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Material engineering to fabricate rare earth erbium thin films for exploring nuclear energy sources

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ABSTRACT

High vacuum evaporation and cold-rolling techniques to fabricate thin films of the rare earth lanthanide-erbium have been discussed in this communication. Cold rolling has been used for the first time to successfully fabricate films of enriched and highly expensive erbium metal with areal density in the range of 0.5–1.0 mg/cm^2. The fabricated films were used as target materials in an advanced nuclear physics experiment. The experiment was designed to investigate isomeric states in the heavy nuclei mass region for exploring physics related to nuclear energy sources. The films fabricated using different techniques varied in thickness as well as purity. Methods to fabricate films with thickness of the order of 0.9 mg/cm^2 were different than those of 0.4 mg/cm^2 areal density. All the thin films were characterized using multiple advanced techniques to accurately ascertain levels of contamination as well as to determine their exact surface density. Detailed fabrication methods as well as characterization techniques have been discussed.

1. Introduction

Rare earth elements have always found applications in a wide range of fields. They are extremely useful to conduct researches that have both, direct impact on the society's development as well as improve understanding of fundamental concepts. Thus, lanthanides such as praseodymium, neodymium, dysprosium, erbium and holmium find importance in basic research as well as applied research. Rare earth metal oxides with specific and unique optoelectrical properties have been used to develop oxygen-storage components, ceramic pigments, catalysts, low resistance materials, high-temperature fuel cell, gas turbines, diesel engines and photocatalysts [1–10]. In the field of water purification, for example, photocatalytic performance of the nanostructured praseodymium zirconate, praseodymium oxide, pure praseodymium cerate, neodymium oxide, holmium oxide and $Dy_2Sn_2O_7$-SnO_2 were studied for degradation of contaminants such as eriochrome black T dye, eosin Y, erythrosine, and methyl orange [11–17]. Also, lanthanide stannate ($Ln_2Sn_2O_7$) were found to be useful in various applications, like lithium ion batteries, electric conductors, and radioactive waste management [18–23]. Besides such applied research applications that directly benefit the society, lanthanides also aid basic research that helps understand complicated subjects: various isotopes of enriched neodymium oxide thin film sandwiched between carbon foils were used to investigate the behavior of optical model potential parameters around the Coulomb barrier [24].

Presently it has become imperative to develop multiple alternative sources of energy to cater to the need of an ever growing world population and industry. A lot of developmental work has been going on in the fields of renewable energy sources, like photovoltaic and wind farms. However, efficiently extracting energy from these has been a constant challenge and thus it is still an open field when it comes to proposing energy sources that are efficient, abundant as well as have a very high yield. One option, in which research has been going on since the last few decades [25] is to extract large amounts of energy stored in nuclear isomeric states. The transitional energies from such states are in the range of a few million electron volts which are emitted in the form of electromagnetic radiation (gamma rays), much higher than those emitted by chemical batteries or solar devices. In order to achieve a sufficient concentrated population of nucleons at a higher energy state for stimulated emission to lower energy states to occur, identifying excited long-lived nuclear states (isomers) is necessary. Developing devices which generate coherent radiation of ultra short wavelengths by stimulating nuclear transitions of sufficiently high energy and yield is a challenging task that requires in-depth basic research. Theoretical predictions suggest presence of multiple isomeric states in heavy mass

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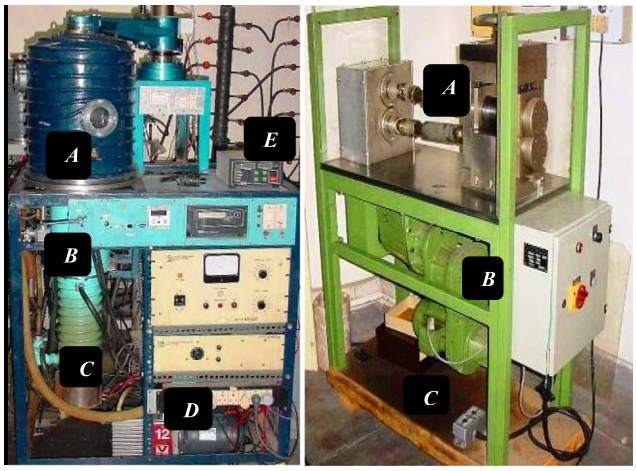

Fig. 1. (l) High vacuum thin film deposition system; labels: (A): bell jar, (B): LN_2 trap, (C): diffusion pump, (D): roughing pump, (E) deposition rate monitor (r) Rolling machine; labels: (A) specially hardened rollers, (B): motor control system, (C): pedal control.

region of the periodic table [26]. Nuclear physics experiments of stable elemental beams combined with rare earth metal targets can address the heavy mass region and increase knowledge about isomers present in this region. In order to enhance the understanding of nuclear structure dynamics close to the proton drip line, fabrication of the sparsely abundant (1.6%), 73.6% enriched erbium (mass 164) isotopic films were carried out to be used in an experiment. High vacuum deposition technique of physical vapor deposition was used for the first time to fabricate such films from their metallic form, without using any substrate heating [27] or oxide reduction methods [28,29]. Also, unlike previously ever reported, erbium's high malleability was exploited to cold roll the metal into thin sheets of superior quality, uniform thickness and stability, usable as target materials. The experimental design, where multiple nuclei (fragments) produced during the beam-target interaction need to fly through a mass recoil separator, required that the films be moderately thick. The fragments would lose too much energy while leaving thick films whereas too thin films would bring down the overall reaction cross-section.

Thin films were made using two major techniques and three different methodologies were tried out. The first technique of using high vacuum (low-pressure plasma environment) in an adiabatic deposition chamber ensured a reduced foreign particle count per volume in comparison to atmospheric pressure conditions. This technique ensures *(a)* comparatively lower number of inelastic impact processes with possible contaminants; *(b)* longer mean free path for the isotopes' vapor particles before condensation on the substrate material. A couple of methods were applied using this technique to fabricate films of the isotope with surface density ranging from 0.45–0.9 mg/cm^2. The second technique involved cold-rolling erbium sheets into very thin films of the order of 0.6–0.9 mg/cm^2 under atmospheric conditions, with great care keeping in view the metal's soft nature. The facility pictures of the different equipments available at the Target Laboratory of Inter University Accelerator Center (IUAC), New Delhi, India, are displayed in Fig. 1(l), (r).

In Section 2, discussed are the detailed practical techniques used to fabricate stable thin ^{164}Er films using both techniques—high vacuum deposition as well as cold-rolling under atmospheric conditions. Section 3 mentions characterization methods used to precisely measure thickness and purity levels. Section 4 includes conclusions about the film fabrication and characterization methods.

2. Fabrication of thin erbium films

The primary aim was to develop ^{164}Er films of a thickness appropriate for the experimental purpose (~700–800 μg/cm^2). As per previous literature available [27–29], as well as availability of facility at IUAC, the high vacuum evaporation thin film deposition technique was used. Owing to the very high cost of the enriched isotopic material due to low abundance, film fabrication methods with naturally abundant isotopes of erbium were tried before using enriched isotopes.

Fig. 2. Shredding of erbium from substrate during deposition.

2.1. High vacuum deposition

To obtain thin films by physical vapor deposition, diffusion pump based coating unit present at IUAC, as shown in Fig. 1(l), was used. This deposition unit consists of a resistive heating setup as well as a single pocket electron-beam gun with 2 kW power supply, facilitating the simultaneous fabrication of multi-layered thin films. A quartz crystal based deposition rate monitor working on the principle of piezoelectric oscillation frequency modulation [30] is present in-situ which assists in estimating the film thicknesses (in units of nanometer). In order to achieve high vacuum within the deposition chamber strong pumps are necessary; a roughing pump is augmented by an oil diffusion pump which in-turn is equipped with a cold trap where liquid nitrogen can be poured to avoid back-streaming of oil into the chamber. Typical vacuum levels ~10^{-10} bar can be maintained throughout cycles of depositing multiple layers of thin films onto the substrate material.

2.1.1. Self-supported erbium thin films

Attempts were made using the high vacuum evaporator to fabricate self-supported erbium thin films. A method was devised to counter the water-reacting issue of erbium by trying to float erbium thin films in an alcohol bath. It is worth mentioning that this activity was successfully undertaken in the past [29] using a method to reduce the oxide of erbium and then fabricating thin films using zapon as a coating agent. Here, the attempt was to coat glass slides with a thin film of a parting agent and then without breaking vacuum, using a second in-situ resistive heating setup to deposit erbium. Extensive trials were undertaken to realize this aim; however it was found that erbium does not stick to the parting agent. Both $BaCl_2$ and NaCl were tried as parting agents in this case. Fig. 2 clearly shows erbium shredding off of the substrate, displaying inability of the parting agent to hold on to an erbium layer.

As observed while trying to fabricate self supported films, erbium's electro-positivity results in its reaction with cold as well as hot water. Rapid reaction with water forming erbium hydroxide, $Er(OH)_3$, and hydrogen gas [$2Er + 6H_2O \rightarrow 2Er(OH)_3 + 3H_2\uparrow$] means that after the films are made, they cannot be efficiently floated off the substrate onto water. Hence to address this issue, it was thought wise to blanket the erbium thin film with carbon layers on both sides. Carbon has been shown before in many cases to be the ideal choice owing to its chemical stability as well as high sublimation point, which makes it safe to be used in cases of ion-beam bombardment experiments.

2.2.

2.2.1. Deposition of parting agent and carbon layer

A set of glass slides, ultrasonically cleaned to remove any impurity, was used as the solid base for thin films to be deposited upon. In order to