Assessing Near Surface Diffusion of Volatile Elements in Nuclear Fuels and Nuclear Fuel Surrogates

R.J.W. Frost[1,*], N. Wikström[1], D. Adrovic[1], K.D. Johnson[2], D.A. Lopes[3], J. Valdez[4], E. Sooby[4], S. Patnaik[5,6], M. Giamouridou[5], E. Charatsidou[5], J. Oscarsson[7], P. Olsson[5], D. Primetzhofer[1,7]

[1] *Department of Physics and Astronomy, Uppsala University, Uppsala, SE-751 20, Sweden*
[2] *Westinghouse Sweden AB, Sweden*
[3] *Nuclear Energy and Fuel Cycle Division, Oak Ridge National Laboratory, USA*
[4] *Department of Physics and Astronomy, University of Texas at San Antonio, San Antonio, TX USA*
[5] *Department of Physics, KTH Royal Institute of Technology, Stockholm, Sweden*
[6] *Idaho National Laboratory, Idaho Falls, ID, USA*
[7] *The Tandem Laboratory, Uppsala University, Uppsala, Sweden*

The diffusion of gaseous fission products such as Xe and Kr in nuclear fuel represents a significant performance and safety parameter due to their high fission-yields, particularly high neutron absorption cross-sections and the potential to create pressure build up within fuel cladding [1]. The study of the diffusion behaviour of these noble gas species is, however, an experimental challenge, due difficulties in adding gas species to the fuel matrix as well as accessing techniques which can monitor gas concentrations and diffusion on nanometer scales. In the majority of cases, diffusion parameters are derived from irradiated materials [2], either through measurements in the plenum, or by annealing in Materials Tests Reactors and Post Irradiation Examination facilities. Although effective, the use of irradiated material requires significant infrastructure and resources to operate, and thus limits sample throughput. Presented here, is an extension of previous work [3] to combine medium energy ion implantation with time-of-flight elastic recoil detection analysis, to investigate the diffusion of volatile elements in high-density sample matrices including surrogates and nuclear fuels. ZrO_2, CeO_2, UO_2 and UN samples are implanted with Kr and Xe, and the depth-dependent distributions of these fission products (FP) are studied before and after thermal annealing. This work contributes towards providing benchmarking data for the validation of short-length scale atomistic models, such as density functional theory and molecular dynamics, which in turn contribute significantly to the Accelerated Fuel Qualification initiative [4]. The technique described here has the potential to significantly reduce cost and timescale of studying FP diffusion in nuclear fuels as well as aid in qualification, when compared to traditional methods.

References

[1] H. Bailly, D. Menissier, C. Prunier, The Nuclear Fuel of Pressurized Water Reactors and Fast Reactors: Design and Behavior, CEA, Eyrolles, 1996.

[2] G. Lawrence, A review of the diffusion coefficient of fission-product rare gases in uranium dioxide, Journal of Nuclear Materials 71 (1978) 195–218.

[3] R.J.W. Frost, D.A. Lopes, K.D. Johnson, S. Patnaik, J. Oscarsson, M. Sortica, P. Petersson, D. Primetzhofer, "Evaluating the diffusion of Kr in UO2 and ADOPT™ using Time-of-Flight Elastic Recoil Detection Analysis (ToF-ERDA)", (under review).

[4] K. Terrani, N. Capps, M. Kerr, C. Back, A. Nelson, B. Wirth, S. Hayes, C. Stanek, "Accelerating nuclear fuel development and qualification: Modelling and simulation integrated with separate-effects testing", Journal of Nuclear Materials (2020).

* Presenting author email: rob.frost@physics.uu.se

Study of the $^{11}B(p, a_0)^8Be$ and $^6Li(p, ^3He)^4He$ reactions in the low energy range, suitable for NRA purposes

E. Taimpiri[1,2,*], M. Axiotis[1], H. W. Becker[3], V. Foteinou[3], M. Kokkoris[2], A. Lagoyannis[1]
F. Maragkos[2,3]

[1] *Tandem Accelerator Laboratory, Institute of Nuclear and Particle Physics, N.C.S.R. "Demokritos", Aghia Paraskevi, 15310 Athens, Greece*

[2] *Department of Physics, National Technical University of Athens, Zografou campus, 15780 Athens, Greece*

[3] *Central Unit for Ion Beams and Radionuclides, Ruhr University Bochum, 44801 Bochum, Germany*

Boron (81.1% ^{11}B and 19.9% ^{10}B) and lithium (92.5% ^7Li and 7.5% ^6Li), two of the lightest elements in nature, are widely used in many technological as well as industrial applications. As a result, their detection and accurate depth profile determination, via Ion Beam Analysis (IBA) techniques, is important. Nuclear Reaction Analysis (NRA) constitutes the most effective of such techniques, as it results in isolated peaks (due to the generally high Q-values) with negligible background and also provides the possibility for simultaneous analysis of several low Z elements in near-surface layers of materials. The use of the NRA technique is further enhanced when the light elements under study co-exist in complex matrices. Especially, when these techniques are performed at projectile energies in the 100 keV – 300 keV range (Medium Energy Ion Scattering technique – MEIS), additional advantages are offered. More precisely, due to the limited penetration of low energy protons in matter, MEIS is highly sensitive to the surface layers of a sample, reaching depth resolutions on the order of a few nanometers. However, there is a significant lack of differential cross-section datasets, not only for low proton energies but also over a wide range of backscatter detection angles. The aim of the present work is to enrich the literature by providing reliable differential cross-section data in this uncovered area. For that reason, the $^{11}B(p, a_0)^8Be$ and $^6Li(p, ^3He)^4He$ reactions were thoroughly studied in a wider energy and angular range.

The experiments were conducted at the 4MV Dynamitron Tandem Laboratory of the Central Unit for Ion Beams and Radionuclides (RUBION) of the Ruhr University Bochum in Germany. For proton energies of 150 keV up to 340 keV the 450 kV single-stage accelerator was used, with the measurements being extended up to 1000 keV at the 4 MV Dynamitron Tandem accelerator. The energy step varied from 2 keV to 20 keV. For the detection of the charged particles, three Silicon Surface Barrier (SSB) detectors were placed at the angles of 130°, 140° and 160° and two Passivated Implanted Planar Silicon (PIPS) detectors at 150° and 170°. Two different targets were used for the two reactions under study. The first was a thin ^6LiF (95 % ^6Li & 5% ^7Li) layer evaporated on a ^{nat}C substrate, while the second one was a ^{nat}B thin layer, evaporated on a ^{nat}Cu substrate, with a top layer of Au. The obtained results of the present work were compared with existing ones found in the literature.

* Presenting author email: taimpiri@inp.demokritos.gr

Ion Beam Analysis of Medieval Enamelled Artefacts *

Zsófia Kertész[1†], Bernát Rácz[2], Máté Szarka[1], Anikó Angyal[1],
Boglárka Dönczö[1], Enikő Furu[1], Enikő Ivett Papp[1], Szabolcs Rosta[3], Zita Szikszai[1]

[1] *HUN-REN Institute for Nuclear Research, Debrecen, Hungary*
[2] *Central European University (CEU), Vienna, Austria*
[3] *Katona József Museum of Kecskemét, Kecskemét, Hungary*

In this work, the results of ion beam analysis of the Reliquary of Pétermonostora are presented. This exceptional masterpiece consists of two large enamel plaques and three decorated gilded copper strips. The Reliquary, dating from the 12th century, is most certainly a product of the rich Romanesque artistic culture of the Meuse Valley [1].

The objective of the ion beam analysis was to determine how the composition of the metal and enamel relates to the historical artwork centres of the Meuse Valley, Rhineland, Limoges, and Hildesheim and to compare our results with other IBA studies carried out on enamel artefacts of these workshops [2].

Particle induced X-ray (PIXE) and gamma-ray (PIGE) emission measurements were performed at the external microbeam of the ATOMKI 2MV Tandetron accelerator [3]. A focused proton beam with 3 MeV energy and 500-800 pA current was used for the irradiation of the selected measurement points. Elemental composition for $Z \geq 9$ was determined for all the different colours and metal decorations in the case of the two plaques and a smaller portrait of a saint also of Mosan provenience.

The results of the PIXE-PIGE analysis will be compared with micro-XRF measurements, and we discuss the differences arising from the two approaches in terms of the characterization of enamels.

Figure 1. PIXE-PIGE measurement of an enamel plaque of the Reliquary of Pétermonostora at the external microbeam of the ATOMKI TANDETRON accelerator

References

[1] Rácz, B.: The Reliquary of Petermonostora: A Twelfth-Century Mosan Phylactery Discovered in Hungary, MA Thesis, Central European University, (2023)
[2] Biron, I.: "Émaux sur Métal–du IXe au XIXe Siècle." *Histoire, technique et matériaux* (2015).
[3] Török, Zs, et al. Nuclear Instruments and Methods in Phys. Res. B **362** (2015): 167-171.

* The work was carried out in the frame of ERIHS-hu and was supported by GINOP-2.3.3-15-2016-00029.

† Presenting author email: zsofi@atomki.hu

KM3NeT material analysis with IBA at CIRCE

M.L.Mitsou[1,2,†] , C.Santonastaso[1,2],R.Buompane[1,2], L.Gialanella[1,2], P.Migliozzi[2],F. Marzaioli[1,2], C.M.Mollo[2], D. Vivolo[1,2], M. De Cesare[3,2], F.Molitierno[1,3]

[1] *Mathematics and Physics Department, Campania University, L. Vanvitelli,viale Lincoln, 5, Caserta,Italy*

[2] *INFN (National Institute for Nuclear Physics), Napoli section,via Cinthia, Napoli, Italy*

[3] *CIRA (Italian Aerospace Research Center), via Maiorise,1,Capua (Caserta), Italy.*

The KM3NeT detection units, located in the deep-sea water of Mediterranean Sea, operate in harsh conditions of salt water and hydrostatic pressure up to 400 bar. The equipment has been carefully designed to operate in such environment for an estimated lifetime of 20 years. The prediction of a possible extension of the expected lifetime requires a study of the long-term penetration through microcracks and water diffusion. In this contribution we present the experimental study performed with the 3 MV Tandem Accelerator at CIRCE (Center of Isotopic Research on Cultural and Environmental heritage), Department of Mathematics and Physics, Campania University "L. Vanvitelli", Caserta, Italy through NRA (Nuclear Reaction Analysis), a technique able to investigate the concentration depth profile of a single isotope by inducing nuclear reactions. Here, we present the results on water diffusion in the Low-Density Polyethylene (LDPE) matrix of the Vertical Electrical-Optical Cables (VEOC) [1]. Usually, the water and hence the hydrogen depth profile is measured with NRA exploiting the reaction $^{15}N(p,\alpha\gamma)^{12}C$ [2]. However, since the polyethylene contains a significant fraction of hydrogen, the depth profile has been investigated by the reaction $^{15}N(d,p)^{16}N$ induced on the polyethylene samples exposed to deuterated water. The samples were exposed to D_2O up to 400 bar in a small volume hyperbaric chamber. This chamber was used for three expositions, one of 80 days, one of 7 days, and the latter of 40 days. The reactions were induced on both exposed and not exposed samples using accelerated ^{15}N beams in an energy range between 7 MeV and 15 MeV. The radiations emitted from the interactions were detected using two NaI scintillators in close geometry. The results indicate a sufficient sensitivity to the diffusion process, allowing us to constrain the diffusion model parameters.

References

[1] S.Pilli *et al*., *Mechanics of time Dependent Materials*, Measuring time dependent diffusion in polymer matrix composites (2012).

[2]W.A Lanford *et al.Nuclear Instruments and Methods in Physics Section B: Beam Interactions with Materials and Atoms*,Analysis for hydrogen by nuclear reaction and energy recoil detection (1992).

† Presenting author email: marialucia.mitsou@unicampania.it

Applications of ^{53}Mn and ^{60}Fe Accelerator Mass Spectrometry at CologneAMS

Timm-Florian Pabst[1], Gereon Weingarten[1], Markus Schiffer[1], Steven Binnie[2], Alfred Dewald[1], Tibor Dunai[2], Abdallah Karaka[1], Stefan Heinze[1], Erik Strub[3], Dennis Mücher[1]

[1] *University of Cologne, Institute for Nuclear Physics, Zuelpicher Str. 77, 50937 Cologne, Germany*

[2] *University of Cologne, Institute for Geology and Mineralogy, Zuelpicher. Str 49a, 50674 Cologne, Germany*

[3] *University of Cologne, Division of Nuclear Chemistry, Zuelpicher Str. 49b, 50674 Cologne, Germany*

Medium-heavy cosmogenic radionuclides provide valuable tools for age determination and astrophysical studies. Measuring such nuclides via Accelerator Mass Spectrometry (AMS) is currently only possible by utilizing tandem accelerators with high terminal voltages of 10 MV or more. While machines of that size get less common, there is still a variety of applications of measuring nuclides like ^{53}Mn and ^{60}Fe. In this talk we will discuss the measurement of ^{53}Mn and ^{60}Fe at the 10 MV AMS facility in Cologne.

Due to ^{53}Mn's half life of 3.74 Ma, the combination of ^{53}Mn/^{3}He has the potential to extend the cosmogenic nuclide burial dating beyond the limits of the commonly used ^{26}Al/^{10}Be pairing. Particularly in hyper-arid and arid regions where fossil-based dating is extremely limited, the measurement of ^{53}M/^{3}He concentrations in iron-titanium oxides can be used to date sedimentary deposits into the Miocene.

The Colognes tandem accelerators high terminal voltage, combined with a gas-filled magnet, allows for highly sensitive detection of ^{53}Mn. Systematic optimization, including the development of a Bragg detector and enhanced stability of the accelerator voltage via active slit control, has resulted in stable conditions for the measurement of ^{53}Mn, achieving a low blank level of ^{53}Mn/^{55}Mn < 10^{-13}.

In addition to ^{53}Mn, we will discuss the measurement of ^{60}Fe, an isotope of great importance for understanding galactic processes. For investigating the frequency of supernovae reaching earth, ^{60}Fe can play a role in discovering the remnants of these events. Furthermore the neutron capture rate of ^{59}Fe(n,γ)^{60}Fe is important for constraining the ^{26}Al/^{60}Fe ratio in the galaxy. Here AMS is an opportunity of adding a direct measurement to further minimise restricting uncertainties.

This presentation will outline the challenges, technical innovations, and implications of using AMS for the measurement of ^{53}Mn and ^{60}Fe in both geochronological and astrophysical contexts.

Presenting author email: tpabst@ikp.uni-koeln.de