

Experiment Proposal

1.Characterization of nuclear ceramics (zirconia,spinel and silicon carbide) using RBS and NRA techniques

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Nuclear waste management will be one of the most difficult challenges facing industrialized countries during the coming decades. One possible option for the elimination of the excess plutonium arising from nuclear power plants and dismantled nuclear weapons is its incineration in a light-water reactor. To that purpose, the concept of an inert matrix fuel has recently been advanced. Crystalline ceramics, particularly zirconia, spinel and SiC were identified as potential candidates due to their high melting point, good thermal conductivity, absence of phase transformations at high temperatures, stability against radiation, good mechanical properties, oxidation resistance, low solubility in water, retention of radiotoxic elements and adequate neutronic properties. Rutherford backscattering spectrometry with light ions, typically $1-3\text{ MeV }^1\text{H}$ or ^4He ions, is a often used technique for depth profiling of elements concentrations. Furthermore, under channeling conditions, the technique may be exploited to give information about the sample structure and lattice locations of impurities. Channeling is a powerful technique which is widely used for ordered samples characterization.

In the extensive use of elastic backscattering for materials characterization purposes, ^4He particles up to several MeV have been for long considered as most convenient projectile. This often gives sufficient mass and depth resolution. However, the case of more complex film structures, with compound materials, have put higher demands on both the mass and the depth resolution in the analysis.

The RBS technique has also its limitations. Mass resolution for heavy elements and sensitivity for light elements are poor, and, except for the surface, mass determination is not unambiguously possible. The analysis of light elements in a heavier matrix is often impossible, because of the energy overlap of the beam ions scattered by light surface atoms and by heavier bulk atoms deeper in the sample. Furthermore, small amounts of light elements are difficult to analyze, because of the Z^2 dependence of the Rutherford cross section.

It is well known that the mass and depth resolution, as well as the sensitivity may be improved by using heavier and more energetic ions. In particular, mass separation for medium and heavy elements is improved significantly by heavy ions RBS. The expression for the energy separation as a function of the projectile mass M_1 , the projectile energy E_0 and the target mass M_2 can be written as:

$$\Delta E \cong \frac{2M_1E_0}{M_2^2}(1 - \cos\theta) \quad (1)$$

where θ is the backscattering angle. This is valid for $M_1/M_2 \ll 1$. The expression suggests the use of higher mass projectiles and higher bombarding energies. There are, however, some drawbacks,

because the resolution of the silicon detector is worsened and, due to $1/E_0^2$ dependency of the cross section, the counting rate reduces. Subsequently, longer analyzing time has to be used if higher energies are needed. In order to avoid the worsened resolution of silicon detectors for heavy ions the measurement of backscattered ions energy using a time of flight spectrometer can be used.

A severe disadvantage of conventional RBS is low sensitivity for light elements. The Rutherford scattering cross section is proportional to the square of the nuclear charge of the target nucleus. Therefore, the scattering peaks from light elements such as C, N and O are superimposed on a relatively high background due to backscattering from heavy elements in the sample. In recent years, high energy ^1H and ^4H backscattering has been utilized to overcome this difficulty and to quantify the stoichiometry or to profile the light elements in the heavy bulk samples. In the high energy backscattering experiments, ^1H and ^4He ions of 3–9 MeV (or even more) are used as incident projectiles. The elastic scattering cross section for light elements becomes a nuclear rather than a Rutherford interaction, called non-Rutherford backscattering or nuclear resonance elastic scattering. The non-Rutherford backscattering can be used to enhance the sensitivity for light elements. For example, at ^4He energies of 3.045, 4.265 and 3.72 MeV the elastic backscattering cross sections for O, C and N are 25, 150 and 6 times larger than their corresponding Rutherford cross sections, respectively.

The nuclear ceramics will be deposited by laser ablations (Pulsed Laser Deposition – PLD) as thin films with thickness between some hundreds of nanometers and a few microns.

The PLD method involves the interaction between the laser beam and the target's material: the laser beam heats, melts, evaporates the material, a plume expansion to the normal target's direction is following and produces a plume into material's vapours. Through this plume, the material is transferred on the substrate as a thin film. The depositions can be made in vacuum or gas atmosphere, and the substrate can be heated.

The pulsed laser depositions is a versatile and flexible technique, having a intrinsic advantage to obtain complex oxides as thin films, in particular the stoichiometric compounds transfer from the target to the substrate. An important advantage is the possibility to decrease the deposition temperatures of those compounds, due to the high-energy species generated in the ablation plume.

For a considerable improvement of the thin films surface, as well as crystallinity, to the PLD method, a radio frequency (RF) discharge can be added. This discharge leads to the reactivity increasing on the substrate, due to the ionised and excited species coming from the RF beam, and which acts on the thin layer in the space between the laser pulses, when the atoms are settled down to form a thin film.

This technique was proved to be useful to obtain thin films from different materials: metals, supraconductors, semiconductors, and biocompatible materials.

The experimental deposition set-up contains a Nd:YAG laser with four harmonics (266 nm, 355 nm, 532 nm and 1064 nm), a reaction chamber, a pumps system that can go down to 10^{-6} mbar, a target's rotation-translation system and a heater that can go up to 850°C.

Starting from a zirconium metallic target in oxygen reactive atmosphere, ***ZrO₂ (zirconia)*** ceramics layers will be obtained by laser ablation. The deposition parameters as laser wavelength, laser fluence, oxygen pressure and RF power will be varied.

Another ceramic compound that can be obtained either from a MgAl_2O_4 target ablated in argon or oxygen environment or from alternative ablation of magnesium and aluminium targets in oxygen atmosphere, is ***MgAl₂O₄ spinel***. The deposition conditions will be varied in order to obtain MgAl_2O_4 crystalline thin films with properties suitable for the previous propose applications. The silicon carbide (SiC) thin films will be deposited by PLD or RF-PLD, from Silicon and Carbon targets in vacuum or argon atmosphere; the influence of deposition parameters has been investigated.

We intend to use RBS and NRA techniques to characterize the thin layers of the above mentioned materials. Both RBS with 10 MeV carbon and NRBS with 4.5 MeV ^4He will be used. The

measurements will be performed using a dedicated target chamber. The experiments will be performed at our Tandem using a standard backscattering setup. The energy of the ^4He beam used for measurements will be calibrated by resonances. The ions scattered at 167° will be detected by a Si detector having 17 keV resolution. The results will serve also for two research projects PN 09 37 01 05 and 71-103.

2.Characterization of materials using RBS and NRA techniques

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The importance of RBS analysis on $\text{Sr}_x\text{Ba}_{1-x}\text{Nb}_2\text{O}_6$ and $(\text{Na}_{1/2}\text{Bi}_{1/2})\text{TiO}_3\text{-BaTiO}_3$ thin films growth by PLD and RF-PLD

Strontium barium titanate, $\text{Sr}_x\text{Ba}_{1-x}\text{Nb}_2\text{O}_6$ (SBN:x), is a ferroelectric material with interesting optical and electrical properties, having a relatively complicated crystal structure of tetragonal tungsten-bronze type (TTB). Because of high cost of single-crystal production, the growth of epitaxially SBN thin films has become very attractive. There are many reports in the literature about the properties of bulk SBN, but the problems which appear for SBN in thin film form are far from being clarified.

Sodium bismuth titanate in solid solution with barium titanate - $(\text{Na}_{1/2}\text{Bi}_{1/2})\text{TiO}_3\text{-BaTiO}_3$ belong to a new class of lead-free ferroelectrics which are design to replace the most used ferroelectric, $\text{PbZr}_{(1-x)}\text{Ti}_x\text{-PZT}$, mainly because of environmental reason. The European Union has already some dead-lines (2012 for the directive WEE regarding the use of hazardous elements) for using PZT in electronic telecom industries. For NBT-BT thin film form, there are few publications but the results are far from being comparable with those in bulk form, the main problem is to reproduce the complex stoichiometry of the target into the thin films, otherwise the dielectric and ferroelectric properties are not very good.

By using pulsed laser deposition and radiofrequency assisted pulsed laser deposition, we are able to grow highly oriented strontium barium niobate ($\text{Sr}_x\text{Ba}_{1-x}\text{Nb}_2\text{O}_6$) and NBT-BT thin films on

MgO (100) and Pt/Si (100), starting from stoichiometric single-crystal and ceramic targets, respectively.

The composition of the SBN and NBT-BT thin films plays an important role in tailoring the properties of thin films for a specific application. In this context, the RBS method is suitable to show that the films have stoichiometric composition identical to the target material. By controlling the stoichiometry of the thin films, we can achieve the scientific base necessary to publish several papers in important ISI journals.

We intend to use RBS and NRA techniques to characterize the thin layers of the above mentioned materials. Both RBS with 10 MeV carbon and NRBS with 4.5 MeV ^4He will be used. The measurements will be performed using a dedicated target chamber.