SIMULATIONS ON NICKEL TARGET PREPARATION AND SEPARATION OF Ni(II)-Cu(II) MATRIX FOR PRODUCTION OF RADIOISOTOPE 64Cu

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ABSTRACT

SIMULATIONS ON NICKEL TARGET PREPARATION AND SEPARATION.OF Ni(II)-Cu(II) MATRIX FOR PRODUCTION OF RADIOISOTOPE ^{64}Cu. *The simulations on Nickel target preparation and separation of Ni(II)-Cu(II) matrix has been carried out as a preliminary study for production of medical radioisotope Cu-64 based on nuclear reaction of ^{64}Ni (p,n) ^{64}Cu. The nickel target preparation was performed by means of electroplating method using acidic solution of nickel chloride - boric acid mixture and basic solution of nickel sulphate – nickel chloride mixture on a silver- surfaced-target holder. The simulated solution of Ni(II) – Cu(II) matrix was considered as the solution of post-proton-irradiated nickel target containing both irradiated nickel and radioactive copper, but in the presented work the proton irradiation of nickel target was omitted, while the radioactive copper was originally obtained from neutron irradiation of CuO target. The separation of radioactive copper from the nickel target matrix was based on anion exchange column chromatography in which the radiocopper was conditioned to form anion complex $CuCl_4^{2-}$ and retained on the column while the nickel was kept in the form of Ni^{2+} cation and eluted off from the column. The retained radioactive copper was then eluted out the column in the condition of dilute HCl changing back the copper anion complex into Cu^{2+} cation. It was found that the electroplating result from the acidic solution was more satisfied than that from the basic solution. By conditioning the matrix solution at HCl 6 M, the radioactive copper was found in the forms of Cu^{2+} and $CuCl_4^{2-}$ while the nickel was totally in the form of Ni^{2+}. In the condition of HCl 9 M, the radioactive copper was mostly in the form of $CuCl_4^{2-}$ while the nickel was found as both Ni^{2+} and $NiCl_4^{2-}$. The best condition of separation was in HCl 8 M in which the radioactive copper was mostly in the form of $CuCl_4^{2-}$ while the nickel was mostly in the form of Ni^{2+}. The retained $CuCl_4^{2-}$ was then changed back into Cu^{2+} cation form and eluted out the column by using HCl 0.05 M. The γ-spectrometric analysis showed a single strong peak at 511 keV which is in accord to γ-annihilation peak coming from positron decay of Cu-64, and a very weak peak at 1346 keV which is in accord to γ-ray of Cu-64.*

Keywords *: Nickel target preparation, Radioisotope Cu-64, Separation of Ni(II)-Cu(II) matrix, Nuclear reaction of ^{64}Ni (p,n) ^{64}Cu, Anion exchange chromatography.*

ABSTRAK

SIMULASI PREPARASI TARGET NIKEL DAN PEMISAHAN MATRIK Ni(II)-Cu(II) UNTUK PRODUKSI RADIOISOTOP ^{64}Cu. *Simulasi proses preparasi target nikel dan pemisahan matrik Ni(II)-Cu(II) telah dipelajari sebagai langkah awal dalam penguasaan teknologi produksi radioisotop Cu-64 melalui reaksi nuklir ^{64}Ni (p,n) ^{64}Cu. Target nikel disiapkan melalui elektroplating suasana asam larutan nikel klorida – asam borat dan elektroplating suasana basa larutan nikel klorida – nikel sulfat pada keping penyangga target dengan permukaan perak. Larutan simulasi matrik Ni(II) – Cu(II) dianggap sebagai larutan target nikel pasca iradiasi yang mengandung nikel dan tembaga radioaktif, tetapi dalam percobaan ini iradiasi nikel tidak dilakukan sedangkan tembaga radioaktif dihasilkan dari aktivasi target CuO menggunakan berkas neutron. Pemisahan radioisotop Cu dari matrik target nikel dilakukan berdasarkan teknik kromatografi kolom penukar anion dengan mengkondisikan spesi Cu berada dalam bentuk komplek anion $CuCl_4^{2-}$ sedangkan spesi Ni berada dalam bentuk kation Ni^{2+}. Spesi komplek anion $CuCl_4^{2-}$ yang tertahan dalam kolom resin penukar anion kemudian dielusi keluar kolom dengan mengubahnya kembali menjadi kation Cu^{2+} dalam kondisi HCl encer. Hasil percobaan menunjukkan bahwa elektroplating menggunakan larutan nikel suasana asam memberikan deposit elektroplating nikel yang lebih memuaskan dibandingkan dengan larutan nikel suasana basa. Pengkondisian larutan matrik dalam HCl 6 M menunjukkan bahwa spesi tembaga berada dalam bentuk Cu^{2+} dan $CuCl_4^{2-}$, sedangkan spesi nikel dalam bentuk kation Ni^{2+}. Dalam kondisi HCl 9 M, spesi tembaga berada dalam bentuk $CuCl_4^{2-}$, sedangkan spesi nikel dalam bentuk Ni^{2+} dan $NiCl_4^{2-}$. Kondisi pemisahan yang terbaik adalah dalam HCl 8 M yang memberikan kondisi untuk spesi tembaga berada dalam bentuk $CuCl_4^{2-}$, sedangkan spesi nikel dalam bentuk kation Ni^{2+}. Selanjutnya spesi $CuCl_4^{2-}$ yang tertahan di dalam kolom diubah kembali*

menjadi Cu^{2+} dan dielusi keluar kolom dengan HCl 0,05 M. Pemeriksaan spektrometri-γ menunjukkan puncak kuat pada energi 511 keV yang sesuai dengan energi radiasi γ-anihilasi radioisotop ^{64}Cu dan puncak lemah pada 1346 keV yang sesuai dengan energi radiasi γ dari radioisotop ^{64}Cu.

***Kata kunci** : Pembuatan target nikel, Radioisotop ^{64}Cu, Pemisahan matrik Ni(II)-Cu(II), Reaksi nuklir ^{64}Ni (p,n) ^{64}Cu, Kromatografi penukar anion.*

INTRODUCTION

Radioisotope ^{64}Cu has a half-life of about 12.7 hours and decays by various ways, i.e. : positron emission (17.86 %), beta decay (39.0 %), electron capture (43.075 %) and internal conversion (0.475 %), emitting 0.5787 MeV of β^-, 0.6531 MeV of β^+ and 1.34577 MeV of γ radiations[1]. It has been known as medical radioisotope widely used in the form of various radiopharmaceuticals for both diagnostic and therapeutic purposes due to its emitted γ-annihilation and β^- radiations[2-6]. However, the potency of the application of this radioisotope has not been able to be expoloited at the domestic nuclear medicine facilities, mostly due to the lack of technical capabilities on the production of the radioisotope and its radiopharmaceuticals.

Radioisotope ^{64}Cu can be produced in a reactor or cyclotron. In a reactor, ^{64}Cu is obtained using either thermal or fast neutrons *via* (n,γ) or (n,p) reaction on ^{63}Cu or ^{64}Zn targets respectively. The method based on simple nuclear reaction of ^{63}Cu (n,γ) ^{64}Cu even though using a high-enriched ^{63}Cu target will produce a low specific activity of ^{64}Cu which is not suitable for medical applications. The (n,p) nuclear reaction on high-enriched ^{64}Zn target is another alternative method for producing ^{64}Cu in a nuclear reactor[7]. This method needs fast neutron fraction but the irradiation facility for the fast neutron fraction is not available in BATAN's G.A. Siwabessy reactor. Study on this reaction in BATAN's G.A. Siwabessy reactor, therefore, has a problem related to the formation of ^{65}Zn as the main produced radionuclide from the (n,γ) reaction which will be produced in higher yield than the expected ^{64}Cu. The nuclear reactions based on bombardment using proton beam in cyclotron is accordingly preferred to produce carrier-free ^{64}Cu suitable for medical purposes. The nuclear reaction of ^{68}Zn (p,αn) ^{64}Cu has been studied and reported by Bonardi et al[8], but this method needs a high energy of the proton beams (more than 30 MeV) those are not suitable with the BATAN's cyclotron machine. The nuclear reaction of ^{64}Ni (p,n) ^{64}Cu[9,10] which needs a lower energy of proton beams those are available in BATAN's cyclotron machine (less than 25 MeV) might be a better choice, although the high-enriched ^{64}Ni is very expensive due to its very low natural abundance (only 0.926 %)[11].

As a preliminary step to master production technology of ^{64}Cu, the presented work is aimed to study the separation of radioactive copper from a matrix of proton irradiated natural nickel as the target material instead of the high-enriched ^{64}Ni. The target material can be provided as electroplated deposit on silver-surface-target holder that is suitable for proton irradiation in the BATAN's Cyclotron Facility. Based on the nuclear reaction of ^{64}Ni (p,n) ^{64}Cu, the radionuclidic separation of ^{64}Cu from the matrix of Ni target by means of ion exchange column chromatography was studied.

It has been known that the chemical matrix of Ni(II)-Cu(II) mixture can be separated by using Dowex anion exchange resin based on the ability of Cu(II) in forming a chloro-complex anion at a certain condition in which the Ni(II) is still in the form of Ni^{2+} cation[12]. The chloro-complex anion of Cu(II) is thus expected to be bound on the anion exchanger resin while the Ni(II) cation is not, so that the later can be easily eluted out from the column. Due to technical and economical reasons, the presented work was carried out by using unirradiated natural nickel target, while the radioactive copper was simulated using neutron-irradiated natural CuO. The result of this presented work is expected to be applicable for routine procedure in separation of radioisotope ^{64}Cu from the matrix of proton-irradiated Ni target based on nuclear reaction of ^{64}Ni (p,n) ^{64}Cu.

EXPERIMENTAL

Material and equipment

All chemical materials were pro analysis grade and used without further purifications. Copper oxide (CuO), nickel chloride ($NiCl_2.6H_2O$), hydrochloric acid (HCl), dimethyl glyoxime (DMG) were produced by E.Merck, while Dowex AG 1X8 (Cl^-) was obtained from Bio-Rad. De-mineralized water (aqua DM) was obtained

from the Water Purification Facility at the Center of Multi Purpose Reactor, BATAN, Serpong. Quartz tubes of neutron-irradiation grade used as irradiation ampoules, as well as alumunium capsules for irradiation, which were made of highly pure aluminium, were supplied by local private company. For obtaining ^{64}Cu, an appropriate amount of CuO was irradiated in the rabbit system of the G.A. Siwabessy reactor (BATAN, Serpong).

Separation of Ni(II) – Cu(II) matrix by means of ion exchange column chromatography was performed using Econo-column 737-1010 (Bio-Rad USA). A gamma-spectrometry system equipped with a multi channel analyzer (Canberra 1000) and an HP-Ge detector (Canberra Industries) was used for radionuclidic analysis. Prior to its use, the spectrometer was calibrated using standard sealed-sources of ^{133}Ba (302.85 and 356.01 keV), ^{137}Cs (661.64 keV) and ^{60}Co (1173.23 and 1332.51 keV) from Du Pont[13]. A well-type dose calibrator (Atom Lab) was used for radioactivity measurement.

Preparation and dissolution of Nickel target

Two types of electroplating solution of nickel target were prepared by adopting procedures written by Kopeliovich[14]. The first was by dissolving 20.24 g of $NiCl_2.6H_2O$ and 4.5 g of H_3BO_3 in about 100 mL of water giving the pH of solution less than 3. It was then followed by dilution to 150 mL with water. The second electroplating solution was prepared by dissolving 40 g of $NiSO_4.6H_2O$ and 5 g of $NiCl_2.6H_2O$ in about 100 mL of water, followed by pH adjustment to more than 7 by addition of NH_4OH solution. The solution was finally diluted to 150 mL with water. The resulting solutions were transferred into a closed polyethylene bottle and kept as nickel-electroplating solutions until used.

Preparation of target was carried out by electroplating of the nickel solution on a silver-surface-target holder. A known-weight-silver-surface-target holder was placed as a side wall of the electroplating cell as shown in Figure 1, and functioned as a cathode. A piece of Pt-anode was placed at another side of the cell. The nickel-electroplating solution was then poured into the electroplating cell, and the electroplating steps were performed by applying varieties of currents. After finishing the electroplating, the solution was retransferred into the header by means of vacuum pressure. The target holder was released from the cell, rinsed with water and dried before being weighed to measure the amount of the resulting nickel deposit. The target was ready to be irradiated with a proton beam in cyclotron irradiation facility, but in case of this simulation procedure, the irradiation step was omitted.

For the dissolution of the nickel target, the target holder, which has the electroplated nickel on its surface, was placed as the bottom of the dissolution cell. About 10 mL of HCl (varieties in molarities) was then transferred into the dissolution cell. The cell was then heated for up to 80 - 90°C with occasionally shaking. Fews drops of H_2O_2 were added to speed the dissolution process. The resulting nickel solution was then transferred into the header by vacuum pressure and then transferred into receiving bottle for further process (namely Solution I).

Neutron irradiation and dissolution of Copper target

An amount of about 50 mg of CuO powder was placed in a quartz irradiation ampoule. The ampoule was then closed by welding and placed in an inner aluminium capsule. The inner capsule was then closed by welding and sent into rabbit system facility of the G.A. Siwabessy reactor for irradiation. The irradiation was performed for about 1 hour after which the irradiated CuO target was brought into a receiving hot cell. The quartz ampoule was then removed from the inner aluminium capsule, transferred into processing glove box and broken at the tip to transfer its content (post-irradiated CuO) into a 100 mL glass beaker. The irradiated CuO was then dissolved in 10 mL of HCl (varieties in molarities). The resulting solution was named as Solution II and cited for radioactivity measurement and γ-spectrometric analysis. For further separation experiment, this solution was mixed in an equal volume with the Solution I to get simulated matrix solution of post-irradiated nickel target containing radioactive copper.

Separation of ^{64}Cu from the simulated Ni(II) – Cu(II) matrix solution

As this work is aimed to study separation technique for irradiated target matrix based on nuclear reaction of ^{64}Ni (p,n) ^{64}Cu, the simulated solution of Ni(II) – Cu(II) matrix was considered as the solution of post-proton-irradiated nickel target containing both irradiated nickel and radioactive copper. In the presented work the proton

irradiation of nickel target was omitted, while the radioactive copper was originally obtained from neutron irradiation of CuO target.

The solution of nickel target (Solution I) was mixed with the solution of post-irradiated copper target (Solution II) in equal volume composition (5 mL each). The 10 mL of resulting solution mixture was loading into chromatographic-Econo-column 737-1010 containing 4 cm-height of pre-conditioned-Dowex 1X8 (Cl^- 100 – 200 mesh) anion exchanger resin in corresponding to the effluent to be used. The eluting fraction of the loaded matrix solution was collected in the waste receiver. The column was then eluted with 1×10 mL of concentrate HCl (molarities were varied in corresponding to that of the loaded solution) followed by further elution using 2×5 mL of 0.05 M HCl. Each eluate fraction was subjected to radioactivity measurement and γ-spectrometric analysis.

Radioactivity measurement and radionuclidic analysis

The radioactivity measurement was carried out using a well-type dose calibrator. To simplify the measurement procedure, the geometric consideration was ignored. Radionuclidic analysis was conducted to the Solution II (post-neutron-irradiated CuO) and the resulting dilute HCl. fractions (retrieved from the column after eluting it with HCl 0.05 M). A 5 to 20 μL of sample was pipetted out, spotted and absorbed on 2-cm-diameter filter paper. The filter paper was dried and put into a small plastic bag and then counted at the energy range of 150 – 1600 keV by using a calibrated γ-spectrometer to gain the γ-ray spectra.

Chemical analysis

The chemical analysis was performed to see whether the radioactive copper fraction was chemically contaminated with nickel. An aliquot of the sample solution was spotted on a piece of filter paper followed by spotting solution of 1 % DMG in acidic ethanol and then the paper was held over ammonia vapour. A red spot or colouration was produced for a positive test result[15].

RESULTS AND DISCUSSION

Natural nickel comprises five isotopes[11]. Their natural abundance, (n,γ) and (p,n)-nuclear reactions and products are shown in Table 1. There are three radioisotopes and two stable isotopes of nickel those can be produced by (n,γ)-nuclear reaction on natural nickel target and five isotopes by the (p,n)-nuclear reaction. However, based on the combination of natural abundance and half-life of the corresponding produced radioisotopes, the ^{64}Cu will be expected to be the most dominant product compared to the others. There is no radioactive nickel would be worried to contaminate the resulting radioactive copper.

From the (n,γ)-nuclear reaction, the production of longer half-life nickel radioisotopes (^{59}Ni and ^{63}Ni) will be insignificant in the applied duration time of irradiation, whereas the shorter half-life of nickel radioisotope (^{65}Ni) will quickly decay and almost none will be left by the end of the process. From the (p,n)-nuclear reaction, the producing copper radioisotopes, except the ^{64}Cu, have short half-lives so that it can be expected that the ^{64}Cu radioisotope is the only radioactive copper that is important to be considered. Based on the above-mentioned reasons, this presented study omitted the irradiation of nickel, neither by neutron nor proton, while the ^{64}Cu was obtained from ^{NAT}Cu (n,γ) ^{64}Cu instead of ^{NAT}Ni (p,n) ^{64}Cu nuclear reaction. In the other side of view, the natural Cu comprises two isotopes, ^{63}Cu and ^{65}Cu, with the abundance of 69.17 % and 30.83 % respectively. The later will produce ^{66}Cu on (n,γ)-nuclear reaction that can be ignored during this study because its half-life is only 5.12 minutes[11].

Table 1. The natural isotopes of nickel and their (n,γ) and (p,n)-nuclear reactions product.

ISOTOPE	ABUNDANCE (%)	(n,γ)-PRODUCT			(p,n)-PRODUCT		
		ISOTOPE	$t_{1/2}$	E_γ (keV)	ISOTOPE	$t_{1/2}$	E_γ (keV)
^{58}Ni	68.077	^{59}Ni	7.5×10^4 a	No γ	Cu-58	3.2 s	511, 1454
^{60}Ni	26.223	^{61}Ni	stable	--	Cu-60	23 m	511,1332
^{61}Ni	1.140	^{62}Ni	stable	--	Cu-61	3.4 h	511,283
^{62}Ni	3.634	^{63}Ni	100 a	No γ	Cu-62	9.74 m	511,1173
^{64}Ni	0.926	^{65}Ni	2.52 h	1481	Cu-64	12.7 h	511,1346

NOTE : All of copper radioisotopes produced by (p,n)-nuclear reaction on natural nickel are β^+-emitters, so they release annihilation radiation observed as γ-ray at 511 keV.

The electroplating method is the most suitable technique in solid target preparation for irradiation in BATAN's Cyclotron facility. In the presented work, the nickel electroplating was performed based on the method reported by Kopeliovich[14]. The electroplating cell and the silver-surface-target holder systems used were similar to those for preparation of thallium target in production of ^{201}Tl[16] as shown in Figure 1. The results of repeated electroplating procedures with varieties conditions are resumed in Table 2.

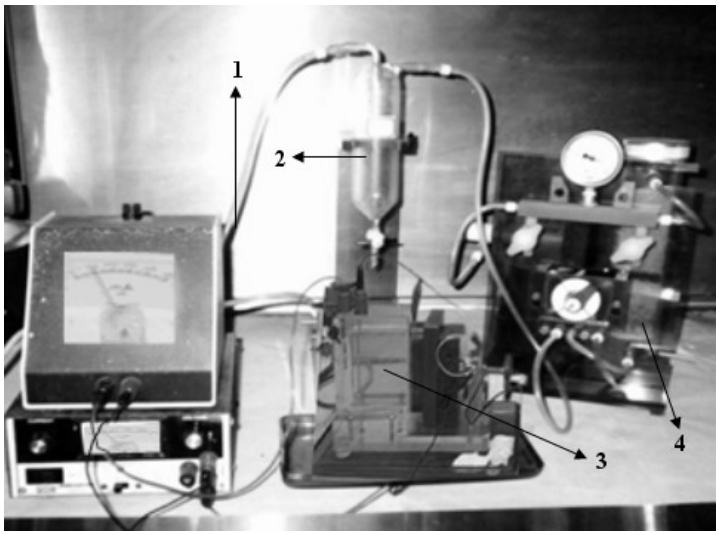

Figure 1. Nickel electroplating cell with supporting devices. 1. Volt and ampere meter, 2. Header for solution, 3. Electroplating cell containing nickel solution with target holder at the right side, 4. Mini vacuum pump,

It can be seen from Table 2 that the better conditions for nickel electroplating were found to be using the electroplating solution of pH < 3 (containing boric acid and nickel chloride) and the applied current of 100 - 120 mA. This result was then used for further experiments. The resulting nickel deposits were dissolved in varieties molarities of HCl those are corresponding to molarities of HCl used for simulation of column chromatographic separation.

The simulated solution of post-proton-irradiated nickel must contain both nickel (as target material) and radioactive copper (as the result of proton irradiation on nickel target). Accordingly, a 10 mL of matrix solution containing equal volume of Solution I (nickel target solution) and Solution II (post-irradiated copper solution) was loaded into the anion exchanger column. The separation fractions comprised three eluates, *i.e.* 1 × 10 mL of waste, 1 × 10 mL of concentrate HCl (varieties in molarities) and 2 × 5 mL of HCl 0.05 M. The distributions of radioactive copper in the resulting fractions are summarized in Table 3.

Table 2. Electroplating result in simulation of nickel target preparation[1)].

No.	Current (mA)	pH	Amount of deposited Ni (mg)	Deposit thickness (mg/cm^2)	Remark on the resulting deposit
1.	100	>7	430.0	30.71	Bad fluctuation, bad quality of surface, mechanically unstable
2.	100	>7	180.0	12.86	
3.	100	>7	323.0	23.07	
4	80	<3	366.3	26.16	Needs longer time to get suitable thickness, unstable current
5	100	<3	443.6	31.69	Good fluctuation, good quality of surface, mechanically stable, the applied current was more stable
6	100	<3	417.9	29.85	
7	120	<3	526.8	37.63	
8	120	<3	499.9	35.71	
9	140	<3	587.9	41.99	Bad quality of surface, mechanically unstable
10	160	<3	680.4	48.60	

NOTE : 1). All electroplating procedures were performed for 5 hours.

Table 3. Distribution of radioactive copper in the resulting separation fractions

Molarities of HCl used to dissolve the target	6 M	8 M			9 M
Volume of matrix solution loaded into the column (mL)	10	10	10	10	10
Radioactivity of matrix solution loaded into the column (mCi [%])	249 [100]	8.97 [100]	2.54 [100]	2.50 [100]	0.741 [100]
Distribution of radioactivity after loading the matrix solution					
Column (%)	96.79	99.55	98.43	98.80	93.25
Eluate (%)	0.23	0.11	0.50	1.61	6.33
Activity loss (Error, %)	+2.98	+0.34	+1.07	–0.41	+0.42
Distribution of radioactivity after eluting the column with corresponding concentrate HCl (6 M, 8 M or 9 M)					
Column (%)	65.74	87.63	80.71	97.20	90.28
Eluate (%)	29.56	9.96	15.28	2.80	8.07
Activity loss (Error, %)	+1.49	+1.96	+2.44	–1.20	–5.10
Distribution of radioactivity after eluting the column with dilute HCl (0.05 M, 2×5 mL)					
Column (%)	0.38	3.68	0.41	26.48	13.17
Eluate-I (%)	62.97	74.92	77.89	68.03	77.60
Eluate-II (%)	0.21	0.21	0.51	1.63	6.73
Activity loss (Error, %)	+2.18	+8.82	+1.90	+1.06	–7.22

The resulting data summarized in Table 3 were found to be unsatisfactory in term of statistical precision and accuracy. This was suspected because of the geometric of samples measurement that was not taken into consideration. Nevertheless, the qualitative trend of separation is consistent in which the radioactive copper was bound on the anion exchanger at the condition of concentrate HCl (8 M and 9 M) due to the formation of anion complex of $CuCl_4^{2-}$ and then eluted off the column by elution using dilute HCl (0.05 M) because the anion complex changes back into Cu^{2+} cation. In the condition of HCl 6 M, both ionic species of copper are maybe formed[17] so that the radioactive copper is significantly observed in both column and HCl 6 M eluate. The observed phenomena can be explained as below :

1. $Cu^{2+}_{(aq)} + 4\ HCl_{(8\ M\ or\ 9\ M)} \rightarrow CuCl_4^{2-}{}_{(aq)} + 4\ H^+_{(aq)}$
2. $CuCl_4^{2-}{}_{(aq)} + 2$ Dowex-$(Cl^-) \rightarrow [Dowex]_2$-$(CuCl_4^{2-}) + 2Cl^-_{(aq)}$
3. $[Dowex]_2$-$(CuCl_4^{2-}) + 2\ HCl_{(0.05\ M)} \rightarrow 2$ Dowex-$(Cl^-) + Cu^{2+}_{(aq)} + 4Cl^-_{(aq)} + 2H^+_{(aq)}$.

The above-mentioned phenomena seem to agree with the published data presented by Jentzsch and Frotsher showing that copper will form anion complex in HCl with concentration of higher than 4 N but not in HCl with concentration of lower than 1.5 N[12]. In other report, Wanty et al[17] stated that in the condition of HCl 7 M, copper species can be in the form of $CuCl_2$ or $CuCl_4^{2-}$. Therefore, in the condition of HCl 6 M the copper spesies can be in the forms of both cationic and anionic species.

Table 4. DMG-spot test analysis on separated eluate fraction.

Molarities of HCl used to dissolve the target	6 M	8 M			9 M
Color visualization on the concentrate HCl eluat	Green	Green	Green	Green	Greenish pale
DMG spot test on HCL 0,05 M fraction	–	–	–	–	+

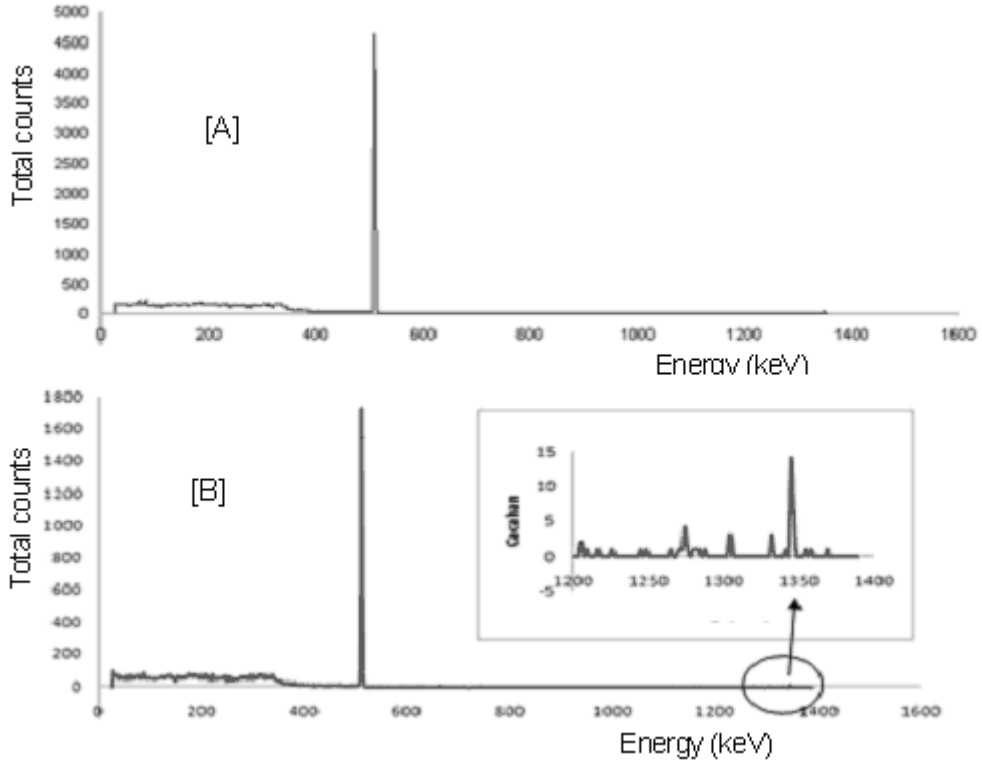

Figure 2. Typical γ-spectra of post-neutron-irradiated CuO solution [A] and radioactive copper fractions in HCl 0.05 M [B].

On the other side, the nickel will be in the form of anion complex in HCl with concentration between 10 to 11 N. In HCl with concentration of less than 7.5 N, however, the anion complex of nickel is not formed[12]. Accordingly, it can be expected that the nickel is not trapped on the anion exchanger in the condition of HCl with a concentration of less than 7.5 M. In order to check the existence of nickel in the eluted radioactive copper fractions, the dimethylglyoxime (DMG) spot test was then applied. The result of spot test by using DMG in the HCl 0.05 M fractions which were assumed to contain radioactive copper is summarized in Table 4.

The spot test data indicate that in either HCl 6 M or 8 M, nickel was not trapped on the column as it was in the form of $Ni^{2+}_{(aq)}$ cation having strongly green color. The column was washed free of nickel by using corresponding concentrate HCl. When the resin was then eluted with dilute HCl (0.05M), no more Ni^{2+} was left so that the DMG spot test gave negative result. The use of HCl 9 M, on other side, showed that the formation of anion nickel complex $NiCl_4^{2-}$ has begun to take place. A part of nickel was still in the form of Ni^{2+} giving greenish pale color of HCl 9 M eluate, the other part was in the form of nickel anion complex $NiCl_4^{2-}$ which was then bound in the anion exchanger resin and then changed again into Ni^{2+} cation form when the resin was eluted using dilute HCl solution, so that the DMG spot test was positive giving red coloration.

1. $Ni^{2+}_{(aq)} + 4\ HCl_{(\geq 9M)} \rightarrow NiCl_4^{2-}{}_{(aq)} + 4\ H^+_{(aq)}$
2. $NiCl_4^{2-}{}_{(aq)} + 2$ Dowex-$(Cl^-) \rightarrow [Dowex]_2$-$(NiCl_4^{2-}) + 2Cl^-_{(aq)}$
3. $[Dowex]_2$-$(NiCl_4^{2-}) + 2\ HCl_{(dilute)} \rightarrow 2$ Dowex-$(Cl^-) + Ni^{2+}_{(aq)} + 4Cl^-_{(aq)} + 2H^+_{(aq)}$.
4. $Ni^{2+}_{(aq)} + 2\ DMG_{(EtOH)} \rightarrow Ni(DMG)_{2\ (red)} + 2\ H^+_{(aq)}$
   $NH_4OH_{(g)}$

In term of the radionuclidic analysis results, the radioactive copper fractions were subjected to γ-spectrometric analysis at the range of E_γ between 150 keV to 1600 keV. The typical γ-spectrum is presented in Figure 2 as compared to the γ-spectrum of post irradiated copper solution. Both spectra seem to be very similar showing single peak at about 511 keV as γ-annihilation radiation in accord to the data presented in Table 1. A

very small peak which can be observed under partial magnification of the spectra shows the γ-energy of about 1346 keV that was emitted by the radioactive Cu-64.

CONCLUSSION

Nickel target for production of radioactive ^{64}Cu based on the nuclear reaction of ^{64}Ni (p,n) ^{64}Cu was prepared by electroplating of acidic nickel chloride solution containing boric acid on a silver-surface-target holder. The nickel target is dissolved in solution of HCl 8 M and mixed with solution of radioactive copper from neutron-irradiation of CuO target assuming as matrix solution containing post irradiated nickel and radioactive copper, specifically ^{64}Cu as the product of the proton irradiation of nickel target.

The simulation of Ni(II) – Cu(II) separation procedure which had been performed in the presented study was found to be applicable as a separation method for production of radioactive ^{64}Cu based on the nuclear reaction of ^{64}Ni (p,n) ^{64}Cu. The resulting ^{64}Cu was able to be separated from the post-proton-irradiated nickel matrix by means of anion exchange chromatography in which the copper was conditioned in HCl 8 M to form tetrachlorocopperate-(II) anion, $CuCl_4^{2-}$, which was bound to the anion exchanger, while the nickel was kept in the form of Ni-(II) cation, Ni^{2+} which was eluted out of the column The retained $CuCl_4^{2-}$ was then eluted out from thr column as Cu^{2+} cation by using HCl 0.05 M.

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