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Surface Wear Studies in Some Materials Using α-induced Reactions

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Abstract The radio-activity produced during the irradiation of ^{63,65}Cu, ^{59}Co, ^{93}Nb and ^{121,123}Sb targets with α-particles have been measured using activation technique. The yields of radioactive isotopic products ^{66,67,68}Ga, ^{61} Cu, ^{96g,m}Tc and ^{123,124,126}I have been determined in the energy range $\approx$ 10-40 MeV using stacked foil technique. Radioactive counting of samples was performed with a high-resolution gamma-spectrometer. As light ion beams produce an extremely narrow layer of activities in the surface of a material, these reactions may be useful for thin layer activation study.

Keywords: activation technique, excitation functions, alpha induced reactions, thin layer activation analysis.

1. INTRODUCTION

The thin layer activation (TLA) technique has become a widely used sensitive tool to study the surface wear of micron order in metallic surfaces in various engineering components. It is known that reliability of industrial equipments, transport systems, power plants both nuclear as well as conventional etc., is considerable influenced by the process of wear, corrosion and erosion. As such, the development of effective methods for the detection, measurements and monitoring of these processes are quite important. This is an important aspects in the sense that dangerous accidents during operation of industrial installation, and production losses may be avoided due to machine break-down. The charged particle activation for measurement and monitoring of corrosion and wear is important particularly when the parts of the surface are not really accessible. In TLA method, radioactivities in thin layers of the metallic foils are induced by bombarding them with light charged particles like protons, deuterons and α-particles. The incident charged particles may produce radio-active residues in the target material through nuclear reactions. The yield of the radio-active residues as a function of the depth gives the profile of surface wear. The yield of a particular radio-isotope may be determined from the measured cross-sections for that reaction responsible for the formation of the residue. The cross-sections for the production of

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residual nuclei by medium energy nuclear reactions are of importance not only for the development of reactor technology but also for many fields of basic and applied sciences including astrophysics, environ- mental sciences, medicine, accelerator technology, space sciences etc. More recently, these cross-sections are also in demand in order to know the transmutation probabilities required for the proposed accelerator driven systems (ADS) popularly known as energy amplifiers [1]. The nuclear data requirements for the accelerator driven technologies are extreme with respect to both the target element coverage and type of reaction data. As such, more detailed and accurate measurements are needed to provide these data. The TLA technique requires the accurate knowledge of charge particle induced reaction cross-sections. Several investigations are found in literature, where attempts have been made to determine these reaction cross-sections. [2,3] have studied the excitation functions (EF's) for several isotopes of Sc, Cr, V and ^{nat}Zr and have obtained the calibration curves for TLA technique. Furthermore, [4], have also given the experimentally measured cross-sections for α-induced reactions on ^{nat}Pt. Attempts have been made [2,4] to explain the experimental data with calculations done using various computer codes. [5] have studied TLA of Cr with deuterons. They [5] have indicated that Cr is an element which is added in many metallic materials including coatings to improve corrosion resistance. The technological developments give rise to the use of Cr as a material for various application purposes. Similarly, various isotopes of Sn [6] are used in the treatment of tumour and in positron emission tomography (PET) imaging technique. As such, the TLA studies of various isotopes used in medical technology are widely useful. The light ion implanted samples have been used for the evaluation of metallic samples [7]. Information on the wear level of some engine components with additional data on transfer and adhesion of material have also been obtained [7].

In general, authors [6,8,9,10] have also made an attempt to verify the measured cross-sections using theoretical model codes. Though, considerable data is available in literature on nucleon and light ion induced reactions but the cross-section values measured by different groups of workers for the same reaction, generally, do not agree. Further, most of the older measurements have been done using detectors and electronics of low resolution. As such, there is a demand of new, reliable and self-consistent cross-section data taken by detectors of high resolution. Further, the value of cross-section for a specific reaction depends on the reaction mechanism and therefore, accurately measured cross-section data may also be used to test the reaction mechanism. In view of the above, a programme of precise measurement and analysis of cross-sections for the alpha-induced reactions in several elements has been undertaken [11,12,13,14]. In order to complete the TLA experiment and to calculate the amount of wear by using nuclear tracing technique, it is important to know the accurately measured EF's for the nuclear reactions of interest and the half-lives, γ-ray energies etc., of the produced isotopes. In the present work, TLA technique has been explored using gamma spectroscopy for a large number of reactions in several isotopes, which may be of interest for the reactor technology. The measured EFs have been used to provide

the practical yield curves and the activity versus depth distribution curves are obtained as a direct tool to deduce the surface wear of different materials.

2. EXPERIMENTAL DETAILS

Stacked foil activation technique has been employed for determining the yield of the radioactive isotopes at different energies. In this technique a stack of few sample foils is bombarded normal to the beam direction so that after passing through each foil of the stack the beam energy gets reduced and successive foils are irradiated at decreasing beam energy. The samples for irradiation were prepared from natural elements of spectroscopic purity better than 99.9 %. In case of $^{63,65}Cu$, ^{59}Co and ^{93}Nb targets the thin metallic foils were used. The samples of size 12×12 mm^2 and of thickness $\approx$5-30 mg/cm^2 were cut from these metallic foils. However, in case of Sb, the samples were prepared by vacuum evaporation on thin aluminum backing of thickness $\approx$ 6.75 mg/cm^2. The thickness of antimony deposition was $\approx$1 mg/cm^2. Samples were fixed individually on aluminum holders of 2.5×2 cm^2 having concentric holes of 1 cm diameter at their centre. Stacks of samples were irradiated separately using $\approx$40 MeV α-particle beams at the Variable Energy Cyclotron Centre (VECC), Kolkata, INDIA. In order to achieve wide energy variation, in some cases the Al degraders of suitable thicknesses sandwiched between the successive samples, were used. The beam currents of $\approx$100-400nA have been used depending on the individual case. Keeping in view the half-lives of interest, the stacks were irradiated for the optimum times ranging from about 30 min to few hours. After the irradiation, the activities of the radioactive products in each foil of the target stack were measured non-destructively using 100 c.c. HPGe detector coupled to a multi-channel analyzer. Different radio-active residues produced in the irradiated foils, have been identified from their characteristic gamma rays and measured half-lives. The efficiency versus energy curves of the detector for γ-rays of different energies were determined using various standard γ-sources including ^{152}Eu source of known strength at several source-detector separations. The measured intensities of the identified γ-rays from the residual nuclei of interest were used to compute the reaction cross-sections. The details of the experimental technique employed in the present work, the method of the data reduction and the details of error analysis are given elsewhere [11]. Relevant data on isotopes $^{66,67,68}Ga$, $^{96g,m}Tc$, ^{61}Cu and $^{123,124,126}I$ for application in thin layer activation technique is given in Table 1.

3. MEASUREMENT OF CROSS-SECTION

3.1. Yield curves

The yields of $^{66,67,68}Ga$, $^{96g,m}Tc$, ^{61}Cu and $^{123,124,126}I$ isotopes have been obtained, from the intensity of characteristic γ lines, at different energies of α-particles, i.e., at different depths from the stacked foil activation experiment carried out for the measurement of the cross-sections of the reactions.

Table 1: Relevant data on isotopes $^{66,67,68}Ga$, ^{61}Cu, $^{96g,m}Tc$ and $^{123,124,126}I$ for application in thin layer activation technique.

Isotope	Half-life	γ-ray		Reaction	σ*max*
		Eγ (keV)	Iγ (%)		(mb)
^{66}Ga	9.49 h	833.6,1039.4	6.1,37.9	$^{63}Cu\ (\alpha,n)^{66}Ga$	19.25
^{67}Ga	3.26 d	184.6,209	20.4,2.3	$^{65}Cu\ (\alpha,2n)^{67}Ga$	45.35
-	-	300.2,393.5	16.6,4.6	-	-
^{68}Ga	67.71 m	1077.3	3	$^{65}Cu(\alpha,n)^{68}Ga$	26.48
^{61}Cu	3.33 h	282.9,373	12.5,2.2	$^{59}Co\ (\alpha,2n)^{61}Cu$	23.07
-	-	588.5,656	1.2,10.7	-	-
-	-	908.6,1185.2	1.2,3.7	-	-
^{96g}Tc	4.28d	778.2,812.5	99.8,82	$^{93}Nb\ (\alpha,n)^{96g}Tc$	21.82
-	-	849.9,1126.8	98,15.2	-	-
^{96m}Tc	51.5 m	1200.2	1.1	$^{93}Nb\ (\alpha,n)^{96m}Tc$	51.20
^{123}I	13.23h	159,529	83.3,1.4	$^{121}Sb\ (\alpha,2n)^{123}I$	989.47
^{124}I	4.176d	602.7,722.8	61,10	$^{121}Sb\ (\alpha,n)^{124}I$	215.0
^{126}I	12.93d	388.6,666.4	34.1,33.1	$^{123}Sb\ (\alpha,n)^{126}I$	148.53

The cross-sections were measured in the energy range from threshold to ≈ 40 *MeV* by measuring the activities induced in the foils of the stack. The activities induced in each foil of the stack were used to deduce the yields of the isotopes of interest. The stopping power of α-particles has been used to obtain the incident energy on each foil. The yield of the nuclides ^{66,67,68} *Ga*, $^{96g,m}Tc$, ^{61}Cu and $^{123,124,126}I$ per micron thickness against the depth as well as incident energy are shown in Figs. 1-4, respectively. Since, a typical EF curve is smooth without any resonance structure, the experimental points were fitted using the smooth fitting option of the ORIGIN software. As can be seen from these figures, the curves obtained by fitting the data may be used to obtain the yield for a particular isotope at different depths in the material under consideration. In order to compute the net yield of a particular isotope in the thick target by the absorption of 40 *MeV* α-particles, the integral area of the curve from the front surface to the final depth from where the given isotope production starts (∼ threshold of reaction) has been obtained. As can be seen from these figures, the different isotopes of a given

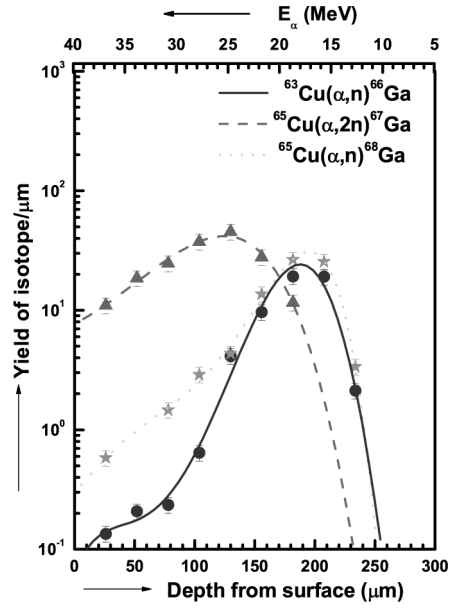

Figure 1: Yield curves of isotopes $^{66,67,68}Ga$ from Cu material.

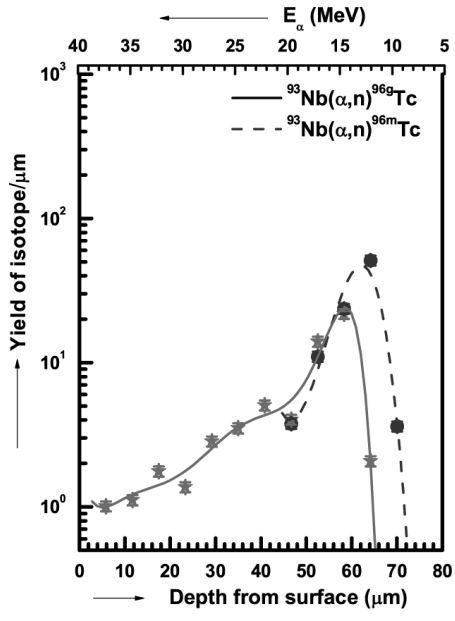

Figure 2: Yield curves of isotopes $^{96g,m}Tc$ from Nb material.

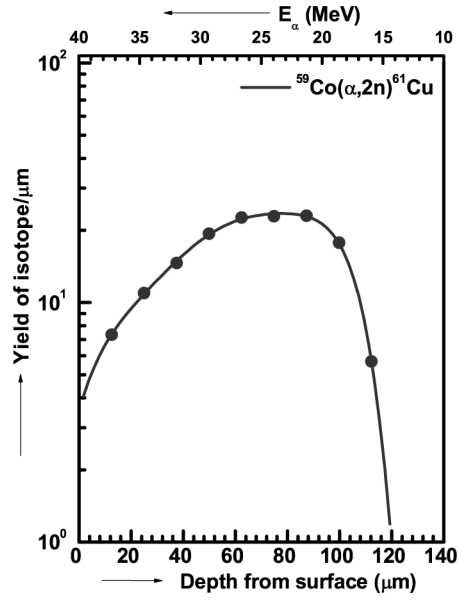

Figure 3: Yield curve of isotope ^{61}Cu from Co material material.

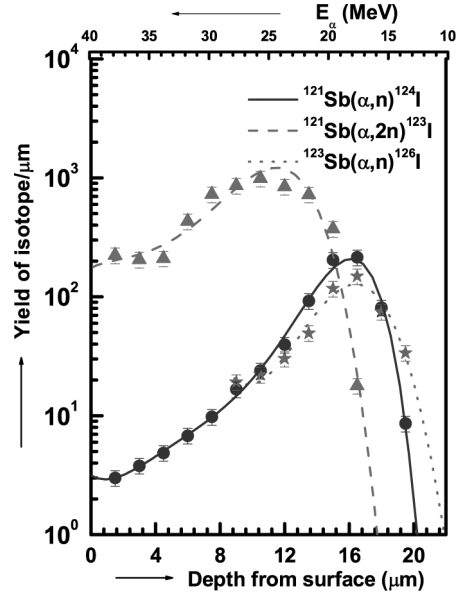

Figure 4: Yield curve of isotope $^{123,124,126}I$ for Sb material.

element have different yields depending on the reaction channel and the Q-value for the reaction. The area under the curves gives the total yield of the isotope generated in the thick target by absorption of 40 MeV energy of the αparticles.

3.2. Calibration curves and surface wear computation

As indicated in Figs. 1-4, the total yield of the isotope in the given material may be obtained from the integral area under the yield curves. After the removal of certain depth of material (decreasing the thickness corresponding to the each foil of the stack one by one) the percentage of the remaining activity is computed graphically and the calibration curves so deduced from the yield curves are shown in Figs. 5-8. As a matter of fact, these calibration curves may be considered to correspond to pure metals bombarded with 40 *MeV* α-particles normal to the beam direction. In practice, the actual material may not be 100% pure and α-irradiation may not be always possible at 90° angle of incidence. As such, transformations [15] may be required to be carried out on the X-axis of the calibration curves. Thus the material composition and the angle of incidence of the projectile to the surface of sample should be taken into consideration. If the surface of the samples has been irradiated at 90° angle of incidence, as in the present case, then only the transformation corresponding to the material composition may be carried out according to the composition of several materials by using Bragg formula [16], as given below,

$$\frac{1}{R} = \frac{f_1}{r_1} + \frac{f_2}{r_2} + \frac{f_3}{r_3} + \frac{f_4}{r_4} \ldots\ldots\ldots \frac{f_i}{r_i} \tag{1}$$

where, R is the range in the different materials, f_i the weighted fraction of the i^{th} component, and r_i the range in the i^{th} component.

If A is the total activity, and A_{n-1} is the activity after removing thickness δx_1 , then the ratio $A_{n-1}|A$ may be given as,

$$\frac{A_{n-1}}{A} = \frac{\sum_{i=2}^{n} \sigma_i \delta x_i}{\sum_{i=1}^{n} \sigma_i \delta x_i} \tag{2}$$

As can be seen from the above expression, the ratio is not depending on the number of target atoms, beam current and irradiation time. Thus the ratio A_{n-1}/A may be considered as the mathematical basis for determination of the quantity of material (thickness) removed due to surface wear in thin layer activation method.

The statistical variation of the count rates before and after the wear run may limit the sensitivity of the technique. In the present work, out of the nine isotopes viz, $^{66,67,68}Ga$, $^{96g,m}Tc$, ^{61}Cu and $^{123,124,126}I$, the yield of ^{123}I has been found to be maximum and that of ^{66}Ga is minimum as per the cross-sections given in Table 1. The sensitivity can

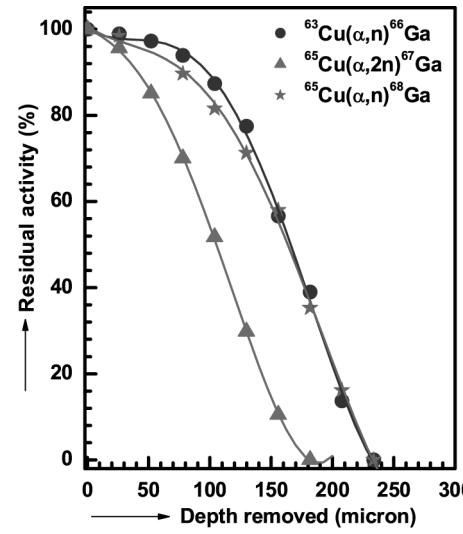

Figure 5: Calibration curves of isotopes $^{66,67,68}Ga$ from Cu material.

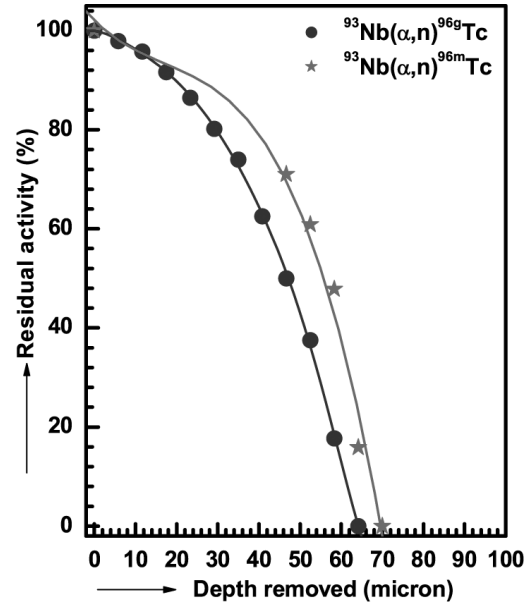

Figure 6: Calibration curves of isotopes $^{96g,m}Tc$ from Nb material.

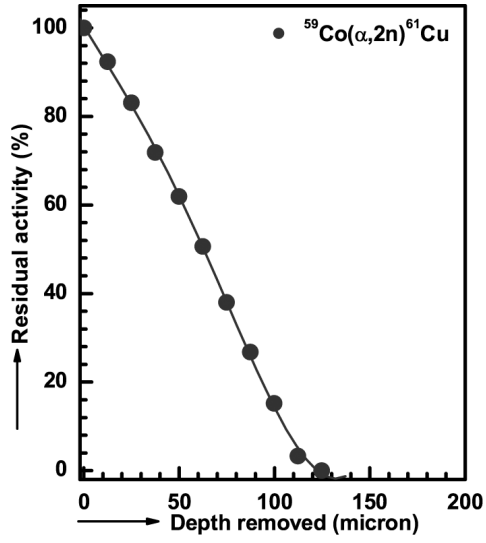

Figure 7: Calibration curve of isotope ^{61}Cu from Co material.

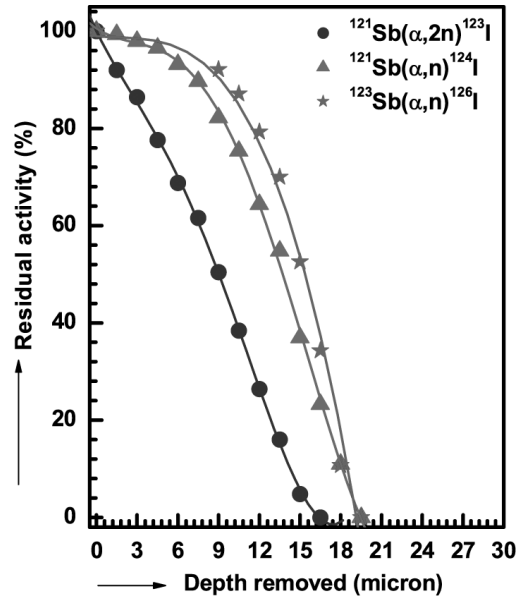

Figure 8: Calibration curve of isotope $^{123,124,126}I$ for Sb material.

be enhanced by changing the suitable experimental parameters, viz, increase in the beam current and time of irradiation. It may be pointed out that the beam current and the time of irradiation should be optimized so that the properties of the material are not affected after the irradiation. In this work $^{63,65}Cu$, ^{59}Co, ^{93}Nb and $^{121,123}Sb$ materials were irradiated to the beam current of ≈100-400 *nA* with a beam spot ≈5 mm diameter for ≈ 10 h, equivalent to $\approx 10^{14}$ ions/cm^2 , without almost any change in the properties, such as hardness, of the material, as evident from the physical appearance of the samples. Similar observations have also been reported in proton and deuteron induced irradiations. It may also be noticed from *Figs*. 5–8 that the rate of change of activity per unit thickness is more for the higher slope curve i.e., the errors associated in higher slope curve will be less. The overall error has been estimated from the errors of cross-section in generating the calibration curves and the activity measurement of the irradiated samples and has been found to be ≤ 5-15%.

4. CONCLUSION

In the present work an attempt has been made to develop the TLA technique using α-particles to determine the surface wear in the micron order in different materials. The calibration curves with percentage of residual activity against the removal of thickness may be utilized to investigate the surface wear of materials used in the nuclear power reactors. The investigation for the surface wear data of such materials used in power reactors is useful from the point of view of operational performance and safety of the plants.

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