Separation of Radioisotope Iodine-131 from Radiotellurium-131 using Dowex-1x8 resin column chromatography

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Abstract

Iodine-131 (^{131}I) radioisotope is generally produced by irradiating natural tellurium targets in a nuclear reactor. Tellurium-130 (^{130}Te) will absorb a neutron particle and activate to form ^{131}Te which will then decay to form ^{131}I. The resulting ^{131}I radioisotopes are then separated using two methods, namely wet distillation and dry distillation. However, these methods have some disadvantages, such as requiring complex devices and producing contaminated ^{131}I during the evaporation process. To overcome these disadvantages and to make the process simpler, a separation method using an ion exchanger resin column has been developed. The purpose of this research is to study the parameters that influence the separation method such as resin exchange capacity and iodine-tellurium distribution coefficient.

Research started with irradiating a TeO_2 target in a nuclear reactor through a core reaction $^{130}Te\ (n,\gamma)\ ^{131}Te$. The next steps were to determine the resin of anion exchange capacity, the distribution coefficient and the yield using column chromatography separation method with Dowex 1x8 resin and tetrabutylammonium bromide (TBAB) as an eluent. The eluate was injected followed by the addition of NaOH 2 N to produce Na^{131}I. From this research, we determined a resin exchange capacity of 3.42 meq/gram and a distribution coefficient of 60.29 mL/gram. Separation using the column chromatography method can produce Na^{131}I with a percentage yield of 90.63% clear solution, ranging from pH 6-7, with a radionuclide purity of 99.18% and radiochemistry purity of 99.05 ± 0.13%.

Keywords: Iodine-131, dowex 1x8 resin, tetrabutylammonium bromide (TBAB), column chromatography.

Introduction

Cancer is a disease that occurs from abnormal cell growth, turning normal cells into cancerous cells. Thyroid cancer is a type of cancer affecting the thyroid gland. Based on data from the Republic of Indonesia's Ministry of Health, approximately 8.2 million people died from cancer in 2012. Statistics from 2010-2013 show that thyroid cancer is one of the top 10 cancers in Indonesia and the numbers are increasing every year.

Among the risk factors for thyroid cancer are thyroid gland infection from mumps, obesity and iodine deficiency[1,2]. One of the effective methods to treat thyroid cancer is radiotherapy using iodine-131 (^{131}I). This method is better than the other available methods because ^{131}I radioisotopes work optimally and specifically on the thyroid gland. Moreover, this method is also painless for the patient and has a high success rate[1].

The ^{131}I radioisotopes have a half-life of 8 days as a β-ray transmitter (E_β = 0.606 MeV) and a γ-ray transmitter ($E\gamma$ = 365 keV) which can be developed by irradiating natural tellurium as a target in a nuclear reactor. Natural tellurium (^{130}Te, abundance 34%) will absorb a neutron particle and will activate to form ^{131}Te which will then decay to form ^{131}I. The resulting ^{131}I radioisotopes are then separated from the remaining inactivated tellurium target and from other radionuclides[3-6]. Currently, the most common ^{131}I separation methods are wet distillation and dry distillation. However, these methods have some disadvantages such as requiring complex devices and having a higher chance to produce contaminated ^{131}I during the evaporation process.

To overcome these disadvantages and to make the process simpler, a separation method using an ion exchanger resin column has been developed. The alternative option is to separate ^{131}I from other radionuclides using the column chromatography method with anion exchanger Dowex 1x8. This method is an efficient technique because of its low likelihood of contamination, its simple process and its ability to produce a high product yield. In a study by Chattopadhyay and Das[7], ^{131}I separation process using NaOH as a solvent and Dowex as a resin has been successfully done. However, the research did not consider parameters that can affect the separation method which are the resin exchange capacity and the iodium-tellurium distribution coefficient.

A research done by Kandil et al[8] determined the resin exchange capacity and distribution coefficient on HCl and NaOH with solvent concentration variation and shaking time variation. However, that research was performed using Amberlite as the resin. Therefore, further study was needed to determine the distribution coefficient using the Dowex 1x8 resin.

Our research seeks to separate ^{131}I using the column chromatography method with the Dowex 1x8 resin and also to determine the resin exchange capacity and distribution coefficient.

Material and Methods

Materials and Apparatus: Materials used in this research are tellurium dioxide (TeO_2; Sigma-Aldrich), Dowex resin 1x8 Cl^- form 100-200 mesh (Cl^- form; Sigma-Aldrich), silver nitrate ($AgNO_3$), sodium chloride (NaCl), hydrochloric acid (HCl), sodium hydroxide (NaOH), isopropyl alcohol (C_3H_7OH), ammonium hydroxide (NH_4OH), nitric acid (HNO_3), potassium chromate (K_2CrO_4), ethanol (C_2H_5OH), tetrabutilammonium bromide (TBAB), methylen chloride (MTC), methanol (CH_3OH), potassium nitrate (KNO_3), sodium nitrate ($NaNO_3$), sulfuric acid (H_2SO_4), sodium oxalate ($Na_2C_2O_4$), potassium permanganate ($KMnO_4$), sodium sulfide (Na_2S), methyl orange, phenolphthalein and bromocresol green. All materials are of E. Merck proanalysis grade. Apparatus used in this research are a magnetic stirrer, a pH meter, a UV-visible spectrophotometer, a Multi-Channel Analyzer (MCA), a Single Channel Analyzer (SCA), a Dose Calibrator and common laboratory glassware. The reactor used in this research is TRIGA 2000 Bandung.

Determination of Anion Resin Exchange Capacity: Dowex 1x8 resin (5 grams) was placed in a separation funnel and then rinsed with water. A burette filled with 500 mL of 0.3 M HCl was placed above the funnel and streamed at a constant rate of 20 mL/minute. The resultant effluent was collected in a 500 mL Erlenmeyer (Erlenmeyer A). After setting away the effluent, 10 mL of water and a few drops of methyl orange were added into Erlenmeyer A. Meanwhile, some isopropyl alcohol was added into a burette and dripped into the resin in Erlenmeyer A resulting in the solute color changing to yellow with a $pH < 3.9$. Collected effluent was set away and the resin must be kept wet.

The Erlenmeyer A flask was flushed with water and used again as a container, whilst in the burette 250 mL of 0.5M NH_4OH was added and dripped into the resin. After that, in the burette, 250 mL of water was added and dripped into the resin, so the flask now contains a mixture of NH_4OH and water. 100 mL of effluent was then moved into another Erlenmeyer (Erlenmeyer B) and 3 drops of 0.05% methyl orange and a few drops of 0.02M HNO_3 were added. The solute color changed to red. Then, a few drops of 0.5M NH_4OH were added to the solute, changing the colour to yellow. The solute was then further added with 1 mL of 5% K_2CrO_4 and then titrated with 0.1M $AgNO_3$ until the color changed (Titration volume counted as F).

Erlenmeyer A was flushed with water and used again as a container, whilst in the burette, 100 mL of 25% NaCl and 400 mL of water were added and dripped into the resin so that Erlenmeyer A will contain a mixture of NaCl and water. 100 mL of effluent was moved into another Erlenmeyer (Erlenmeyer C) and 1 drop of 0.1% bromocresol green was added. The solute was titrated with 0.3M HCl until the color changed (Titration volume counted as G).

Erlenmeyer A was flushed with water and used again as a container, whilst in the burette, 500 mL of 2% $NaNO_3$ was added and dripped into the resin so that Erlenmeyer A will contain 500 mL of $NaNO_3$. 25 mL of effluent was moved into another Erlenmeyer (Erlenmeyer D) and a few drops of 0.05% phenolphthalein and a few drops of 0.02 M HNO_3 were added until the color turned colorless. Then, 1 mL of 5% K_2CrO_4 was added into Erlenmeyer D and the solute was titrated with 0.1M $AgNO_3$ until the color changed (Titration volume counted as H). The resin exchange capacity was calculated with following formula: (ASTM D2187-94, 2004)[9].

Resin exchange capacity =
$([(10 \times (F+H) \times N] - 10 (G \times Na)])/W =$ milliequivalent/gram
where F = $AgNO_3$ average volume for NH_4OH titration, G = HCl average volume for NaCl titration, H = $AgNO_3$ average volume for $NaNO_3$ titration, W = resin's wet weight, N = $AgNO_3$ normality and Na = HCl normality.

Target TeO_2 Irradiation: Target TeO_2 (1 gram) was placed into a container with an inner aluminium lining. The container was then placed into a second container with an outer aluminium lining to be irradiated. Target TeO_2 was irradiated in TRIGA 2000 Bandung reactor with CTP (Centre Timble Position) using 10^{13} n/cm^2/s neutron flux. The irradiation process took 94.3 hours. The irradiated target was dissolved in HCl and NaOH [10].

Determination of Distribution Coefficient

Solvent Concentration Variation: Five prepared vials were added with 1 mL of solution from the target TeO_2 irradiation procedure. Solvent at a concentration variation of 2, 4, 6, 8 and 10 N, was added to the vials to make a total 5 mL. 50 mg of Dowex 1x8 was then added to each vial and the vials were shaken using a magnetic stirrer for 1 hour. After the shaking was done, each vial was filtered using No. 1 Whatmann filtration paper. Filtrate from each vial was measured using gamma spectrometry (Multi Channel Analyzer). This procedure was repeated using NaOH as a solvent with the same concentration variation[8].

Shaking Time Variation: 6 prepared vials were added with 5 mL of solution from the target TeO_2 irradiation procedure. Solvent with optimum concentration was then added to make a total of 5 mL. 50 mg of Dowex 1x8 was then added to each vial and the vials were shaken using a magnetic stirrer with a shaking time variation of 20, 40, 60, 80, 100 and 120 minutes. After the shaking was done, each vial was filtered using No. 1 Whatmann filtration paper. Filtrate from each vial was measured using gamma spectrometry (Multi Channel Analyzer). The procedure was repeated using NaOH as a solvent with the same concentration variation[8].

Separation of ^{131}I Radioisotopes: 1 mL of irradiated target TeO_2 from the Target TeO_2 Irradiation procedure was placed into a glass chromatography column (∅, 1 cm x 5 cm) that was pre-filled with 0.5 grams of Dowex 1x8. The column was washed using 10 mL of water and 5 mL of ethanol, then eluted with 5 mL of 0.63 mM TBAB solution in methylene chloride (MTC). Eluate was collected in 10 fractions (each fraction contained 5 mL eluate). Each fraction was dried using the syringe evaporation technique until completely dry. 1 mL of 2 N NaOH was then added into each fraction to produce Na^{131}I[7].

Determination of Na^{131}I Purity

Radionuclide Purity: Into a clear vial, 5 µL of Na^{131}I solution was added and the radioactivity was measured using MCA for 5 minutes. This procedure was repeated for each fraction obtained from column chromatography separation. Besides measuring using MCA, radioactivity can also be measured using Dose Calibrator as comparison.

Radiochemistry Purity: To measure purity, Whatmann No. 1 chromatography paper was used. The dimensions of the paper used were 1.5 cm x 16 cm. Numbers starting from -2, -1, 0 until 14 were written down in order, every 1 cm apart using a pencil. Filtrate from the separation procedure (1.2.4) was spotted at point 0 (zero) on the chromatography paper and dried at room temperature. After it had completely dried, the chromatography paper was hung on a chromatography column filled with 75% methanol allowing the solvent to reach point 14. After it had reached the point, the chromatography paper was lifted up, dried at room temperature and cut up to 1 cm pieces. The radioactivity was measured for each piece of the paper using the SCA instrument set up for measuring Na^{131}I radioisotopes.

Results and Discussion

Anion Resin Exchange Capacity: The purpose of determining the anion resin exchange capacity was to clarify whether the resin still has the same exchange capacity as the resin's catalog capacity. The procedure used in this research was based on the standard procedure published by ASTM code D2187-94 in 2004[9]. The result of this research indicates that the anion resin exchange capacity was 3.42 meq/gram (3.50 meq/dry gram from catalogue). The experimental data did not have too many differences. Therefore, it can be concluded that the resin used in this research has a good anion exchange capacity and can be used to separate ^{131}I using column chromatography.

Target TeO_2 Irradiation: The irradiation process done to target ^{130}Te in its oxide state (TeO_2), was activated with a neutron resulting in radioactive tellurium (^{131}Te) with 25 minutes half-life and will decay releasing β-rays and turn into ^{131}I with ±8 days half-life.

Determination of Distribution Coefficient (Kd): The purpose of determining the distribution coefficient was to find out the best solvent, the appropriate concentration of the solvent and the shaking time for the separation process using the column chromatography method. The distribution coefficient results obtained have been plotted in figure 2 and figure 3.

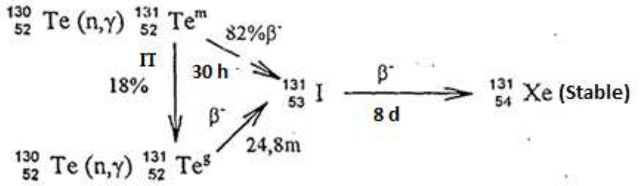

Figure 1: Decay scheme ^{131}Te – ^{131}I [6]

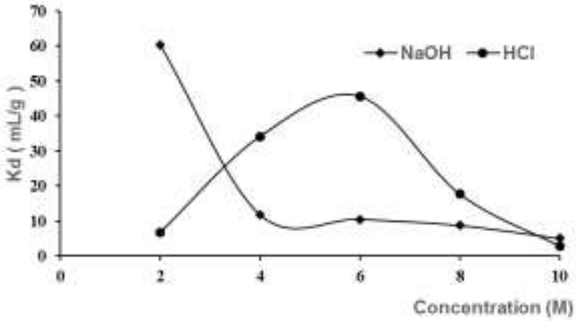

Figure 2: Distribution coefficient – solvent concentration variation chart

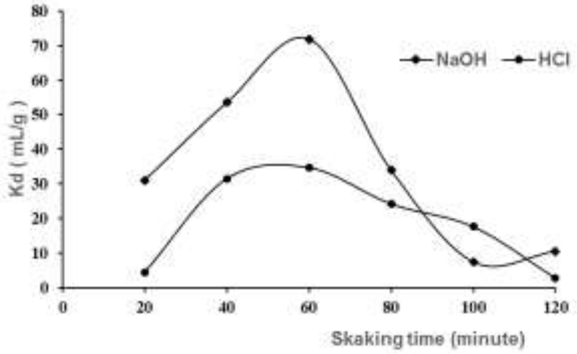

Figure 3: Distribution coefficient – shaking time variation chart

The values plotted in figure 2 and figure 3 are mean values from performing the procedure three times. As such, the statistic validity of this research could be accepted. As seen in figure 2, the biggest distribution coefficient for solvent concentration variation on NaOH is 2 N. As seen in figure 3, the biggest distribution coefficient for shaking time variation both on NaOH and HCl is 60 minutes. Results in figure 2 and figure 3 show that the radioactive samples used in this research were well distributed if NaOH solvent with 2 N concentration was used as a solute and the samples is shaken for 60 minutes. This means, the bigger is the distribution coefficient value, the easier it is for ^{131}I radioisotopes to be separated from iodium-tellurium mixture irradiation products.

A good distribution will affect how much the radioisotopes bind to resin and make the separation process much easier. This procedure has also been performed and proven to be effective in research done by Kandil et al.[8]

Separation of ^{131}I Radioisotopes: Elution process using TBAB leads to the binding of ^{131}I into TBA-^{131}I. The collected eluate was dried using evaporation in a water bath (the drying process of ^{131}I from organic solvent was stopped when there was no more vapor; indicating that a residue of ^{131}I was collected). Then, 1 mL of 2 N NaOH was added into the residue so a clear solution of Na^{131}I was produced with pH 6-7. The elution profile of ^{131}I separation is shown in figure 4.

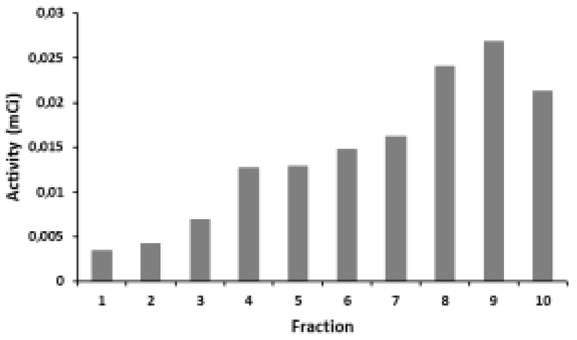

Figure 4: Na^{131}I radioisotopes elution profile

Figure 4 shows an increasing activity from the first fraction to the ninth fraction, showing that ^{131}I starts getting separated from the resin when eluted, but decreased on the tenth fraction indicating that the existence of ^{131}I in resin starts to disappear (separated from the resin). By repeating this procedure three times, the separation yield was 90.63% ± 2.84%.

Determination of Na^{131}I Purity: Determination of the Na^{131}I purity began with checking the pH of the obtained Na^{131}I; the results of which were pH 6-7 and also a clear solution. Radionuclide purity has been plotted in and presented in figure 5.

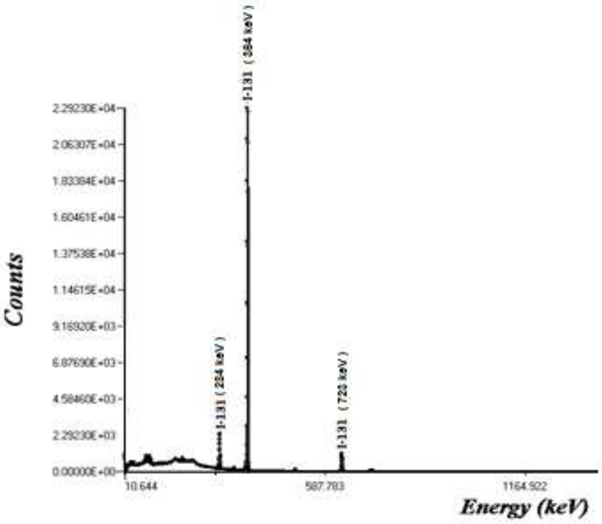

Figure 5: Na^{131}I radionuclide gamma spectrometry

Figure 5 is a plotted chart illustrating the radionuclide purity of Na^{131}I measured using the MCA instrument. The highest count of ^{131}I radioisotopes was 364 keV. Based on a calculation, Na^{131}I radionuclide purity is 99.18%.

The purity test process of ^{131}I solution was done using paper chromatography method with Whatmann No. 1 chromatography paper for the stationary phase and 75% methanol for the mobile phase. The ^{131}I was expected to be in an iodide (I$^-$) form and in 0.7-0.9 Rf range.

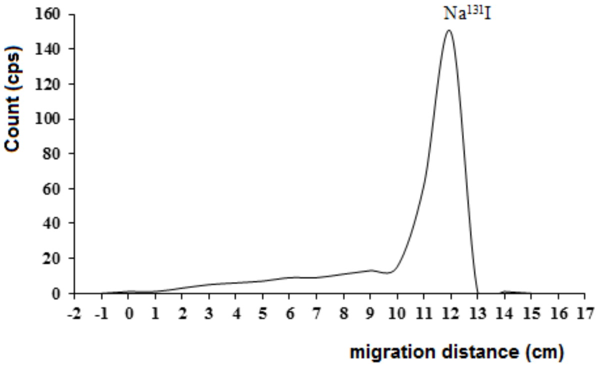

Figure 6: Na^{131}I radioisotopes chromatogram with Whatmann No. 1 for the stationary phase and 75% methanol for the mobile phase.

Figure 6 shows a migration from point 0 (zero) to point 12 and a drastic decrease from point 13 to point 16. The results show that Na^{131}I has a value of 0.75 Rf[6]. Additionally, Na^{131}I radiochemistry purity is 99.05 ± 0.13%.

Conclusion

Separation of Na^{131}I radioisotopes from tellurium target was obtained using column chromatography technique with anion exchanger Dowex 1x8. From this research, we determined the resin exchange capacity to be 3.42 meq/gram and the distribution coefficient to be 60.29 mL/gram. The technique used to determine the distribution coefficient used 2 N NaOH solvent with a shaking time of 60 minutes.

Separation using column chromatography method can produce a 90.63% ± 2.84 % yield percentage of Na^{131}I, a clear solution with a pH 6-7, radionuclide purity of 99.18% and radiochemistry purity of 99.05 ± 0.13%. In conclusion, Na^{131}I produced from the technique researched can be used as a radioisotope for medical purposes.

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