# **Production and separation of 97Ru and coproduced 95Tc from 12C-induced reaction on yttrium target**

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**Summary.** An attempt has been made to produce no-carrieradded (nca)  $97$ Ru for the first time from the  $12$ C-induced reaction on yttrium target. A natY foil of 10 mg/cm<sup>2</sup> thickness was irradiated by  $75 \text{ MeV}$  <sup>12</sup>C beams for  $3.5 \text{ h}$  and a thick target yield of  $514 \text{ kBq}/\mu\text{A}$  h for <sup>97</sup>Ru and 338 kBq/ $\mu\text{A}$  h for  $95Tc$  was achieved. The nca  $97Ru$  and coproduced nca  $95Tc$ were then radiochemically separated from the bulk target by liquid–liquid extraction (LLX). Liquid cation exchanger di-(2 ethylhexyl)phosphoric acid (HDEHP), liquid anion exchanger trioctylamine (TOA) were used in this study. The technique offers efficient chemical separation of the products, resulting in very high separation factors.

# **1. Introduction**

Owing to rich coordination chemistry radioactive rutheniumlabelled complexes have found different biomedical applications. In the past, complexes were labelled mostly with neutron rich  $^{103}$ Ru (39.247 d), which is produced in a reactor by neutron activation on the natural  $102$ Ru (abundance 31.6%). However due to comparatively long half-life, low specific activity, high energy  $\gamma$ -rays: 497.085 keV (91.0%) and 610.333 keV (5.76%) and  $\beta$ <sup>-</sup>-emissions, it is not ideal for *in-vivo* applications. Alternatively, proton rich isotopes of ruthenium, <sup>94</sup>Ru (51.8 min), <sup>95</sup>Ru (1.643 h), <sup>97</sup>Ru (2.83 d), having suitable decay properties appeared promising. Moreover they can be produced in the no-carrier-added (nca) state.

Proton rich <sup>97</sup>Ru has electron capture decay mode and it emits only two intense low energy  $\gamma$ -rays: 215.70 keV (85.62%) and 324.49 keV (10.79%), hence found potential among others for labelling compounds for delayed studies in diagnosis as well as therapy [\[1](#page-5-0)[–6\]](#page-5-1). Applications demonstrate <sup>97</sup>Ru-DTPA (diethylenetriamine pentaacetic acid) as cerebro-spinal-fluid imaging agent, a superior analogue of  $^{111}$ In-DTPA [\[2\]](#page-5-2);  $^{97}$ Ru-PIPIDA (N- $\alpha$ - (p-isopropylacetanilide) iminoacetic acid) as hepatobiliary diagnostic agent, a good alternative of  $99m$ Tc-PIPIDA [\[3\]](#page-5-3); 97Ru-transferrin as an efficient agent for tumour and/or ab-scess localization [\[4\]](#page-5-4); <sup>97</sup>Ru-bleomycin acts as a chemotherapeutic agent [\[5\]](#page-5-5), *etc.* In fact, 97Ru-labelled compounds offer better results in some cases than that labelled with  $\frac{99 \text{m}}{2}$ C,  $111$ In or  ${}^{67}$ Ga. Moreover, a variety of radiopharmaceuticals can be prepared with  $97$ Ru comparable to  $111$ In,  $99$ mTc and  ${}^{67}Ga$  [\[1\]](#page-5-0). Therefore investigation is required to produce high purity 97Ru for its successful applications.

There have been several attempts to produce <sup>97</sup>Ru. Apart from neutron activation product,  $97$ Ru have been produced in different light ion  $(p, \alpha \text{ and }^3\text{He})$  induced reactions on natural and enriched targets [\[6–](#page-5-1)[17\]](#page-5-6). Effort has also been made to produce it even by  ${}^{99}Te(p, 3n)^{97}Ru$  reaction using a metallic target of long lived <sup>99</sup>Tc (2.111  $\times$  10<sup>5</sup> yr) [\[9–](#page-5-7)[11\]](#page-5-8). In spite of some attractive criterion, such as high yield (∼ 1 mCi/μA h) of  $97$ Ru can be achieved in the  $99$ Tc(p, 3n)<sup>97</sup>Ru reaction, this particular route is not appropriate for regular production as the target is radioactive, not easily available and expensive.

In late seventies a group of scientists from Brookhaven National Laboratory produced nca <sup>97</sup>Ru through high energy (200 MeV) proton induced spallation reaction on high purity Rh foil. According to their claim more than 100 mCi activity of  $97$ Ru can be produced in this method per day [\[6\]](#page-5-1). It is expected that several radionuclides other than  $97Ru$ will be produced under the experimental condition about which the authors remained silent. However, utility of this technique is limited due to the scarcity of high energy proton beam. Later in early eighties another group from Crocker Nuclear Laboratory made an attempt to produce  $97$ Ru using lower energy protons (67.5 MeV) on Rh foil. The yield of  $97$ Ru was measured as 1.36 mCi/ $\mu$ A h at the end of bombardment (EOB). In addition to <sup>97</sup>Ru, production of several coproduced radionuclides, <sup>95</sup>,95m,96Tc, <sup>99</sup>,100,101Pd, 97,97m,98,99,99m,100,101,101m,102Rh, was confirmed by γ-ray spectrometry [\[7\]](#page-5-9).

Recently we proposed a new route to produce high purity 97Ru by impinging 32 MeV 7Li projectiles on a Nb foil *via* <sup>93</sup>Nb(<sup>7</sup>Li, 3*n*)<sup>97</sup>Ru reaction where ∼ 1 MBq/μA h yield of 97Ru was achieved. Minute activity of <sup>95</sup>,96Tc produced in the target matrix was chemically separated from  $97$ Ru [\[18\]](#page-5-10).

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Several techniques have been developed for separation of ruthenium  $[19]$ . Separation of  $\frac{97}{2}$ Ru has been achieved from the proton irradiated Rh or Tc metal target by distillation of  $97$ RuO<sub>4</sub> from concentrated H<sub>2</sub>SO<sub>4</sub> medium [\[6](#page-5-1)[–8,](#page-5-12) [10\]](#page-5-13). The chemical yield of  $97$ Ru was found to vary between 95 and 98%. Distillation method was found efficient for separation of 97Ru from Pd and Rh, but not for Tc as it is also distilled as technetium oxides,  $TcO_4$ ,  $Tc_2O_7$ , *etc.* with  $RuO_4$ . The complete chemical procedure also takes long time (> 5 h).

The separation of nca  $97$ Ru has also been achieved from the  $\alpha$ -particle or <sup>3</sup>He-particle irradiated natural Mo target by solvent extraction, ion exchange and precipitation technique [\[13–](#page-5-14)[16\]](#page-5-15). These techniques are relatively simple and faster compared to the distillation method. The precipitation technique selectively separated <sup>97</sup>Ru from bulk Tc or Mo metals when coprecipitated with Fe-oxides (Fe<sub>2</sub>O<sub>3</sub>, Fe(OH)<sub>3</sub> and  $Fe(OH)_{2}$ ) which does not coprecipitate Tc or Mo [\[14\]](#page-5-16). The ion exchange technique was also applied successfully to separate nca <sup>97</sup>Ru from the molybdenum target. Target was dissolved in  $H_2O_2$  and kept into 1 M HNO<sub>3</sub>. Active solution was passed through the tin dioxide column followed by an anion exchange column of Bio-Rad AG1-X8 in the nitrate form. The  $SnO<sub>2</sub>$  column adsorbs bulk Mo and Tc quantitatively while retention of <sup>97</sup>Ru is less than 5%. Further purification occurs in anion exchange column. The method leads to pure  $97$ Ru with  $> 95\%$  efficiency [\[16\]](#page-5-15).

Ion exchange phenomenon has also been exploited by Lahiri *et al.*  $[20, 21]$  $[20, 21]$  $[20, 21]$  to separate nca  $97$ Ru from coproduced Tc, Nb and bulk Mo in liquid–liquid extraction (LLX) using liquid anion exchanger trioctylamine (TOA) or liquid cation exchanger di-(2-ethylhexyl)phosphoric acid (HDEHP) along with neutral liquid, tri-butyl phosphate (TBP) dissolved in cyclohexane as organic phase in com-bination with HCl [\[20,](#page-5-17) [21\]](#page-5-18). We also reported separation of nca 97Ru from bulk Nb and coproduced Tc by both LLX using HDEHP dissolved in cyclohexane as organic phase and SLX using cation exchanger resin DOWEX-50 as solid phase along with aqueous HCl. The LLX appeared slightly better than SLX in terms of radiochemical yield and purity of  $97$ Ru [\[18\]](#page-5-10).

As a part of our continuous endeavour, this paper reports a new production route of  $97$ Ru. For the first time, it describes production of nca  $97$ Ru from  $12$ C-induced reaction on yttrium target and its subsequent radiochemical separation from the bulk target and coproduced nca 95Tc using LLX.

## **2. Theoretical study**

In order to investigate the feasibility of production of  $97Ru$ . we have estimated excitation functions of the evaporation residues theoretically from  $^{12}C + ^{89}Y$  reaction using the statistical model code PACE4 [\[22\]](#page-5-19) in the 35–85 MeV incident projectile energy range. Fig. [1](#page-1-0) shows the excitation functions of the residues: <sup>99</sup>,98,97,96Rh, 97Ru, <sup>93</sup>,94,95,96Tc and 93mMo. The cross section of 97Ru is rather low; the maximum cross section was found to be 300 mb at 65 MeV projectile energy. The cross section of  $97Rh$ , which decays to  $97Ru$ , peaked at the same energy with a maximum of ∼ 400 mb. It is clear from the figure that there is adequate possibility of production of other radionuclides such as <sup>98</sup>Rh, <sup>97</sup>Rh, <sup>93</sup>Tc, <sup>94</sup>Tc,



<span id="page-1-0"></span>**Fig. 1.** Theoretical excitation functions of the residues produced in  ${}^{12}C + {}^{89}Y$  reaction as estimated from PACE4.

 $95$ Tc,  $93$ mMo and  $95$ Ru, apart from  $97$ Ru, if  $50-75$  MeV incident projectile energy is considered. However, only <sup>95</sup>Tc activity is expected to be present along with  $97$ Ru after a day as all other short-lived radionuclides will decay by the time. Hence,  ${}^{12}$ C-induced reaction on  ${}^{89}Y$  with appropriate projectile energy followed by efficient chemistry can produce high purity 97Ru. Considering all facts and with an aim to reduce the production of <sup>95</sup>Tc, 75 MeV projectile energy was chosen for this experiment.

# **3. Experimental**

# **3.1 Production of 97Ru and 95Tc**

A pure natural yttrium  $(^{89}Y)$  foil (99.9% purity) was procured from Alfa Aesar, a Johnson Matthey Company, and was rolled properly to produce a self supporting foil of 10 mg/cm2 thickness. The yttrium target was bombarded by 75 MeV  $^{12}C^{6+}$  ions for 3.5 h up to a total dose of 1 mC at the BARC-TIFR Pelletron accelerator facility, Mumbai, India. The exit energy of the projectile was 62 MeV as estimated using the Stopping and Range of Ions in Matter (SRIM) [\[23\]](#page-5-20). The total dose of irradiation was measured by an electron suppressed Faraday cup stationed at the back of the target. The recoiled products, if any, in the beam direction were stopped completely in an aluminum catcher foil.

**Table 1.** Nuclear decay data [\[24\]](#page-5-21) of the radionuclides produced in <sup>12</sup>C + <sup>89</sup>Y reaction. The *γ*-ray energies marked in bold were used in this study.

<span id="page-1-1"></span>

Product	$T_{1/2}$	$E_v$ keV $(I_v \% )$
$98$ Rh	$8.72 \,\mathrm{min}$	652.6 $(97.0)$ , 1164.3 $(4.9)$
$97$ Rh	$30.7 \text{ min}$	421.5(74.6), 840.1(12.0), 878.8(9.0)
$\rm^{97m}Rh$	$46.2 \text{ min}$	<b>189.2</b> (48.5), 421.5 (12.7)
$96$ Rh	$9.90 \,\mathrm{min}$	832.5 (100), 631.7 (74.5), 685.5 (95.7),
		741.9 (29.4)
$97$ Ru	2.83d	215.7(85.6), 324.5(10.8)
$95$ Ru	1.64h	336.4 (69.9), 1096.8 (20.9), 626.8 (17.8)
$^{93}$ Tc	2.75h	1362.9 (66.2), 1520.3 (24.4)
94Tc	$293 \,\mathrm{min}$	702.7 (99.6), 849.7 (95.7), 871.0 (99.9)
$^{94m}$ Tc	$52.0 \,\mathrm{min}$	871.0 (94.2)
95Tc	20.0 <sub>h</sub>	765.8 (93.8)
96Tc	4.28 d	778.2 (99.7), <b>812.5 (82)</b> , 849.9 (98)
$93m$ Mo	6.85h	263.0 (57.4), 684.7 (99.9)

Yttrium radioisotopes required in monitoring the fate of bulk matrix during radiochemical separation of nca <sup>97</sup>Ru and <sup>95</sup>Tc are barely produced in the above reaction. Therefore, a separate natural yttrium foil of 3 mg/cm2 thickness was irradiated by 22 MeV protons for 2 h at 600 nA beam current to produce  ${}^{88}Y$  (106.6 d) in the yttrium matrix.

After irradiation, the activated yttrium foils were assayed off-line to measure the  $\gamma$ -ray activity of the product radioisotopes using an HPGe detector having a resolution of 2.13 keV at 1332 keV coupled with a PC based MCA, PCA2 (OXFORD). The detector was calibrated using standard sources like, <sup>152</sup>Eu ( $T_{1/2}$  = 13.537 yr), <sup>133</sup>Ba (10.54 yr) and  $137Cs$  (30.0 yr) of known activities and the efficiency was obtained as a function of  $\gamma$ -ray energy. The probable nuclear reactions and nuclear spectroscopic data of the radionuclides produced in the interaction of  ${}^{12}C + {}^{89}Y$  are listed in Table [1](#page-1-1) [\[24\]](#page-5-21).

#### **3.2 Radiochemical separation**

The proton and 12C activated yttrium foils were dissolved separately in minimum volume of 0.1 M HCl. Proton irradiated yttrium containing the radioisotope <sup>88</sup>Y was separated from the nca  $88,89$ Zr, produced by proton activation on yttrium foil, by LLX as reported in our earlier work [\[25,](#page-5-22) [26\]](#page-5-23). Yttrium was extracted quantitatively along with <sup>∼</sup> 20% nca <sup>88</sup>,89Zr into 10% HDEHP dissolved in cyclohexane, leaving  $\sim$  80% nca Zr in 5 × 10<sup>-4</sup> M H<sub>2</sub>SO<sub>4</sub> which was later back extracted from the organic phase by  $9 M H_2SO_4$ . Yttrium solution containing the <sup>88</sup>Y tracer was then evaporated to dryness and kept in 0.1 M HCl.

The 12C activated yttrium solution was spiked with the previously prepared 88Y tracer and evaporated to dryness to attain the same chemical form for both the <sup>88</sup>Y tracer and the bulk yttrium. The residue was then redissolved into 0.01 M HCl to prepare a stock solution that contains ruthenium ( $97$ Ru), technetium ( $95$ Tc) and yttrium. Radiochemical separation was explored by LLX using this active stock.

The LLX was carried out using the liquid cation exchanger, HDEHP, and anion exchanger, TOA dissolved in cyclohexane as the organic phase and HCl as the aqueous phase. Both of these liquid ion exchangers have high selectivity for different metal ions; therefore they were chosen as the extracting agents [\[27\]](#page-5-24). In each extraction 3 mL of organic solution was shaken for 10 min with an equal volume of aqueous HCl and 200 μL active stock solution and was left for 10 min for the complete phase separation. It is worthy to mention that TOA or HDEHP establish rapid equilibrium with aqueous acidic phase in LLX system. Therefore, 10 min time was sufficient to establish the equilibrium. In order to estimate the distribution of activities, a 2 mL aliquot collected from each phase was subjected to γ-ray spectrometric analysis. In each LLX system (TOA-HCl and HDEHP-HCl), the optimum extraction condition was achieved from two series of extractions (i) varying concentration of aqueous phase with respect to a fixed concentration of organic phase (ii) varying concentration of organic phase with a fixed concentration of aqueous phase. All the chemicals used for the separation study were of analytical grade.



<span id="page-2-0"></span>**Fig. 2.** The *γ*-ray spectrum of the <sup>12</sup>C-irradiated yttrium foil after 1 h of EOB.

### **4. Results and discussion**

The  $\gamma$ -ray spectra of <sup>12</sup>C irradiated natural yttrium target collected after EOB were analyzed to assure the production of nca 97Ru and the coproduced radionuclides. Fig. [2](#page-2-0) represents the  $\gamma$ -ray spectrum of 75 MeV <sup>12</sup>C irradiated yttrium target after  $45$  min of EOB. The nca  $97$ Ru was produced in the target matrix along with nca  $^{98}$ Rh (8.72 min),  $^{97}$ Rh (30.7 min), 97mRh (46.2 min), 96Rh (9.9 min), 93Tc (2.75 h), <sup>94</sup>Tc (293 min), <sup>94m</sup>Tc (52.0 min), <sup>95</sup>Tc (20 h), <sup>96</sup>Tc (4.28 d), and  $93m$ Mo (6.85 h). The batch yields of the residual prod-ucts have been tabulated in Table [2.](#page-2-1) Interaction of  $^{12}$ C with 89Y opened up a number of reaction channels as expected from the theoretical study. Compound nuclear reaction is the dominant mechanism in the selected projectile energy range. However, <sup>97</sup>Ru activity will be abundant along with some 95Tc activity after a day as all short-lived residues decayed mostly to stable isotopes.

The maximum yield of <sup>97</sup>Ru was estimated  $\sim$  0.5 MBq/ μA h at EOB which is just half of that measured from <sup>93</sup>Nb(<sup>7</sup>Li, 3*n*)<sup>97</sup>Ru reaction [\[18\]](#page-5-10). The measured yield of <sup>95</sup>Tc is reasonably high,  $\sim$  0.3 MBq/ $\mu$ A h at EOB. However, this target-projectile combination ( ${}^{12}C + {}^{89}Y$ ) shows the possibility of producing high purity nca  $97$ Ru in a thick target reaction, provided a chemical separation is done.

**Table 2.** Batch yields of radionuclides under the experimental conditions.

<span id="page-2-1"></span>

Experimental conditions		Batch yield at EOB		
		Radio- nuclide	Yield $(kBq/\mu A h)$	
Beam	$^{12}$ C <sup>+6</sup> (75 MeV)	$97$ Ru	$514 \pm 42$	
Target	$^{89}Y$ foil (10 mg/cm <sup>2</sup> )	$98$ Rh	$1559 \pm 145$	
Irradiation time	3.5h	$97$ Rh	$3753 + 275$	
Dose	$1000 \mu C$	97mRh	$282 + 25$	
		$96$ Rh	$56 + 5$	
		93Tc	$114 \pm 9$	
		$94$ Tc	$1225 \pm 102$	
		95Tc	$338 + 29$	
		$96T_C$	$0.5 \pm 0.1$	
		$93m$ Mo	$45 + 4$	



<span id="page-3-0"></span>Fig. 3. Extraction profile of nca Ru, Tc and bulk Y in LLX as a function of HCl concentration at a fixed strength of TOA (0.1 M).

Fig. [3](#page-3-0) shows the extraction profile of nca Ru, Tc and bulk Y obtained from LLX as a function of HCl concentration  $(1.0 \times 10^{-4}$  to 1 M) at fixed concentration of TOA (0.1 M). More than 90% nca Tc was extracted into TOA leaving nca Ru and bulk Y in aqueous phase. The extraction pattern of the nca Tc corroborates with our earlier paper [\[28\]](#page-5-25). However, a nominal difference in the extraction pattern of Y was observed. In Ref. [\[28\]](#page-5-25), Y remained in the aqueous phase, but a small amount was extracted in the organic phase, while in the present paper Y quantitatively remained in the aqueous phase. The small difference may be due to difference in bulk Y concentration. For example, in Ref. [\[28\]](#page-5-25), the target thickness was  $4.9 \text{ mg/cm}^2$  whereas in the present case it is 10 mg/cm2 . However, the extraction pattern of Y reported in this article also corroborates with our earlier report [\[29\]](#page-5-26).

No significant effect was observed in this profile due to the variation in HCl concentration. In order to increase the extraction of nca Tc, concentration of TOA was varied from  $1 \times 10^{-4}$  to 0.5 M keeping HCl concentration fixed to pH 4, as shown in Fig. [4.](#page-3-1) Extraction of nca Tc increased with increasing concentration of TOA. The extraction behavior of bulk Y and nca Ru remained unchanged. The optimum separation was achieved at pH 4, where ∼ 98% nca Tc was extracted into 0.5 M TOA leaving Y and Ru quantitatively in the aqueous phase, leading to separation factors (SF) of  $7.6 \times 10^5$  ( $D_{\text{Te}}/D_{\text{Y}}$ ) and  $3 \times 10^6$  ( $D_{\text{Te}}/D_{\text{Ru}}$ ), respectively. Repetition of the extraction for the second time removed entire Tc activity from the aqueous phase. Yttrium mostly forms  $Y^{3+}$  or  $[Y(H_2O)_n]^{3+}$  species [\[30\]](#page-5-27) in low acidity region while Tc forms anionic species, possibly  $[TcCl<sub>5</sub>(H<sub>2</sub>O)]<sup>-</sup>$  or



<span id="page-3-1"></span>**Fig. 4.** Extraction profile of nca Ru, Tc and bulk Y in LLX as a function of TOA concentration at a fixed strength of HCl ( $1 \times 10^{-4}$  M).



<span id="page-3-2"></span>**Fig. 5.** Extraction of nca Ru and bulk Y in LLX as a function of HCl concentration at a fixed strength of HDEHP (1%).

 $[TCCl<sub>6</sub>]^{2-}$  [\[31,](#page-5-28) [32\]](#page-5-29) and is thereby extracted to the organic phase. The nca Ru does not show any anionic chemistry under the experimental condition. Separation of nca Tc and bulk Y agreed well with our previous report [\[28\]](#page-5-25). The nca Tc was stripped back from the organic phase (0.5 M TOA) by 0.1 M DTPA (diethylenetriaminepentaacetic acid) dissolved in 0.1 M NaOH.

After separation of nca Tc, the aqueous solution containing Y and nca Ru was evaporated to dryness and finally kept in minimum volume of 0.01 M HCl. This solution was used as stock solution for separation of nca Ru and bulk Y. The LLX was performed with HDEHP. Extraction behavior of nca Ru and bulk yttrium is presented in Fig. [5](#page-3-2) as a function of HCl concentration ( $1 \times 10^{-4}$  M to 8 M) at a fixed 1% (v/v) HDEHP concentration. Bulk Y was quantitatively extracted into the HDEHP phase in low acidity (pH 4 to 1) leaving nca Ru in the aqueous HCl. However, a sharp fall in the extraction of Y was observed when the HCl concentration was increased beyond 0.1 M. Extraction behavior of nca Ru remained almost unaffected, irrespective of the concentration of HCl throughout the range, though in lower acidity (pH 4–2), about 1% nca Ru was extracted in the organic phase. Best separation was found at 0.1 M HCl, where bulk Y was extracted quantitatively into the HDEHP phase leaving nca Ru quantitatively. Concentration of HDEHP was varied in another set of extraction keeping the concentration of HCl fixed to 0.1 M (Fig. [6\)](#page-3-3). It was observed that bulk Y was extracted quantitatively into the organic phase in all strengths of HDEHP  $(1-10\%)$  and nca Ru remained in the aqueous phase, irrespective of concentration of HDEHP. The



<span id="page-3-3"></span>**Fig. 6.** Extraction of nca Ru and bulk Y as a function of HDEHP concentration at a fixed strength of HCl (0.1 M) in LLX.



**Fig. 7.** Schematic diagram of the production and separation of nca Ru and nca Tc from bulk Y target using LLX.

<span id="page-4-0"></span>**Table 3.** Distribution ratios (*D*) and separation factors (SF) of nca Ru, nca Tc and bulk Y at the optimal experimental conditions.

Experimental condition	D <sub>v</sub>	$D_{\scriptscriptstyle\mathrm{Ru}}$	$D_{\rm Te}$	SF	SF	SF $(D_Y/D_{Ru})$ $(D_{Tc}/D_Y)$ $(D_{Tc}/D_{Ru})$
LLX:						
HCl $(1 \times 10^{-4} \text{ M})/TOA$ $(0.5 \text{ M})$	$1.7 \times 10^{-3}$ 4.3 $\times 10^{-4}$		1300		$7.6 \times 10^{5}$	$3 \times 10^6$
LLX:						
HC1 (0.1 M)/HDEHP (1%)	547	$2.6 \times 10^{-4}$		$2.1 \times 10^{6}$		
HC1 (0.1 M)/HDEHP (5%)	503	$2.7 \times 10^{-4}$		$1.9 \times 10^{6}$		
HC1 (0.1 M)/HDEHP (10%)	566	$2.6 \times 10^{-4}$		$2.2 \times 10^{6}$		

<span id="page-4-1"></span>formation of cationic species of Y is therefore confirmed in low acidity region. Under the experimental condition, no extraction of nca Ru was observed eithor in anion exchanger TOA or in cation exchanger HDEHP. The published literature illustrates that Ru forms complex mixture of Ru(III) and Ru(IV) in HCl solution and Ru forms anionic species in high HCl concentration  $(> 1 M)$  [\[33](#page-5-30)[–35\]](#page-6-0). This is in agreement with the present observation as we have not observed extraction of nca Ru into TOA in lower acidity ( $1.0 \times 10^{-4}$  to 1 M, Fig. [3\)](#page-3-0). On the other hand, nca Ru is not extracted into the cation exchanger HDEHP (Fig. [5\)](#page-3-2) which confirms that nca Ru does not form probably any cationic species in the HCl medium ( $1.0 \times 10^{-4}$  to 8 M).

This led to three optimum conditions carrying high separation factor (SF) for the quantitative separation of nca Ru and bulk Y (i) 0.1 M HCl and 1% HDEHP (SF =  $2.1 \times 10^6$ ) (ii) 0.1 M HCl and 5% HDEHP (SF =  $1.9 \times 10^6$ ) (iii) 0.1 M HCl and 10% HDEHP (SF =  $2.2 \times 10^6$ ). A schematic diagram of the production and separation of nca Ru from bulk Y target is presented in Fig. [7.](#page-4-0) Distribution ratios and separation factors obtained under the optimal experimental condition are presented in Table [3.](#page-4-1) The chemical purification procedure requires only 1 h, which is essentially short compared to other methods. However, at least ∼ 20 h cooling time is required after EOB in order to allow the decay of short lived products.

### **5. Conclusion**

This article reports a new route for the production of high purity 97Ru from 12C-induced reaction on yttrium target. Interaction of the projectile with the target produced several residues, which are mostly short-lived, and it is interesting to note that only <sup>97</sup>Ru and <sup>95</sup>Tc are abundant after a day. Production of 95Tc was expected to be low from the theoretical study at the experimental condition, but thick target yield of <sup>95</sup>Tc was found comparatively high in the experiment. Measured thick target yield of  $97$ Ru is 514 kBq/ $\mu$ A h and that of <sup>95</sup>Tc it is 338 kBq/ $\mu$ A h. The product yield of <sup>97</sup>Ru in the

present reaction is low compared to others  $(p, \alpha)$ <sup>3</sup>He and <sup>7</sup>Li-induced reactions) reported earlier. However, the reaction products, <sup>97</sup>Ru and <sup>95</sup>Tc, obtained in the <sup>12</sup>C + <sup>89</sup>Y reaction will be useful in tracer studies in laboratory scale. The comparatively high yield of 95Tc may be due to the contribution of <sup>89</sup>Y(<sup>12</sup>C, 2*p*4*n*)<sup>95</sup>Tc, <sup>89</sup>Y(<sup>12</sup>C, 6*n*)<sup>95</sup>Rh(ε)<sup>95</sup>Ru(ε)<sup>95</sup>Tc and <sup>89</sup>Y(<sup>12</sup>C,  $p5n$ <sup>95</sup>Ru( $\varepsilon$ )<sup>95</sup>Tc reactions, where reaction thresholds for the production of  $95Tc$ ,  $95Rh$  and  $95Ru$  are 55.1, 65.6, 58.9 MeV respectively. It is interesting to note that signatures of these radionuclides are not observed in the theoretical model calculation.

The methods developed to separate nca Ru and nca Tc from the yttrium target matrix are simple, fast, reliable and efficient. High separation factors were achieved under the optimum condition which in turn provides high purity products, 97Ru and 95Tc, as seen from the corresponding *D* values. More than 98% Tc was separated from bulk yttrium while quantitative separation of nca Ru was achieved in LLX.

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