

Preface

Nuclear materials and their environments require analyses before and during their utilization as well as after service during disposal not only from the current nuclear units but also from planned or foreseen nuclear installations or systems.

Prior analysis, sampling, and sample treatment must be carried out when the analytical technique is not applied in situ, in a non-invasive way, or in an in-line or on-line mode.

The analysis may be carried out in situ, for example using a remote system, or in an underground laboratory in the phase under consideration. The analysis may also be done *ex situ* with transfer of the sample and separation when needed. For all analyses, sample volume, mass or amount, the flux of reagent, the size of the analyzed part of the sample and the acquisition time or time of analysis are key parameters that may affect the detection limit.

The information required—such as the chemical or radioisotope activity, the mass or volume of the sampled and analyzed item, the concentration as fraction or molarity of dopants or contaminants and the type or size of structures in the studied phases—has to be determined in a multi-scale approach at the nuclear scale, atomic or molecular scale, at the microscopic or macroscopic structural scale, at the bulk scale, at the component or system scale, and/or at the environmental or geographical scale according to the requirements of the study. Identification concerns the actinides, fission products or activated products as isotopes or elements, but also their speciation that may not only be done at the molecular scale but also in a broader sense such as at the environmental level. The time scale ranges from the femtosecond, accessible during Free Electron Laser investigations to describe ultra-fast phenomena, through the nanosecond to the mega-second, then to the giga-second or penta-second the timescale of uranium-238 half-life or of the age of the fossil natural geo-reactors. The explored energy range along the analytical methods goes also from the nano-eV (Mössbauer or nuclear magnetic resonance spectroscopy) to the giga-eV (muon-tomography), for example.

Passive and active analytical methods have been revisited in this work, with examples of their utilization in transmission, injection, diffusion or reflection modes. The sampling area, beam size and reagent quantities are either macroscopic, microscopic or nanoscopic in size, while spatial-temporal conditions make excitation incidence versus detection directions possible through solid angles, with synchronous detection or with temporal delay.

In this work the investigated analytical techniques have been classified according to their interactions, if any, between incident waves, particles or injected reagents and the analyzed sample, and, for their detected or recorded signals. For passive techniques, excitations are absent and phonons, photons, leptons, neutrons or ions are detected or quantified for their energy, flux, activity, quantity or mass. For interactive techniques, irradiations or reagent additions are made with phonons, photons, leptons, neutrons or ions with a known energy, flux, activity or mass. The irradiation or injection is done locally while the reception may be carried out in a given space at a given angle from the stimuli direction or the incident beam, instantaneously or after a certain delay after irradiation.

The detection tools are spectroscopy, microscopy, radiography, and tomography. The reaction takes place within or without a specific field such as electrical, magnetic, flow or mechanical acceleration. The detected signal may be the same in nature as the incident one, with the same energy, elastic interaction, or a signal with lower energy and inelastic interaction, with particles being again phonons, photons, leptons, neutrons or ions. In addition to these analytical tools or techniques, neutral species such as atoms or molecules may also be used to interrogate the material. They are treated as ions from a mass and charge point of view. The techniques are classified according to increasing energy of reagents or incident particles or waves. The combination of all excitation or reagent addition and product detections makes the analytic potential very rich to perform the identification of molecules, elements or isotopes, their quantitative determination, and their spatial speciation.

There has been an optimization of techniques and the discovery of new analytical tools over the last century. Some of the techniques are found today to be obsolete, others re-emerge due to new interests; some may be completed by combining the potential of one technique with another. In addition, there has been a constant challenge in pushing the use of the analytical techniques toward lower detection limits, better lateral and depth resolutions, more extreme applications and more flexible uses.

As far as the nuclear materials are concerned, studies must reflect the demanding conditions of temperature, pressure and irradiation under which they are used. These materials act as barriers and their properties are investigated with emphasis on mechanical performances, durability, plasticity and stability when damaged or loaded by dopants or contaminants. These materials range from fuels for thermal or fast reactors, to structural materials. Fuels are analyzed prior to and after irradiation, after their reprocessing for recycling and later as waste forms. Macro-properties such as thermodynamic, thermophysical and mechanical as well as microstructural analysis of these materials have to be analyzed, for example comparing again properties prior to and after irradiation.

As far as the environments of nuclear materials are concerned, one has to think about the way the analyst and the environmental scientist would collaborate together to produce data that can be used by modelers or by authorities. The challenge is to understand the behavior of actinide elements, fission products and other contaminants in the environment. Biogeochemical pathways have to be described, quantified and understood. Transport of actinides, fission products and other contaminants in fluids such as air or water include particulate or colloidal phases. These analyses must be integrated in the analytical strategy as specific species for modeling their biogeochemical behavior.

Data are provided by the analysts for the scientists and the modelers. The problem is to understand the behavior of radionuclides in the systems or the material properties with regard to its integrity. In the environment, contaminant pathways have to be described. The contaminated systems interact with the local environment that may modify radionuclide speciation by physical-chemical processes. The analytical results must be integrated in the study for modeling their chemical and physical properties.

A challenge for future investigations will be to find and develop direct analytical probes for full nuclear material characterization at very low defect, dopant or contaminant concentrations to better characterize the damages, species or structures and to predict their behavior in homogeneous, heterogeneous or complex nuclear materials and in their environments.

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