

# Excitation Function of $^{27}\text{Al} + ^{20}\text{Ne}$ Reaction upto 166 MeV

Dibyasree Choudhury<sup>1\*</sup>, Susanta Lahiri<sup>2</sup>

<sup>1</sup>Department of Basic Science and Humanities, Institute of Engineering and Management, School of UEM, Kolkata 700091, India

<sup>2</sup>Department of Chemistry, Diamond Harbour Women's University, Diamond Harbour Road, Sarisha, South 24 Paraganas 743368, India

**Abstract.** Ultrapure aluminium foils are commonly used behind targets either as a degrader foil or for target-backing in nuclear physics experiments. Interaction of projectile with the aluminium foil is most likely to interfere with the radioisotopes produced by target-projectile interaction. In this study, eight aluminium foils were irradiated with  $^{20}\text{Ne}$  beam upto 166 MeV and the production cross section of the produced radioisotopes such as  $^{43,44}\text{Sc}$ ,  $^{41}\text{Ar}$ ,  $^{34m,38}\text{Cl}$ , etc., were measured. This experiment marked an alternate and cost-effective production route for the clinically important scandium radioisotopes. No-carrier-added  $^{43,44}\text{Sc}$  radioisotopes were selectively separated from bulk Al target and co-produced  $^{24}\text{Na}$  by liquid-liquid extraction technique using Di-2-(ethylhexyl)phosphoric acid (HDEHP)/HCl combination with a high separation factor  $>12,000$ .

## 1 Introduction

Nuclear Physics experiments preferentially require thin targets ( $\sim 1\mu\text{g}/\text{cm}^2$  to  $20\text{ mg}/\text{cm}^2$ ) to minimise energy loss and scattering of outgoing particles, thereby helping in cross-section measurements. Apart from its thickness, few other criteria also need to be fulfilled by the target, such as mechanical strength, uniform thickness distribution, stability and high chemical purity [1]. In order to support the thin and fragile target films, often backings are used in these experiments. Thin ultrapure aluminium foils (99.99% pure) are popularly used as the backing material. Alongside, the Al foils also work as energy degraders, catcher foil or target-heat dissipators [2, 3]. The frequent use of aluminium may be attributed to its easy availability, malleability, high stopping power, low neutron capture cross section and low background activities. Nuclear physicists primarily focus on the products generated by a target-projectile combination, even though the products arising from the used backing material (or the degrader foil, as the case may be) might interfere with the experiment under

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\* Corresponding author: [dibyasree.choudhury@iem.edu.in](mailto:dibyasree.choudhury@iem.edu.in)

study. Therefore, analysing the radioisotopes produced from the backing material (Al foil) is of utmost significance.

In the present paper, we have reported a detailed study of the radioisotopes formed by the irradiation of Al foils (used as target materials) with the heavy ion beam  $^{20}\text{Ne}$ . Heavy ion (HI) induced reactions provide direct access to the neutron-deficient isotopes via multiple charged particle emission, such as p,  $\alpha$ , heavier clusters, opening several reaction channels, thereby leading to valuable cross-section data, essential for optimising isotope production pathways [4]. Few research works using the  $^{27}\text{Al} + ^{20}\text{Ne}$  target-projectile combination are reported. In 1984, Kox et al [5] reported a total cross section of 2130 mb for the  $^{27}\text{Al} + ^{20}\text{Ne}$  reaction at the projectile energy of 30 MeV/nucleon. Another study recently reported the total charge-changing cross section as well as the partial cross section for fragment production of 388 A MeV and 400 A MeV  $^{20}\text{Ne}$  ions, respectively [6,7]. Kumar et al. [8] carried out excitation function measurements of  $^{20}\text{Ne}$ -induced reactions on  $^{27}\text{Al}$  from 50-150 MeV. In this paper, the production of radioisotopes such as  $^{43,44}\text{Sc}$ ,  $^{41}\text{Ar}$ ,  $^{34\text{m}}, ^{38}\text{Cl}$ , etc. was observed and the corresponding excitation functions were studied.

One of the produced radioisotopes,  $^{43,44}\text{Sc}$ , is clinically important, and the production route reported here may be viewed as a feasible, alternative production pathway for these radioisotopes. Radioscandium has attracted the attention of nuclear chemists as a potential popular positron emission tomography (PET) radioisotope. These radioisotopes are relatively long-lived, ( $^{44\text{g}}\text{Sc}$ ,  $T_{1/2} = 4.04$  h and  $^{43}\text{Sc}$ ,  $T_{1/2} = 3.89$  h), having simple decay scheme with a high  $\beta^+$  branching ratio. Additionally, meta-stable  $^{44\text{m}}\text{Sc}$  ( $T_{1/2} = 58.6$  h) can serve as an *in vivo*  $^{44\text{m}}\text{Sc} - ^{44}\text{Sc}$  generator with its ground state [9]. The significance of the positron-emitting  $^{43,44}\text{Sc}$  for long-term PET studies further emphasises the demand for no-carrier-added (NCA) Sc radioisotopes in highly pure form. A separation procedure of NCA Sc from bulk Al by using conventional liquid-liquid extraction (LLX) technique using di-2-(ethylhexyl)phosphoric acid (HDEHP) has been reported.

## 2 Experimental

### 2.1 Reagents

The reagents cyclohexane (for HPLC), and suprapur HCl (30%) were purchased from MERCK. Diethylenetriaminepentaacetic acid (DTPA) and di-2-(ethylhexyl)phosphoric acid (HDEHP) were procured from LobaChemie and ICN Pharmaceuticals, respectively. Aluminium standard of 1000 ppm was procured from the National Institute of Standards and Technology (NIST). De-ionized water (18.2 M $\Omega$ cm) was used throughout the experiment.

### 2.2 Details of Irradiation

Eight Al targets were irradiated in respective stacks with a 170 MeV  $^{20}\text{Ne}$  beam incident energy at the Variable Energy Cyclotron Centre, India. The  $^{20}\text{Ne}$  beam was produced from the Electron Cyclotron Resonance (ECR) ion source. Projectile energy exiting from the target was calculated with the help of the SRIM 2013 code [10]. Table 1 lists the details of irradiation, such as the incident and exit energies, irradiation time, current, etc. The incident beam was collimated on the target holder, the total charge being measured via a Faraday cup in conjunction with a current integrator (Danfysik).

**Table 1. Irradiation details of  $^{27}\text{Al} + ^{20}\text{Ne}$  beam**

| Target | Target thickness (mg/cm <sup>2</sup> ) | Incident energy (MeV) | Exit energy (MeV) | Energy at centre of mass (MeV) | Irradiation time (h) | Integrated Charge (μC) |
|--------|--|-----------------------|-------------------|--------------------------------|----------------------|------------------------|
| Al-1   | 2.0                                    | 170                   | 162.4             | 166                            | 3.95                 | 846                    |
| Al-2   | 7.6                                    | 160                   | 130               | 145                            | 11.60                | 964                    |
| Al-3   | 7.6                                    | 145.5                 | 113.4             | 129                            | 14.75                | 589                    |
| Al-4   | 2.0                                    | 119.8                 | 110.2             | 115                            | 3.75                 | 846                    |
| Al-5   | 1.5                                    | 102.6                 | 94.7              | 99                             | 3.75                 | 846                    |
| Al-6   | 1.5                                    | 84.5                  | 75.8              | 80                             | 11.60                | 964                    |
| Al-7   | 3.5                                    | 75.8                  | 54.2              | 65                             | 14.75                | 589                    |
| Al-8   | 1.5                                    | 54.2                  | 43.4              | 49                             | 14.75                | 589                    |

### 2.3 Offline gamma spectrometry

A series of off-line  $\gamma$ - spectra was taken for 7 days in a p-type HPGe detector (30%) of resolution 2.06 keV at 1.33 MeV in combination with a digital spectrum analyser (DSA 1000, CANBERRA) and Genie 2K software (CANBERRA). The radionuclides were identified by monitoring the decay of the radioisotopes at their signature photopeaks. The detector was calibrated using  $^{152}\text{Eu}$  ( $T_{1/2} = 13.51$  a),  $^{133}\text{Ba}$  ( $T_{1/2} = 10.51$  a),  $^{60}\text{Co}$  ( $T_{1/2} = 5.2$  a) and  $^{137}\text{Cs}$  ( $T_{1/2} = 30.07$  a) sources of known activity.

### 2.4 Calculation of cross section

Post irradiation, the Al targets were cooled for almost an hour, and then removed from the cyclotron facility. The target was placed in a well-shielded fume hood and mounted on a glass plate. Radioisotope cross-sections were measured as a function of  $^{20}\text{Ne}$  energy by using the following activation equation (1):

$$A = n\sigma(E)I(1 - e^{-\lambda T}) \quad (1) [11]$$

where A= Activity (Bq) of a particular radionuclide at EOB

$\sigma$  (E)= cross section of production of the radionuclide at incident energy E

I= Intensity of neon beam in particles/s

n= no.of atoms/cm<sup>2</sup>

$\lambda$ = disintegration constant

T= Duration of irradiation

The total uncertainty arising from efficiency calibration was minimum (~0.2-0.6%). The errors due to counting statistics for the respective radioisotopes are mentioned as follows:  $^{43}\text{Sc}$  (0.6-12%),  $^{44\text{m}}\text{Sc}$  (1.01-20.8%),  $^{24}\text{Na}$  (0.93-4.2%),  $^{38}\text{Cl}$  (5.11-12.6%),  $^{34\text{m}}\text{Cl}$  (0.38-13.26%),  $^{42}\text{K}$  (3.1-14.12%) and  $^{41}\text{Ar}$  (8.25-18.04%).

Target thickness accounted for an error of about 5%. The uncertainties from the beam current, incident beam energy, and related irradiation parameters combinedly contributed to an uncertainty of ~10%. Therefore, the total error was 11.5 to 14.3%.

### 2.5 Radiochemical treatment and ICP-OES measurement

The irradiated foils were unwrapped using tweezers and transferred to a beaker. Thereafter, the foils were carefully dissolved by dropwise addition of 0.1 M HCl under mild heating for 2-3 minutes and continuous stirring. Subsequently, the solution was evaporated to drying

consecutively for 4-5 times, and finally, the radioactive stock was prepared in 10 mL of deionised water.

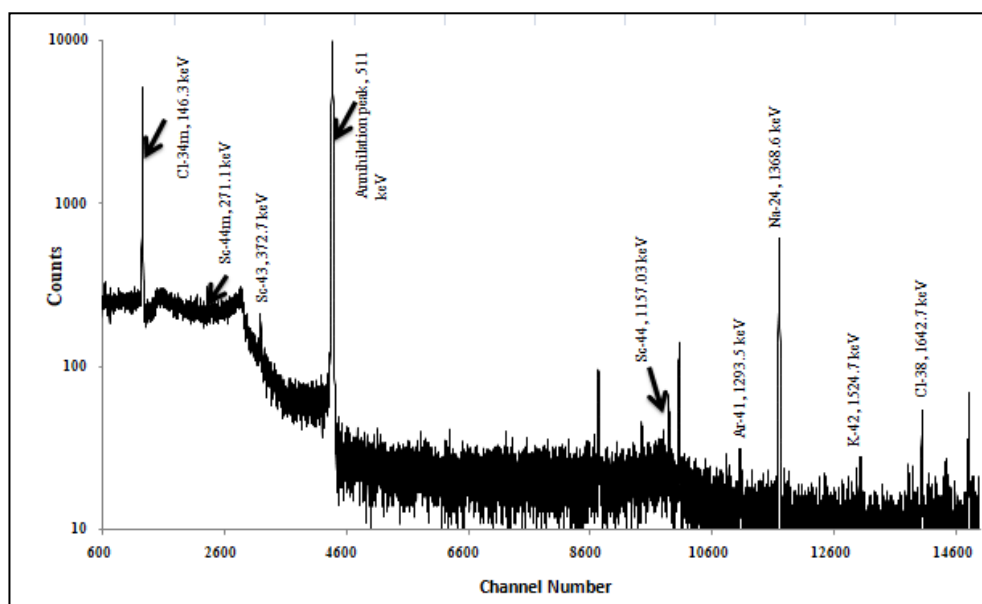
Due to the absence of any Al tracer to monitor bulk Al, it was monitored by Inductively Coupled Plasma Optical Emission Spectroscopy (ICP-OES) (Model: ICAP duo 6500, Thermo-Scientific). The instrumental parameters set for this experiment were: 1150-watt radiofrequency power, 75 rpm pump rate, nebuliser gas flow and auxiliary gas flow rate at 0.6 L/min and 1L/min, respectively. NIST standards were used to pre-calibrate the system.

## 2.6 Radiochemical separation of scandium radioisotopes

Varying concentrations of HDEHP were prepared by addition of cyclohexane in required amounts. 0.5 mL of radioactive stock was added to respective systems containing 3.0 mL HDEHP and 2.5 mL of different HCl concentrations. Each of the systems were shaken well and allowed to settled for about 10 minutes. Subsequent to this, 1.5 mL of the aqueous phase was carefully pipetted out and counted for 2000 s on the HPGe detector. During the measurements, the dead time of the detector was <5%. 1.0 mL of this aqueous phase was then diluted to 4.5 mL for Al detection by ICP-OES. The reference was prepared similarly by the addition of 0.5 mL stock solution to 2.5 mL  $10^{-4}$  M  $\text{HNO}_3$ , and similarly, 1.5 mL was pipetted out and counted for 2000 s.

## 3 Results and Discussions

Production of  $^{43,44,44\text{m}}\text{Sc}$ ,  $^{34\text{m},38}\text{Cl}$ ,  $^{24}\text{Na}$ ,  $^{41}\text{Ar}$ ,  $^{42}\text{K}$  and  $^{43}\text{K}$  radioisotopes was identified upon analysis of the gamma spectrum (Figure 1). Nuclear details of the produced radioisotopes and their corresponding yield are mentioned in Tables 2 and 3, respectively.



**Fig.1.** Gamma spectrum of Al foil irradiated at 160 MeV taken 1h after EOB

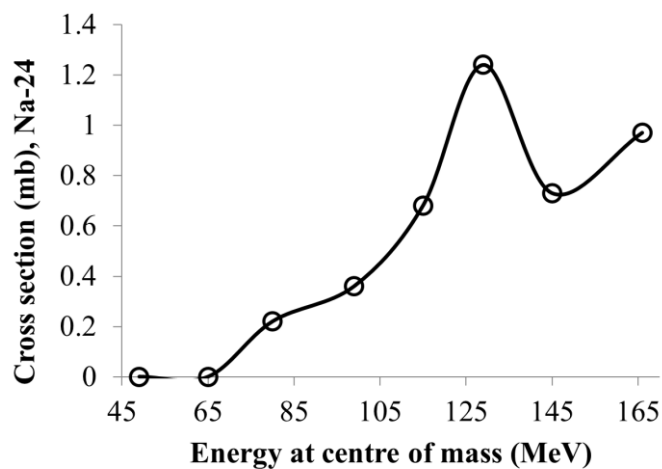
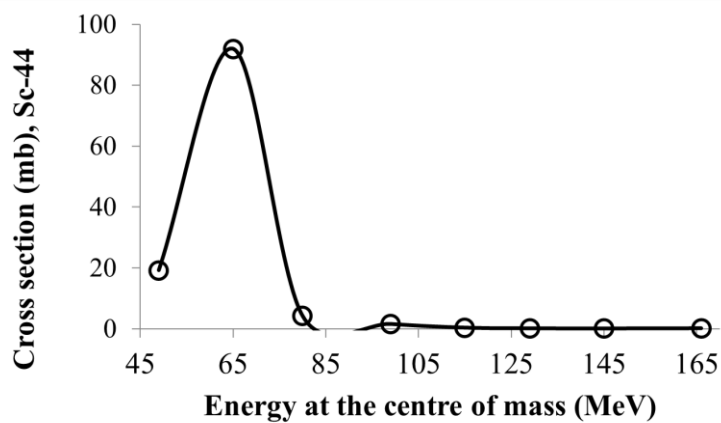
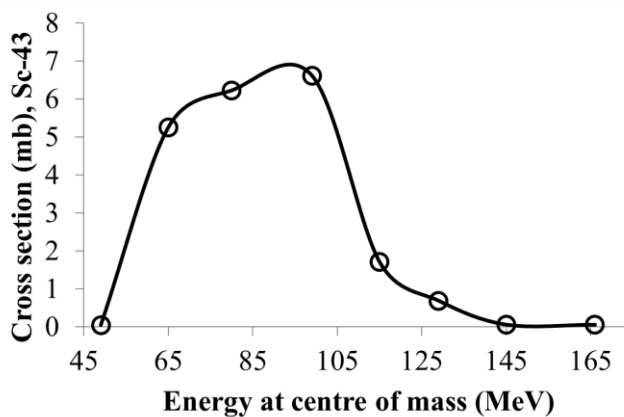
**Table 2. Nuclear data of the produced radioisotopes**  
 (<https://www.nndc.bnl.gov/chart/chartNuc.jsp>)

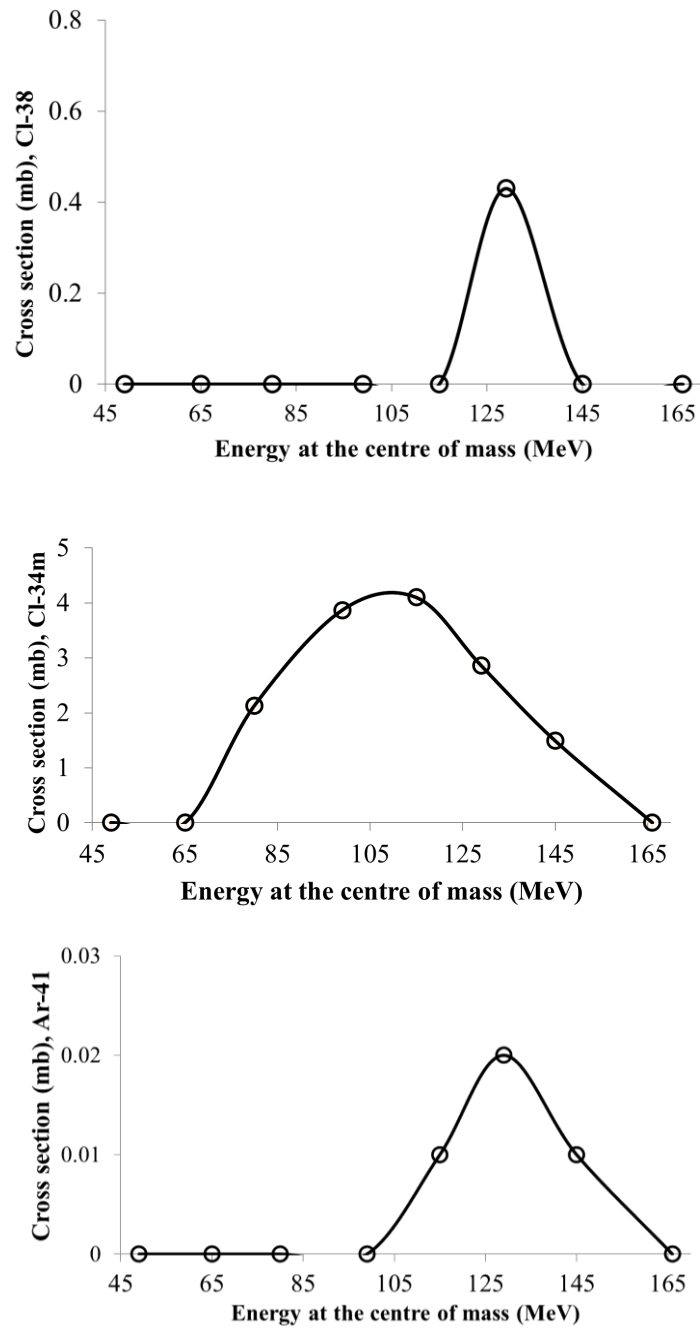
| Radioisotope      | Half-life (T <sub>1/2</sub> ) | Decay mode (%)                       | Principal Gamma energies keV (I,%) | E <sub>Threshold</sub> (MeV) | Probable production route                 |
|-------------------|-------------------------------|--------------------------------------|------------------------------------|------------------------------|---|
| <sup>43</sup> Sc  | 3.89 h                        | ε+β <sup>+</sup> (100)               | 372.7 (23)                         | 32.6                         | <sup>27</sup> Al( <sup>20</sup> Ne,α)     |
| <sup>44</sup> Sc  | 3.92 h                        | ε+β <sup>+</sup> (100)               | 1157.03(99.9)                      | 15.7                         | <sup>27</sup> Al( <sup>20</sup> Ne,2pn)   |
| <sup>44m</sup> Sc | 58.6 h                        | IT (98.8)<br>ε+β <sup>+</sup> (1.2)  | 271.1 (86.7)<br>1157.03 (1.2)      |                              | <sup>27</sup> Al( <sup>20</sup> Ne,2pn)   |
| <sup>42</sup> K   | 12.3 h                        | β <sup>-</sup> (100)                 | 1524.7 (18)                        | 46.0                         | <sup>27</sup> Al( <sup>20</sup> Ne,α2p)   |
| <sup>41</sup> Ar  | 109.3 m                       | β <sup>+</sup> (100)                 | 1293.5 (99.1)                      | 62.1                         | <sup>27</sup> Al( <sup>20</sup> Ne,α3p)   |
| <sup>34m</sup> Cl | 32.0 m                        | IT (44.6)<br>ε+β <sup>+</sup> (55.4) | 146.3 (40.5)<br>1176.6(14.1)       | -                            | <sup>27</sup> Al( <sup>20</sup> Ne,α3n)   |
| <sup>38</sup> Cl  | 37.24 m                       | β <sup>-</sup> (100)                 | 1642.7 (32)                        |                              | <sup>27</sup> Al( <sup>20</sup> Ne,α4pn)  |
| <sup>24</sup> Na  | 14.95 h                       | β <sup>-</sup> (100)                 | 1368.6 (100)                       |                              | <sup>27</sup> Al( <sup>20</sup> Ne,5α2pn) |

**Table 3. Yield of the produced isotopes (in Bq) at the EOB under the experimental conditions**

| Energy (MeV) | <sup>43</sup> Sc | <sup>44m</sup> Sc | <sup>24</sup> Na | <sup>34m</sup> Cl | <sup>38</sup> Cl | <sup>42</sup> K | <sup>41</sup> Ar |
|--------------|------------------|-------------------|------------------|-------------------|------------------|-----------------|------------------|
| 166          | 65.8             | 16.5              | 385              | 0                 | 0                | 0               | 0                |
| 145          | 168.2            | 37.3              | 1066             | 5183              | 0                | 127             | 42.4             |
| 129          | 1680             | 35.0              | 986              | 4807              | 2089             | 314             | 86               |
| 115          | 2079             | 37.4              | 272              | 10143             | 0                | 319             | 23               |
| 99           | 6040             | 127.8             | 106.4            | 7191              | 0                | 182             | 0                |
| 80           | 3748             | 370.3             | 63               | 1469              | 0                | 65              | 0                |
| 65           | 3777             | 11373             | 0                | 0                 | 0                | 0               | 0                |
| 49           | 14.3             | 1020              | 0                | 0                 | 0                | 0               | 0                |

The production of <sup>44</sup>Sc radioisotope may be from <sup>27</sup>Al(<sup>20</sup>Ne,2pn) reaction, or as the daughter product of <sup>44m</sup>Sc (IT 98.61%). Therefore, it was possible to measure the yield and production cross section of <sup>44m</sup>Sc only. The excitation function plots are shown in Figure 2.





**Fig 2.** Excitation Function of  $^{27}\text{Al}+^{20}\text{Ne}$  reaction

The radiochemical separation was done with NCA  $^{43,44}\text{mSc}$  and  $^{24}\text{Na}$  in the matrix, as the other radioisotopes were short-lived and had died down. The cross-section of the other radioisotopes were much lower than that of scandium. The separation of NCA Na and Sc

from bulk Al was monitored. LLX experiments using cation exchanger HDEHP (0.1%) with different concentrations of HCl revealed the extraction of Sc selectively into the organic phase throughout the entire range of acid concentration (Figure 3), leaving Na and Al in the aqueous phase. To get the optimum separation condition, the acid concentration was fixed at 0.1 M, and the strength of HDEHP was varied upto 10% (Figure 4), to observe that the extraction of Al increased with increasing strength of HDEHP, while at this pH condition, Sc and Na showed 100% extraction and non-extraction, respectively. The best separation condition has been mentioned in Table 4.

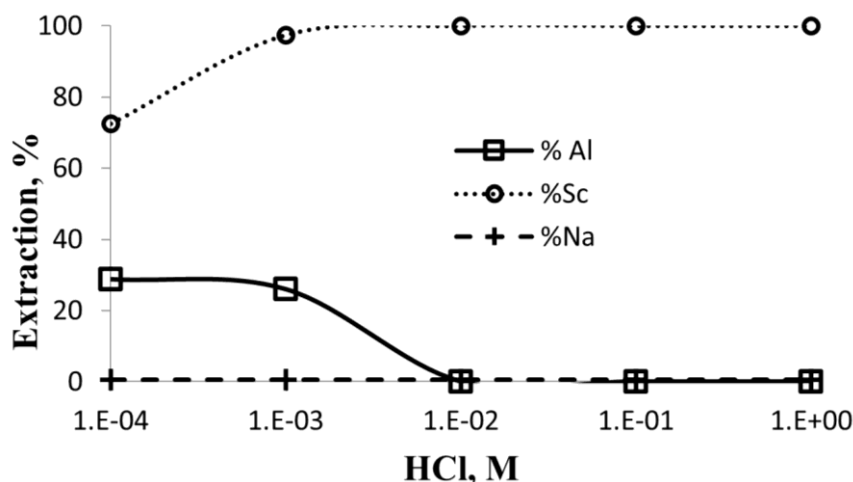


Fig.3. Extraction profile of NCA Sc, NCA Na and bulk Al under varying HCl concentration

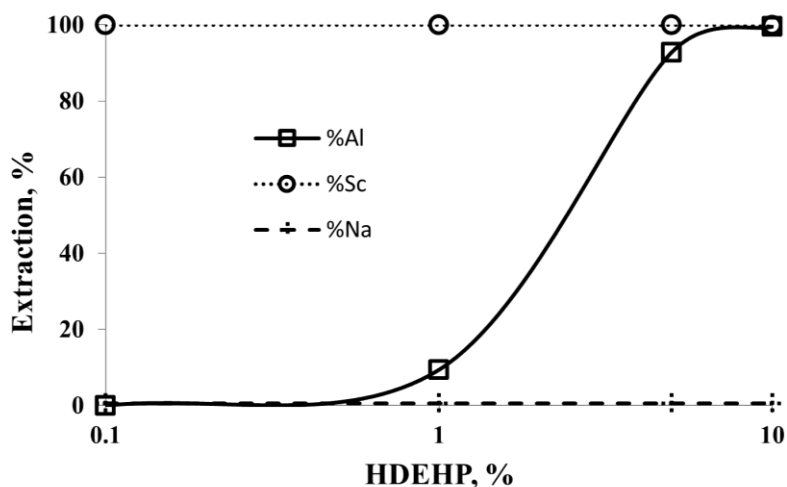


Fig.4. Extraction profile of NCA Sc, NCA Na and bulk Al under varying HDEHP strength

The high extraction of Sc by HDEHP was earlier mentioned by Wang et al [12]. In less acidic conditions, the  $Sc(DEHP)_3$  complex is formed via a cation exchange mechanism through proton transfer mechanism, while in high acidic conditions, Sc gets extracted through a

solvating mechanism. Sc(III) is nominally stable in pure solution below pH 4, but hydrolyses to  $\text{ScOH}_2^+$  at pH 4–7 [13], while Na, existing as  $\text{Na}^+$  did not get extracted under any pH conditions. Extraction of Al by cation exchanger may be due to the existence of Al as  $\text{Al}^{3+}$  and  $\text{Al}_2(\text{OH})_2^{4+}$ . But 0% extraction of Al was found at 1M HCl at all strengths of HDEHP. This may be explained by the fact that with a decrease in pH, Al solely existed as Al(III) and not in different hydroxide forms. Sc was back extracted from the organic phase by the use of alkaline 0.1 M DTPA by 100 % in a single extraction. The LLX experiment therefore, provided a separation route for the NCA Sc radioisotopes from both bulk Al and co-produced NCA  $^{24}\text{Na}$ .

**Table 4. Distribution coefficient and Separation factors at optimum condition**

| Experimental conditions | $D_{\text{Al}}$ | $D_{\text{Sc}}$ | $S_{\text{Sc/Al}}$ |
|-------------------------|-----------------|-----------------|--------------------|
| 0.1% HDEHP+1 M HCl      | 0.2             | 2541            | 12705              |

D= Distribution coefficient =Counts in aqueous (or liquid) phase/counts in organic (or solid) phase

S= Separation Factor, i.e.,  $D_{\text{Sc}}/D_{\text{Al}}$

## Conclusion

An attempt has been made to study the possible radioisotopes produced from the most commonly used degrader foil. Radioisotopes like  $^{43,44}\text{Sc}$ ,  $^{24}\text{Na}$ ,  $^{34\text{m}}\text{Cl}$ , etc., were identified by gamma spectrometry. Excitation function of  $^{27}\text{Al}+^{20}\text{Ne}$  reaction from 49-166 MeV was reported. Alongside, clinically important NCA  $^{43,44}\text{Sc}$  radioisotopes were separated from bulk Al by 100% using a conventional liquid-liquid extraction technique.

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