRIM2010 computer code.

The case is quite different for cross sections for nuclear reactions with γ rays in the exit channel. Although there exists a considerable body of published data in the nuclear physics literature, there is no up-to-date, comprehensive compilation especially dedicated to the IBA community. A number of PIGE cross-section data have already been uploaded to IBANDL by members of the IBA community. In doing so, the IBA community has shown that there is an overwhelming need for compilation, assessment and evaluation of PIGE data. However, a preliminary survey of this body of unevaluated experimental data has revealed numerous discrepancies beyond the uncertainty limits reported by the authors, and ion beam analysts are faced with the dilemma of trying to decide which (if any), amongst the divergent cross section data, they should use.

This state of affairs has been a preoccupation of the IBA community for many years; however they have lacked the resources and coordination to make a concerted effort to improve the situation. As a result of this situation the users of PIGE are forced to implement a semi-quantitative approach. On the basis of the previous experience of other analytical techniques like RBS, EBS, NRA (that already have their data compiled in IBANDL) and NAA (which is reactor based) it is clear that the IAEA could play a unique and crucial role in meeting those needs.

Therefore, the purpose of the present Consultants' Meeting has been to bring together a group of PIGE experts in order to consider appropriate goals and identify possible actions that could be undertaken within the framework of IAEA/NDS structures and frameworks for improving the quality and availability of nuclear data for PIGE.

2. Goals / Scope of the Meeting

- Provide a general review of the status of the scientific activity in the field;
- Carry out an assessment of nuclear data needs for PIGE;
- Determine whether a CRP is the proper instrument to satisfy these needs;
- If a CRP is the proper instrument, determine the scope and the goals of that CRP.

3. Summary of Presentations

All presentations are available at http://www-nds.iaea.org/PIGE-CM/

3.1. PIGE and Resonant PIGE: Past Efforts and Current Needs, M. Kokkoris

Kokkoris divided his presentation into seven parts: in the first part, entitled 'historical aspects', he gave a brief outline on the history and evolution of the PIGE technique, he indicated that more than 200 accelerators in 40 countries are capable of implementing this technique. In the second part, 'status in literature', he elaborated the status of the publications in the field and their relative content noting that in many publications the term Nuclear Reaction Analysis (NRA) is used instead of PIGE. In the third part, 'present status', he described the present situation in PIGE with respect to other competitive Ion Beam Analysis techniques. There is also a brief description of the current situation and related problems concerning cross section data and analysis software, as well as on current trends in low energy nuclear physics laboratories where the majority of the measurements are performed not absolutely, but through comparison with standards due to the lack of a unified source of measured cross sections. In the fourth part, several interesting contributions from European laboratories were presented, while in the fifth, there is a long list of works originating from Greece related to PIGE and γ-ray measurements in general. In addition to the above, in the sixth part, there is a detailed description of the accelerator lab at NCSR 'Demokritos' in Athens, its experimental facilities, recent upgrades and capabilities, relevant to γ-ray measurements suitable for PIGE applications. Finally, in the last part, entitled 'future measurements and perspectives', Kokkoris recommended that the IBANDL database be used to store data for PIGE, he stressed that "…a new CRP is absolutely necessary to stimulate and organize the field" and described its possible content and the scientific problems that need to be addressed within its framework.

Comments:
All the participants congratulated Kokkoris on the excellent historical introduction of the subject.
Chiari: PIGE should be as easy to use as Particle Induced X-ray Emission (PIXE). You need a Database + a standard code, in that case it would be a true complementary technique, both are not destructive and can be applied to cultural heritage objects using an external beam.
Kokkoris: True, but PIGE can not be substituted for profiling.
Pedro de Jesus: One difficulty for the user of PIGE is that data are scarce and angular distributions of the emitted γ ray are difficult to find.

3.2. Development of a PIGE Analysis Methodology at ITN, A. Pedro de Jesus

The Ion Beam Laboratory at the Institute of Nuclear Technology (ITN) has always invested some effort in developing the founding basis of the ion beam analytical techniques that were implemented there. Among them, PIXE and RBS – also in association with channelling and with a micro-beam facility – have now international recognition. As examples of that effort,

PIXE work was preceded by a study of inner-shell ionization and also code development leading to DataPIXE. An integrated package for material analysis, NDF, was also implemented.
Following the same strategy, since 2000 the ITN laboratory has been investing in the development of a PIGE methodology not dependent on the use of standards, based on the ERYA code that calculates γ-ray yields, integrating the excitation functions of the relevant γ ray producing nuclear reactions. These functions, rarely available in the literature as numerical tables, required the measurement of several nuclear reactions.
Some details of the methodology and of nuclear reaction measurements were given, in particular measurements of Li, B, F, Na, Mg and Al for proton energy (E_p) up to 2.4 MeV, Be for $E_p \leq 1.7$ MeV and Li, F and Mg for $E_p \leq 4$ MeV. Furthermore, future work needing to be done in order to complement the existent work was indicated:

- Inclusion of profiling in ERYA;
- Extension of other excitation functions to higher energies e.g. 4 MeV;
- Coincidence methods to clean up the spectra;
- Special problems such as use of an underground laboratory to diminish background.

Comments:
Gurbich and others do not believe that an underground laboratory would diminish the background since most of it is beam-related.
Gurbich: Anticoincidence based on the β^+ emitters (that produce 2 annihilation γ rays) cleans the spectrum without affecting the statistics.
Kokkoris: The line at 6139 keV of fluorine, which is a background component, is sometimes beneficial if you know the fluorine content. It is like a self-calibration.

3.3. PIGE Activity in IPPE, A. Gurbich

For the analysis of carbon, sodium, aluminium and chromium, resonance PIGE has been employed. The excitation functions for the corresponding reactions were measured in the vicinity of resonances favourable for analytical applications. The oxygen analysis using γ rays from direct non-resonant radiative capture was also undertaken. PIGE was used for the analysis of various samples including semiconductor structures and nuclear reactor materials.

Hydrogen analysis using resonance $^{1}H(^{19}F,\alpha\gamma)^{16}O$ reaction was used to study hydrogen penetration into coating layers on the surface of zirconium pipes. Propagation of the spectrometer efficiency calibration on the high energy region was made using cascade gamma quanta from the resonance in the $^{27}Al(p,\gamma)^{28}Si$ reaction with known γ-ray branching. Tickonov's regularization method was applied to resolve the ill-posed problem of the determination of concentration on depth distribution.

A pulsed incident beam was used to substantially enhance the sensitivity of the PIGE analysis due to suppression of the background γ radiation. The advantages of the method were demonstrated for the case of detection of trace amounts of sodium in oxidized silicon samples.

Comments:
Gurbich: Tickonov's method is used when there are several resonances present at different sample depths. He remarked that ^{52}Cr is of technological importance and is difficult to analyze otherwise. He stressed that although there are some PIGE data already in IBANDL, there is a mistake in the representation of thick target data as they are shown as if they were thin target data, also sometimes the data are multiplied by 4π and other times not. In some cases like $^{23}Na(p,p'\gamma)$ there is a discrepancy of around 25% among the different authors.

Chiari: The discrepancy could be due to errors in the detector calibration.

3.4. PIGE for Cultural Heritage and Environmental Applications, M. Chiari

Particle Induced Gamma-ray Emission (PIGE) technique is an invaluable tool, complementary to Particle Induced X-ray Emission (PIXE), to quantify low-Z elements (Na, Mg, Al, Si, ...) in cultural heritage and environmental samples. At the LABEC laboratory of INFN in Florence, PIGE is routinely performed using a proton beam extracted in atmosphere from the accelerator beam line, thus reducing the risk of damage to the samples.

In the analysis of cultural heritage objects the unknown concentrations are typically deduced by comparing the γ-ray yields with those of thick standards of similar composition without knowing the cross section. The crucial point in such calculation is the difference between the stopping power of the unknown sample and that of the standard. Examples concerning the use of PIGE for Na detection to determine sodium concentration in ancient Roman glass mosaic tiles (where crusts and patinas strongly absorb the lowest energy X-rays) and to identify the presence of lapis-lazuli pigments in paint layers will be discussed. In particular, lapis-lazuli (a blue pigment, very precious, mainly composed of lazurite, $3Na_2O{\cdot}3Al_2O_3{\cdot}6SiO_2{\cdot}2Na_2S$) can not be identified by PIXE in canvas and wood paintings due to low energy X-rays absorption in the varnish and in the paint layer itself, and to signal interference from other pigments (such as lead white) that may be mixed with the lapis-lazuli to obtain different shades of colour.

As concerns environmental applications, in particular the study of atmospheric aerosols, PIGE measurements can be used to correct the underestimation of PIXE in quantifying the concentration of the lighter elements, like Na, Mg, Al and Si, due to X-ray self-absorption inside each individual aerosol particle (dimensions up to several micrometers). An accurate measurement of the concentration of crustal elements, namely Na, Mg, Al, Si, K, Ca, Ti, and Fe, is mandatory for the study of airborne mineral dust. Quantitative analysis of dust aerosols is needed since, on a global scale, mineral dust is one of the major components of atmospheric aerosols and has an important effects on the radiative budget of the atmosphere and thus on climate forcing. Examples considering the use of PIGE to analyse Al in dust aerosols collected in-flight over the Sahel desert (for climate change study) and in mineral dust particles archived in Antarctic ice cores (for paleoclimate research studies) were described.

Chiari showed the results of cross section measurements of proton induced γ-ray emission reactions on ^{7}Li, ^{19}F and ^{23}Na for beam energies in the 2 – 6 MeV range, carried out by his group in the past few years.

Comments:
Gurbich: Why do you work by comparing your measurements to standards? Why is there not a proper analysis code for PIGE?
Chiari: People are used to work with standards, in that way they circumvent the problem of not having reliable cross sections and the problems of efficiency calibration of their detectors, etc.
Gurbich: What would be the number of people interested in using a PIGE analysis code with a database of cross sections?
Kokkoris, Chiari: About the same number as SIMNRA (program for the simulation of backscattering spectra for ion beam analysis, see: http://www.rzg.mpg.de/~mam/Download.html) with a number of users around 300. Many people using PIXE would also be able to use such a PIGE code.
Chiari: The detection techniques are evolving in the direction of making PIGE easier; PIGE could be more generally used if data were available to support the analysis.

4. Identification of Nuclear Data Needs

Preliminary survey of the data (see Bibliography, Appendix 3) showed:

- Discrepancies among different authors;
- Lack of data for some important nuclear reactions;
- Insufficient coverage of energy ranges needed for PIGE applications (i.e., up to 5 MeV for proton beams);
- Lack of up-to-date, comprehensive compilation dedicated to the IBA community.

Therefore, the following data needs emerged:

a) Modification of the existing IBANDL database to accommodate data for PIGE in a convenient way for IBA users.
b) Compilation of existing data into IBANDL.
c) Assessment of available data and determination of a priority list for new measurements to complement, extend or improve the quality of the existing ones.
d) Measurement of cross sections mainly related to the analysis of the most important elements for interdisciplinary applications such as sodium, aluminium, boron, fluorine, magnesium, sulphur, beryllium, silicon, nitrogen, oxygen and chromium, using light ion beams. It is desirable that coordinated cross section measurements are undertaken at different IBA laboratories within the framework of a concerted effort.
e) Evaluation of compiled and measured data.
f) Validation of cross section data through benchmark experiments.
g) Development of a unified code using the extended IBANDL database for the implementation of the PIGE analytical technique.

5. Intended Outputs and Outcomes of Proposed CRP

Suggested Outputs:

- Determination of the proper format to store PIGE data in the IBANDL database;
- Compilation of previous data;
- Assessment of the previous data and elaboration of priority list for new measurements;
- New measurements;
- Study the feasibility of evaluation;
- Benchmark experiments;
- A review paper.

Outcomes:
The aim is to increase the analytical capability of the PIGE technique, both for bulk analysis and high-resolution depth profiling, making it fully quantitative and facilitating the access to the available cross section data.

6. Recommendations from this Meeting

It is strongly recommended that the IAEA should initiate a CRP on “Development of nuclear data for PIGE for analytical applications”, if possible with the first RCM in early 2011. This future CRP should address all the nuclear data needs listed in Section 4 above. Taking into account the broad scope of this CRP and the large interest within the IBA community, it is important that as many of the interested Member States as possible participate in the CRP. Therefore, it is recommended that the IAEA advertises this project as widely as possible. A preliminary list of potential institutes is presented in Appendix 4.

International Atomic Energy Agency
Consultants' Meeting on
Assessment of Nuclear Data Needs for PIGE

IAEA Headquarters, Vienna, Austria
1 – 2 March 2010
Meeting Room B0513

AGENDA (draft)

Monday, 1 March

08:30 – 09:30	**Registration (IAEA Registration Desk, Gate 1)**
09:30 – 10:15	**Opening Session**
	Opening Remarks and Welcome (R. Forrest) Introduction: Objectives of this Meeting (D. Abriola) Election of Chairman and Rapporteur Discussion and Adoption of the Agenda (Chairman)
10:15 – 11:00	*Coffee break and Administrative Matters*
11:00 – 12:30	**Participants presentations**
	1) "PIGE and resonant PIGE: Past efforts and current needs" M. Kokkoris (30+15 min) 2) "Development of a PIGE analysis methodology at ITN, Lisboa" A. Pedro de Jesus (30+15 min)
	Coffee break as needed
12:30 – 14:00	*LUNCH*
14:00 – 17:30	**Participants presentations**
	3) "PIGE activity in IPPE" A. Gurbich (30 +15 min) 4) "PIGE for cultural heritage and environmental applications" M. Chiari (30+15 min)
	Round Table: Nuclear Data Needs for PIGE
	Coffee break as needed

Tuesday, 2 March

09:00 – 12:30	**Round Table**
	Database formats, experimental, compilation, assessment, evaluations needs, etc.
	Coffee break as needed
12:30 – 14:00	*LUNCH*
14:00 – 17:30	**Drafting of the Consultants' Meeting Report** Closing of the meeting
	Coffee break as needed

Consultants' Meeting on
"Assessment of Nuclear Data Needs for PIGE"
IAEA Headquarters, Vienna, Austria
1 to 2 March 2010

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