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FEASIBILITY STUDY ON VITRIFICATION OF SPENT ION EXCHANGE RESINS FROM TRIGA REACTOR MALAYSIA

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ABSTRACT

Feasibility studies on the vitrification of spent ion exchange resins combined with glass cullet powder have been conducted using a High Temperature Test Furnace. Bottle glass cullet powder was used as matrix material to convert the ash of the spent resins into a glass. Vitrification of spent ion exchange resins presents a reasonable disposal alternative, because of its inherent organic destruction capabilities, the volume reduction levels obtainable, and the durable product that it yields. In this study, the spent ion exchange resin from the PUSPATI TRIGA reactor of Nuclear Malaysia was combusted in a lab scale combustor and the resulting ash was vitrified together with glass cullet powder in a high temperature furnace to produce a stable spent resin ash embedded in glass. The factors affecting this immobilized waste, such as thermal stability, radiological stability and leachability have all been investigated. However, the outcome of these tests, which include the radionuclide activity concentration in the slag, the optimum conditioning temperature - in relation with volume reduction during vitrification - and the volume mixing ratio of matrix material were reported. It was found that the radionuclides present in spent resins were ^{54}*Mn,* ^{60}*Co and* ^{152}*Eu. The elementary chemical composition (carbon, hydrogen, nitrogen and sulphur) of spent resins was 27.6% C, 5.68% H, 2.04% N and 4.20% S, respectively. The maximum calorific value of spent resins was 1735 kJ/kg. The average activity concentrations of* ^{54}*Mn and* ^{60}*Co in ash at 200°C were 9,411 ± 243 Bq/Kg and 12,637 ± 201 Bq/Kg. flue gases containing* CO_2*, CO,* SO_2 *and NO started to be emitted above 200°C. The optimum conditioning temperature was also the highest tested, i.e. 900°C in 45 minutes, and the best mixing ratio ash to matrix material was also the highest, ie 1:9. Finally, the leaching analysis of slag at 900°C in 45 minutes showed that the leaching activity of* ^{60}*Co was below 0.5 Bq/mL.*

ABSTRAK

Kajian kebolehlaksanaan mengenai penghidratan resin pertukaran ion yang dibelanjakan digabungkan dengan serbuk cullet kaca telah dijalankan menggunakan Tungku Ujian Suhu Tinggi. Serbuk cullet kaca botol digunakan sebagai bahan matriks untuk menukarkan abu resin yang dibelanjakan ke dalam gelas. Vitrifikasi resin pertukaran ion yang dibelanjakan membentangkan alternatif pelupusan yang munasabah, kerana keupayaan pemusnahan organik yang wujud, tahap pengurangan jumlah boleh didapati, dan produk tahan lama yang dihasilkannya. Dalam kajian ini, resin pertukaran ion yang dibelanjakan dari reaktor PUSPATI TRIGA Nuklear Malaysia telah dibakar dalam pembakar skala makmal dan abu yang dihasilkan adalah vitrified bersama dengan serbuk cullet kaca dalam relau suhu tinggi untuk menghasilkan abu yang dibelanjakan yang stabil yang terbenam dalam kaca . Faktor-faktor yang mempengaruhi sisa imobilisasi ini, seperti kestabilan haba, kestabilan radiologi dan kebolehcapaian lejang telah diselidiki. Walau bagaimanapun, hasil ujian ini, yang termasuk kepekatan aktiviti radionuklida dalam sanga, suhu penyaman optimum - berkaitan dengan pengurangan volum semasa penghidratan - dan nisbah pencampuran isipadu bahan matriks dilaporkan. Telah didapati bahawa radionuklida yang terdapat dalam resin yang dibelanjakan adalah 54Mn, 60Co dan 152Eu. Komposisi kimia asas (karbon, hidrogen, nitrogen dan sulfur) yang dibelanjakan adalah 27.6% C, 5.68% H, 2.04% N dan 4.20% S. Nilai maksimum kalori yang dibelanjakan adalah 1735 kJ / kg. Purata aktiviti kepekatan 54Mn dan 60Co abu pada 200oC adalah 9,411 ± 243 Bq / Kg dan 12,637 ± 201 Bq / Kg. gas serombong yang mengandungi CO2, CO, SO2 dan NO mula dikeluarkan melebihi 200oC. Suhu penyaman optimum juga merupakan yang tertinggi diuji, iaitu 900oC dalam masa 45 minit, dan abu nisbah pencampuran *terbaik untuk bahan matriks juga tertinggi, iaitu 1: 9. Akhir sekali, analisis larutan terak di 900oC dalam 45 minit menunjukkan bahawa aktiviti larutan 60Co berada di bawah 0.5 Bq / mL.*

Keywords: ash vitrification, ^{60}Co, ^{54}Mn, leaching, radionuclide, stabilizing, spent ion exchange

ABBREVIATIONS

BDAT = Best Demonstrated Available Technology
DVB = Divinylbenzene
EPA = Environmental Protection Agency
SRTC = Savannah River Technology Centre
RTP = TRIGA PUSPATI

INTRODUCTION

In nuclear industry, ion exchange resins are the most common and effective treatment for removal of radioactive ions in a liquid radioactive waste management system. Hence, they are widely used in nuclear facilities and operations. Generally, most ion exchange resins consist of synthetic organic resins made of linear polystyrene cross-linked with divinylbenzene (DVB). These resins are solid materials insoluble in water, capable of exchanging particular ions within the polymer with ions in solution [1]. After a few cycles, the ion-exchange resin is to be replaced as the exchange capacity is significantly reduced [2]. The ion exchange resins arising from aqueous streams in nuclear facilities may contain fission and activation product such as ^{137}Cs, ^{90}Sr, ^{152}Eu, ^{60}Co, ^{106}Ru and ^{54}Mn with a half-life of 30, 29, 13.2, 5.3, 1, and 0.85 years, respectively [3]. These spent resins could be high in both organic and radioactive material and need special handling and treatment [1, 4].

Disposal of spent resin often becomes an economic problem because of the bulky volume of radioactive waste produced and only relatively few technologies are capable of economically stabilizing this waste. Mostly, the spent resins were stored in drums prior to their disposal. Leaching, which can occur when water contacts such waste, is the main factor that should be monitored. This is important in assessing the release of radionuclides from radioactive waste storage. It can proceed by several concurrent mechanisms such as diffusion, dissolution, erosion, etc., the relative importance of which may change with time, temperature, substances dissolved in the water, matrix material, radionuclides of interest, and other variables [5]. Pre-treatment prior to the conditioning step may reduce the volume of the spent resins. Besides volume reduction, the treatment may also alter the physical and chemical properties of the spent resins. This purpose can be reached by combustion of organic resins, which serves both volume reduction and change of waste properties [1, 6]. Generally preferred at present is to include vitrification in the conditioning. Vitrification is an attractive technology because it is capable of consistently producing a durable, leaching resistant waste form, while simultaneously minimizing disposal volumes through moisture evaporation, destruction of organic, and porosity reduction [8].

A Report from the Savannah River Technology Centre (SRTC) in the U.S.A mentioned that the Environmental Protection Agency (EPA) has declared vitrification to be *the Best Demonstrated Available Technology (BDAT)* for high-level radioactive liquid waste. However, the technology is more recently being applied to low-level and mixed wastes because of the advantages seen for high-level radioactive wastes [9]. In 2001, a bench-scale study of immobilization spent resin using direct vitrification on iron-enriched borosilicate glass as matrix material was accomplished by SRTC in U.S.A. The results from these studies stated that the vitrification method produced homogeneous and durable glasses [8]. Similar study was conducted by Hamodi, et al. on immobilization of spent ion exchange resin arising from nuclear power plant in Pakistan. The vitrification by novel borosilicate glass has been proven as to achieve volume reduction and long-term disposal for liquid radioactive waste. Therefore, the thermal treatment and vitrification of spent resins presents a reasonable disposal alternative because of its inherent destruction capabilities, the volume reduction obtainable, and the production of durable product [10]. In Malaysia, the one and only nuclear reactor is the 1MW research reactor named Reactor TRIGA PUSPATI (RTP). The reactor is located at Malaysian Nuclear Agency and has been operated since 1982.

In the RTP, the reactor water purification system uses a synthetic organic ion exchange resin of amberlite resin type IRN-150. The amberlite resin is a strongly acidic polystyrene ion exchange resin [3]. It basically filters out the non-active and radioactive corrosion products, corrosive ion impurities, and also fission product. The spent resins generated from RTP represent about 50 kg/year. Approximately 3 m^3 of aging spent resin are stored at present in the interim storage for nearly thirty years. The radionuclides content in the spent resins from RTP are ^{60}Co, ^{58}Co, ^{54}Zn, ^{124}Sb and ^{56}Mn [3]. In order to ensure safe storage and disposal, the spent resins from RTP will be treated by special techniques to reduce initial volume and produce a stable end product. Among these treatment techniques are thermal treatment and ash vitrification. Ash vitrification is applied at high temperature with addition of glass cullet powder as matrix material.

MATERIALS AND METHODS

Sampling of Spent Resins

The spent resins were obtained from the TRIGA PUSPATI (RTP) Reactor. The spent resins were firstly collected during reactor maintenance work, then stored in plastic drums. For analysis purposes the spent resins was scoped randomly from the plastic drums for about 5 kg.

Analytical Techniques

The characterization study of spent resins includes elemental composition analysis, activity of radionuclide concentration, thermal characteristics, calorific value, leaching of radionuclide test, and pH and conductivity measurement [1]. The *proximate analysis* is used to determine moisture content, volatile matter, fixed carbon and ash content using furnace and weighing scale. The thermal reaction of the spent resins was measured using a dynamic thermal analyser /thermo gravimetric analyser (DTA/TGA) (DTG-60H, Shimadzu) instrument. The spent resins were heated at a constant heating rate of 5°C/min from room temperature up to 900°C in an N_2 gas atmosphere. To investigate the elemental contents of both the spent resins and the glass cullet powder they were examined by Oxford INCA 400 X-Ray Energy Dispersive Spectrometer (EDS) at 200x magnification. The equipment is integrated with the Quanta 400 ESEM software to acquire the X-ray data for analysis of material samples. The constituents in both of the samples were compared to those of the standard sample material. The calorific value analysis is conducted according to ASTM D 3286-96 and ASTM E 711-87 using a bomb calorimeter (IKA, Model C5000). The radionuclide activity concentrations in the spent resins were analysed by a Canberra p-type high purity germanium (HPGe) gamma spectrometer system with 30% relative efficiency and a resolution of 1.9 keV at 1332.5 keV of ^{60}Co. The detector efficiency was calibrated using a multi nuclide standard in 350 mL plastic container with source containing certified concentrations of ^{241}Am, ^{109}Cd, ^{57}Co, ^{123m}Te, ^{51}Cr, ^{113}Sn, ^{85}Sr, ^{137}Cs, ^{88}Y and ^{60}Co. A Genie-2000 analysis software was used to analyse the spectrum of the samples. The activity concentration of the radionuclides was measured per unit weight or volume ($Bqkg^{-1}$ and BqL^{-1}). The preparation of the radionuclide leaching analysis was performed according to the European Standard leaching test Radionuclide Leaching Test of Spent Resins (pr EN 12457-3:2002).

The samples were submerged and agitated for 6 hours and 18 hours. As for the slag obtained from the vitrification, it was characterized by X-ray diffraction (XRD) analysis by Model X'Pert Pro Diffractometer system to determine the crystallinity of the sample, using CuK he samples were s°–80°, scan speed = 0.2 s/step, increment = 0.02, operating voltage = 40kV and operating current = 40mA). The crystalline phases were identified by comparing peak positions and intensities by referring to the library in the X'Pert Highscore software.

The pH measurements were performed using a calibrated pH meter Hanna Instrument Model pH211. The samples were placed in 250 mL distilled water, and then stirred for 24 hours. Conductivity was measured with a conductivity meter HACH model Session5. The samples were placed in 100 mL distilled water and stirred for 8 hours at room temperature and conductivity measurements were made in every 2 hours.

Combustion of Spent Resins

Approximately 20g of the spent resins were combusted in the Lab Scale Combustor (LSC) at setpoint temperature between 100°C and 500°C. The time of combustion was about 15 - 20 minutes. The LSC was made of two chambers. The first chamber is a glass tube in a tube furnace and the sample is inserted into this tube for combustion. This furnace, when in use, is maintained at 100 to 500°C. The air inlet is connected to a series of gas tanks, which compressed air and N_2. N_2 is used as an inert gas for purging purpose. Most of the time the compressed air is used for combustion and a gas flow meter is used to indicate the flow rate. The other end of the primary chamber is connected to the secondary chamber, which has an air inlet just before the inlet to the secondary chamber to allow for secondary air combustion. The air supply is a branch from the primary air supply. The secondary chamber is also made of a quart glass and is heated by a tube furnace. This furnace is set at a temperature of 950°C and is also filled with 'Alumina Balls' to ensure that the combustion gasses formed during combustion in the primary chamber goes through a 'torturous part' to get the longest residence time possible in the secondary chamber. After the secondary combustion chamber, the off-gasses are forced through a series of impinges to trap the pollutants that are in the combusted gasses through HEPA filters. This is the typical wet method of analysis for the monitoring of combustion gasses practiced in the industry. Finally the LSC is fixed to a vacuum pump to ensure that all gasses produced during combustion are forced to pass through the impingers before release to the atmosphere. The emission is measured by means of a portable flue gas analyser at the exhaust point. The emissions assessed were SO_x, NO_x, CO and CO_2. The ash was collected to record the volume reduction and to analyse the radionuclide concentration using a Canberra p-type high purity germanium (HPGe) gamma spectrometer system. The partitioning of radionuclide was predicted by calculation from the results of ash analysis.

Matrix Material Preparation

A clear glass bottle cullet powder was used as a matrix material. The glass cullet powder was prepared by grounding clear glass bottles cullet and sieving to a size of 250 clear glass bottle cder obtained from the collection of municipal solid waste.

Vitrification of Ashes from Combustion

The ash produced from spent resins in the Lab Scale Combustor (LSC) was immobilized in glass cullet powder. Resins from reactor RTP Nuclear Malaysia contained relatively high concentration of ^{60}Co [3]. Ash vitrification was carried out in the High Temperature Furnace, able to run from room temperature up to 1600°C with an automatic and programmable operation model. The experiment was conducted in a crucible with capacity of 100 millilitres in volume. The ashes from LSC were collected and vitrified at 700 to 1000°C to produce high durability material. The ashes were incorporated in glass at 1:9, 2:8 and 3:7 in volume ratio of ash to glass cullet powder. The heating rate of the furnace was setting at 20°C/min. A glassy surface of slag was obtained after 45 minutes. The slag was collected to record the volume reduction, to analyse the chemical durability leaching test using ASTM C1285-02 Test s for Determining Chemical Durability of Nuclear, Hazardous, and Mixed Waste Glasses and Multiphase Glass Ceramics by Canberra p-type high purity germanium (HPGe) gamma spectrometer system. Multiphase glass ceramic waste forms by X-Ray Diffraction (XRD) analysis for phase identify analysis. The partitioning of radionuclide was predicted by calculation from the results of ash analysis.

RESULTS AND DISCUSSION

Characterization of Spent Resins and Glass Cullet Powder

The spent resins obtained from the TRIGA PUSPATI (RTP) reactor in Nuclear Malaysia contained 35 wt. % of moisture content, 50 wt. % of volatile matter, 13 wt. % of solid charred residue and 2 wt. % of ash, as shown in **Figure 1**. The elemental contents of spent resins and glass cullet powder were analysed using an Oxford INCA 400 X-Ray Energy Dispersive Spectrometer (EDS) system, as shown in **Figure 2** and **Figure 3**, respectively. The spent resins contained 80% of carbon, 7% of oxygen, 6% of hydrogen, 4% of sulphur and 2% of nitrogen. the flue gas of the spent resins will contain carbon monoxide, carbon dioxide, nitrogen oxides and sulphur dioxide. While, the glass cullet powder contained carbon, oxygen, sodium, magnesium, aluminium, silica, sulphur and calcium as shows in **Figure 3**. The activity of radionuclide concentration in the spent resins generated from RTP was detected in **Table 1**. The average calorific value of the spent resin is 17,530 J/g.

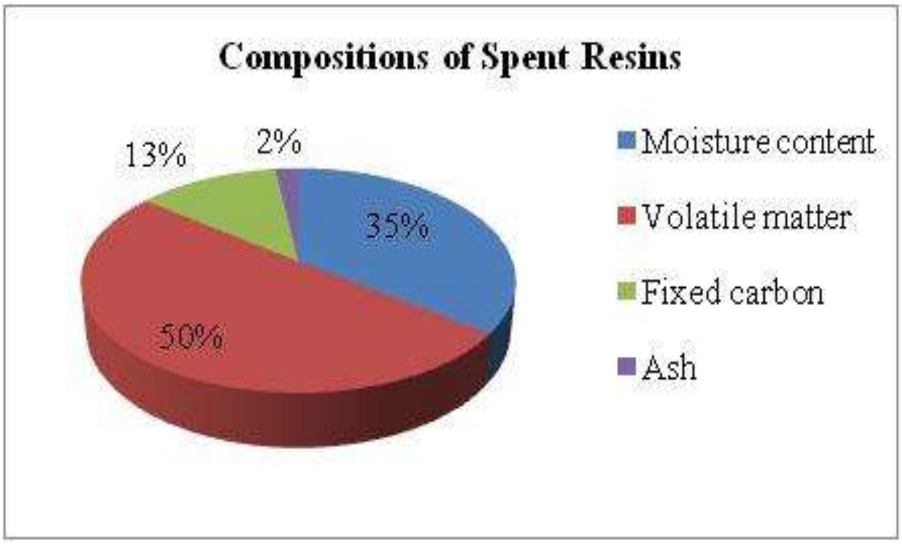


Figure 1: Proximate Analysis of Spent Resins

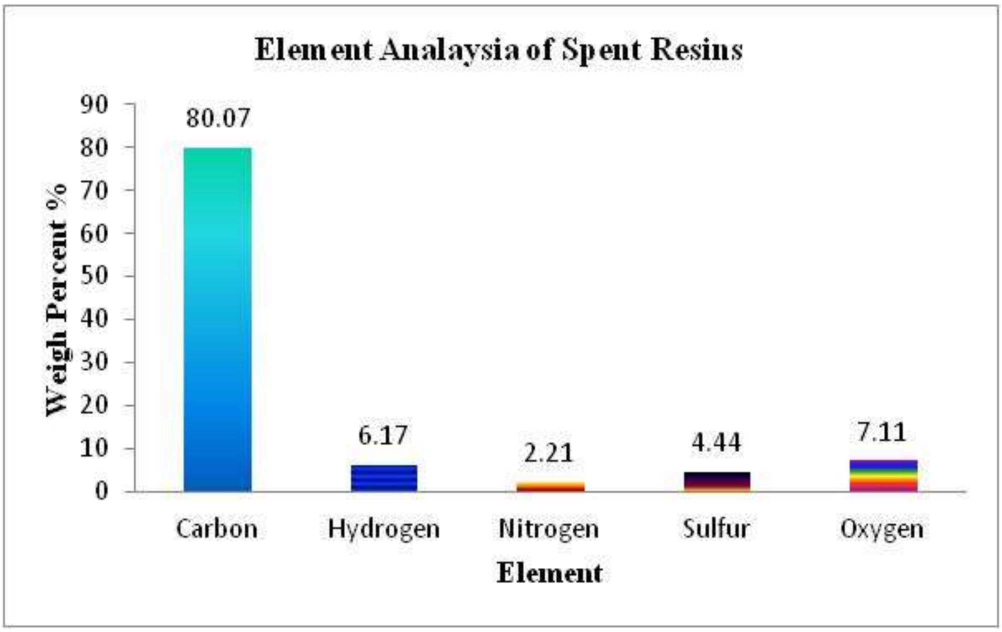
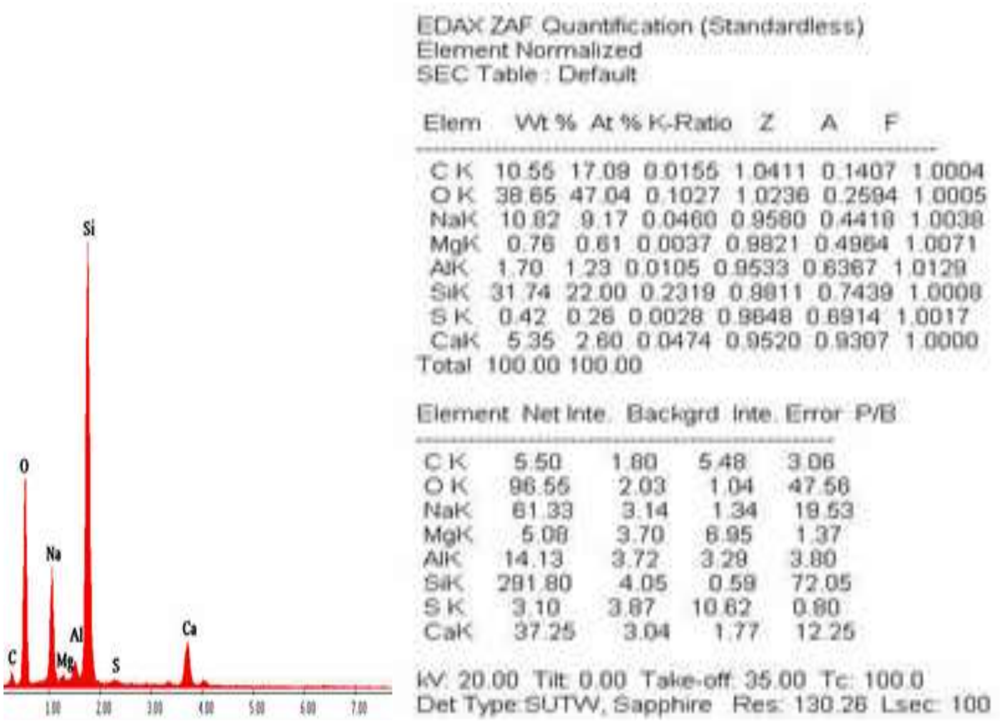


Figure 2: Elemental Analysis of Spent Resins



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EDAX ZAF Quantification (Standardless)
Element Normalized
SEC Table : Default

 Elem    Wt %  At % K-Ratio   Z      A      F
-----------------------------------------------
  C K   10.55 17.09 0.0155 1.0411 0.1407 1.0004
  O K   38.65 47.04 0.1027 1.0236 0.2594 1.0005
  NaK   10.82  9.17 0.0460 0.9580 0.4418 1.0038
  MgK    0.76  0.61 0.0037 0.9821 0.4964 1.0071
  AlK    1.70  1.23 0.0105 0.9533 0.6367 1.0129
  SiK   31.74 22.00 0.2319 0.9811 0.7439 1.0008
  S K    0.42  0.26 0.0028 0.9848 0.6914 1.0017
  CaK    5.35  2.60 0.0474 0.9520 0.9307 1.0000
Total  100.00 100.00

Element  Net Inte.  Backgrd  Inte. Error  P/B
-----------------------------------------------
 C K       5.50      1.80      5.48     3.06
 O K      96.55      2.03      1.04    47.56
 NaK      61.33      3.14      1.34    19.53
 MgK       5.08      3.70      6.95     1.37
 AlK      14.13      3.72      3.29     3.80
 SiK     291.80      4.05      0.59    72.05
 S K       3.10      3.87     10.62     0.80
 CaK      37.25      3.04      1.77    12.25

kV: 20.00  Tilt: 0.00  Take-off: 35.00  Tc: 100.0
Det Type:SUTW, Sapphire   Res: 130.26  Lsec: 100
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Figure 3: Elemental Analysis of Glass Cullet

The results from gamma spectrometer analysis revealed that only activation products (^{54}Mn, ^{65}Zn and ^{60}Co) were detected in the wet spent resins. The confirmation of the presence of these radionuclides in the samples were carefully made, based on the detection of the most abundant and of other supporting major peaks, of half-life and the correction due to the interference of other radionuclides. The highest activity concentration value was found to be ^{60}Co (7859 ± 125Bq/Kg) and the lowest was ^{65}Zn (689 ± 0.45 Bq/Kg). There was no radionuclide present in glass cullet powder.

Table 1: Activity Concentration of Radionuclide in Spent Resins

Radionuclide	^{54}Mn	^{60}Co	^{65}Zn
Concentration Bq/Kg	5814 ± 153	7859 ± 125	689 ± 0.45
Half life	312.5day	5.27year	243.9day

The purpose of the leaching test is to determine the release rates of the nuclides into the water empirically. The results from such analysis might affect the assessment of methods for final disposal. From the radionuclide leaching analysis ^{60}Co was found in the leachate with activity concentration as presented in **Table 2.** These test conditions of the compliance test are significantly harsher than those prevailing in-situ in the environment.

The radionuclide that attached to a sulphuric group has been replaced by H^+ and thus produced a –SO_3H group. The pH results were slightly acidic with average pH value of 5.02 ± 0.08. This result was expected because the resins used at RTP were of a strongly acidic gel type. the resins generally contain bound acid functional group (-SO^3 H) that is highly electronegative and the H^+ of –SO^3 H is a counter ion that making it more electropositive and thus, enables the resins to behave like a strong acid. The pH measurement was also useful for the regeneration of the spent resins, because the effectiveness of the acid used for regeneration depends on the initial pH of the spent resins.

Table 2: The Activity Concentration of ^{60}Co in Leachate.

Run	Activity Concentration of ^{60}Co in Leachate, Bq/mL	
	6 Hours Agitation	18 Hours Agitation
1	1.51 ± 0.21	1.52 ± 0.27
2	1.56 ± 0.33	1.88 ± 0.38

The TGA-DTA analyses of the spent resins detected a slow endothermic drop within the temperature range. The fall was due to the evaporation of water absorbed in the samples. The decomposition temperature of the spent resins was determined to be at 440°C. The electrical conductivity of spent resins leachate as a function of time is showed in **Figure 4**. Conductivity means that the spent resins were capable to absorb the ions contained in distilled water. The conductivity decreased within 60 minutes was mainly due to demineralization of spent resins leachate. The conductivity shows a little tendency to stable after 5 h. This indicates that there was some degree of interaction occurred and small amount of ions were released into the solution. The conductivity results for spent resins leachate was 86.2 Con.

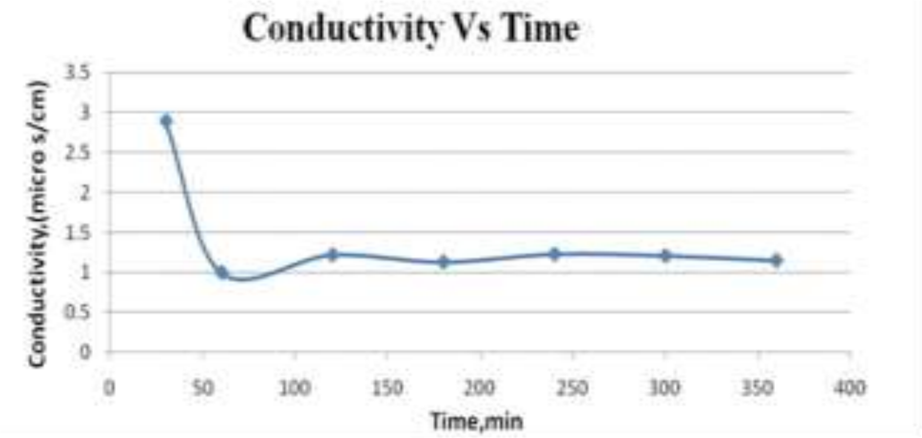

Figure 4: Conductivity of Spent Resins Leachate

Combustion of Spent Resins

During the combustion of spent resins, the off-gases were scrubbed using distilled water. The ash arising from the Lab Scale Combustor is a loose powder, partially soluble in water and acids, and thus must be immobilized [1]. The amount and the nature of the ash will strongly depend on the resins combustion. **Table 3** present the percentage of mass reduction as a function of the combustion temperature. The mass loss at 500°C attains 38% and the resin shows decomposition into carbon monoxide and carbon dioxide gasses [9].

Table 3: Percentage of Mass Reduction

Temperature (°C)	100	200	300	400	500
Mass loss (%)	7.23	14.64	25.46	26.47	38.91

The concentration of the flue gases depends on the operating temperature and the residence time in the chamber. **Figure 5** shows the distribution concentration of the flue gases during the combustion at different temperatures. The experiment showed the carbon dioxide was released at temperature of 200°C and 300°C. Carbon monoxide is formed by the incomplete combustion of materials containing carbon at low temperature. However, at a higher temperature of 400°C carbon monoxide was released along with nitrogen oxide. Nitrogen oxides were formed through the oxidation of diatomic nitrogen in high temperature. The nitrogen oxides were not released at temperature 200°C and 300°C because the temperature was not high enough to release nitrogen oxides. At the temperature 500°C, the emissions were releases 40ppm of carbon dioxide, 69ppm of nitrogen oxides and 43ppm of sulphur dioxide. At this temperature, carbon-14 might have combined with oxygen to form carbon dioxide. The behaviour of the combustion shows the entire element has been converted into gaseous state and emitted as the flue gas at 500°C.

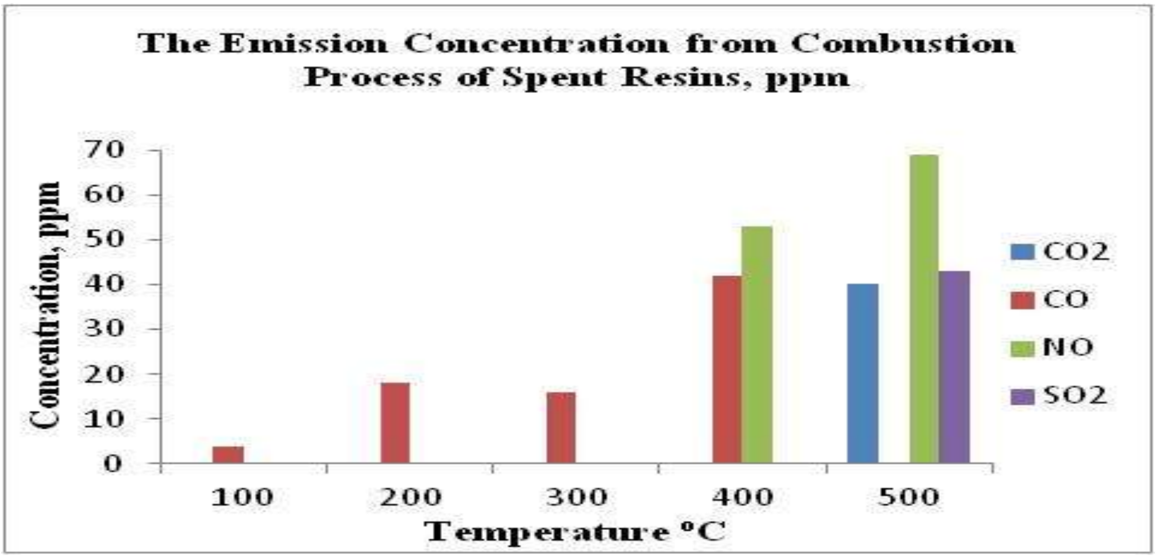

Figure 5: The concentration of Flue Gases during Combustion

The activity of radionuclides in the ashes was determined using gamma-spectrometry and the results are as shown in **Table 4.** The radionuclides detected in the ashes were Manganese-54 (^{54}Mn) and Cobalt-60 (^{60}Co). Four samples of ashes were obtained from the combustion of spent resins at 200°C, 300°C, 400°C and 500°C and analysed. The radionuclide activity concentration is increasing with temperature. The trend can be seen in **Table 4**. At 500 ° C, the concentration of ^{54}Mn and ^{60}Co yields the highest reading. This trend is explained by the thermal decomposition of resin.

Table 4: The Activity of Radionuclide Concentration in Ashes at Different Temperature

Temperature (°C)	Activity Concentration of Radionuclide, Bq/Kg	
	^{54}Mn	^{60}Co
200	9411 ± 243	12637± 201
300	11209 ± 288	15026± 239
400	13018 ± 532	17588 ± 375
500	33048 ± 1344	43287 ± 923

Table 5 presents the results obtained from the radionuclide-leaching test for ash. The results show that the quantities of constituents leaching from the ashes were very small. Most of these were below the detection limit. The leaching concentration value of ^{60}Co at 500°C was higher than at 200°C. However, this ^{60}Co could not be detected when the ashes were encapsulated with glass material. Thus it can be concluded that combustion alone cannot be the final treatment for spent ion-exchange resins. Further treatment such as vitrification should be carried out to make sure that the radionuclide does not leach out.

Table 5: The Activity of Radionuclide Concentration in Leaching Analysis

Temperature (°C)	Concentration of ^{60}Co in Leachate, Bq/mL
200	4.75 ± 0.27
300	2.89 ± 0.24
400	7.56 ± 0.33
500	8.11 ± 0.35

Ash Vitrification

Vitrification is one of the immobilization processes. The objective of immobilization is to convert radioactive waste into a stable monolithic form, which minimizes the probability of radionuclide release to the environment during interim storage, transportation and final disposal. In this study, the ashes from combustion were vitrified in a High Temperature Furnace at temperatures of 700°C, 800°C, 900°C and 1000°C. The ashes were incorporated in the glass cullet powder. The different volume ratio was set at 1:9, 2:8 and 3:7. The effect of mixing ratio was observed in that the volume of glass was about five times smaller than the initial volume of spent resins. **Figure 6** evidences the mass reduction obtained at optimum conditions of temperature and mixing ratio [9]. The oxidation of radionuclide may help in reducign the Reduction/Oxidation (Redox) state of the glass during vitrification. The volatilization of radionuclide from the ash gives the weight loss in the slag and the decreasing concentration of radionuclide. The percentage of mass reduction increases with the temperature.

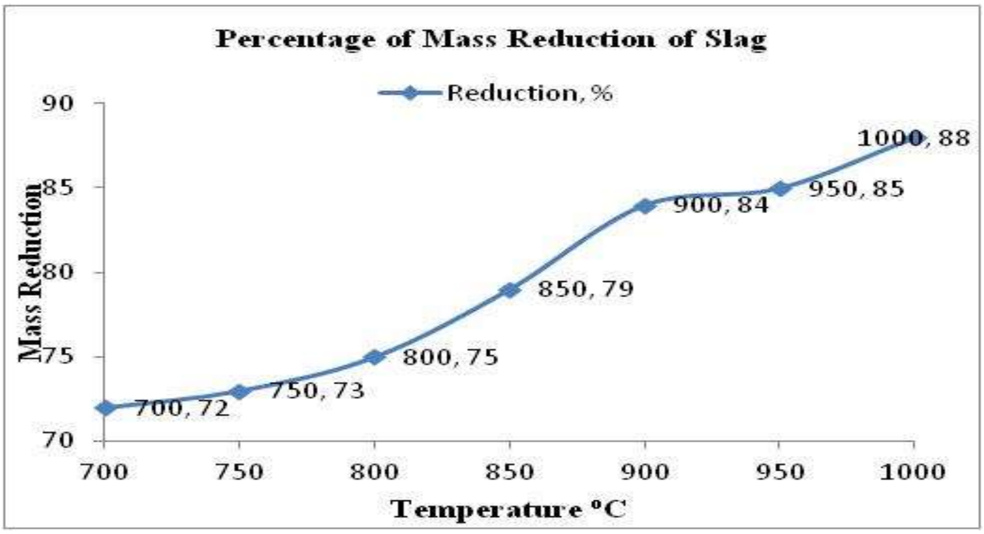

Figure 6: The Percentage of Mass Reduction

The correlation between the concentration of radioactivity and temperature is shown in **Figure 7.** The results were obtained from the activity concentration of radionuclide in slag. The resulting values significantly decrease when the vitrification temperature increases. The radionuclide present in the slag was ^{60}Co. The optimum conditioning temperature was revealed to be 900°C and the preferred volume ratio of ash to glass cullet powder was 1:9.

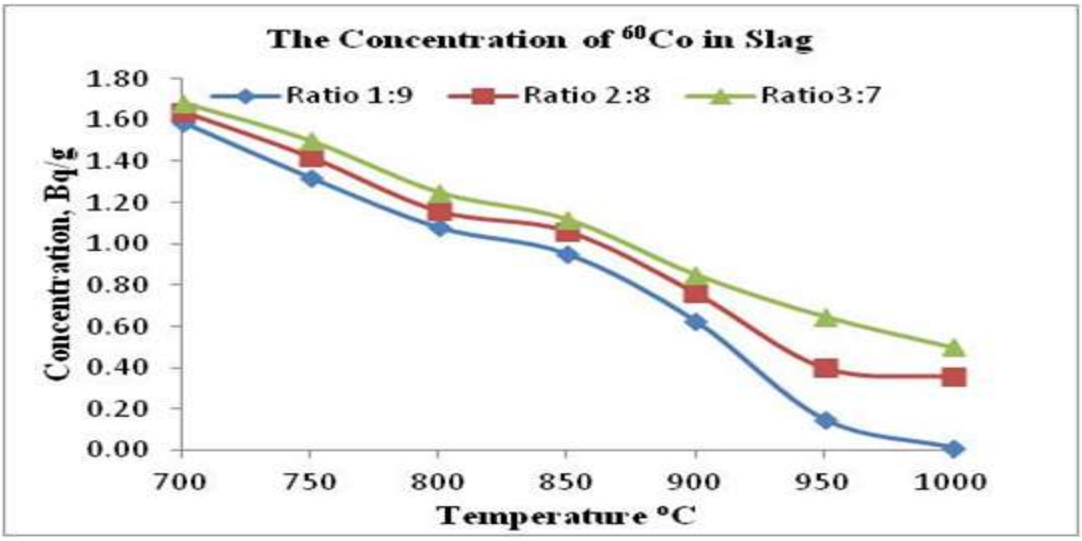

Figure 7: Radioactivity of ^{60}Co in Slag

The identity of the slag phases from vitrification was determined by X-ray diffraction (XRD) analysis using a Model X'Pert Pro Diffractometer system. The scanning range was selected from 10° to 80°and a graphite monochromator and CuKα radiation were used. The collected data were refined using X'Pert Highscore software. The optimum degree of chemical durability of slag was observed for volume ratio 1:9 (ash to glass cullet powder) at 900°C. The XRD pattern suggests that this slag material was amorphous with only minor amounts of crystalline phases. Along with the amorphous phase of vitrified ash crystalline inclusions such as SiO_2 in cristobalite form and quartz are present in the sample vitrified at 900°C with a volume ratio 1:9 of ash to glass cullet powder (**Figure 8**). **Figure 8** shows the XRD analysis at 900ºC for Ratio 1:9 (S7), 2:8 (S8) and 3:7 (S9). The sharp peaks of cristobalite and quartz were more prominent in S7 compared to S8 and S9. The result indicates that the presence of glass cullet powder lowered the activation energy of the reaction system and enhanced the reaction rate of flux material to form durable slag material. Leaching of nuclear waste form is a complex process be affected by many factors, relating to the leaching system, i.e. either the leaching agent or the solid being leached [1]. Leachability increases with rising temperature if the mechanism is based on diffusion. With ^{60}Co, the diffusivity generally increased with rising temperature.

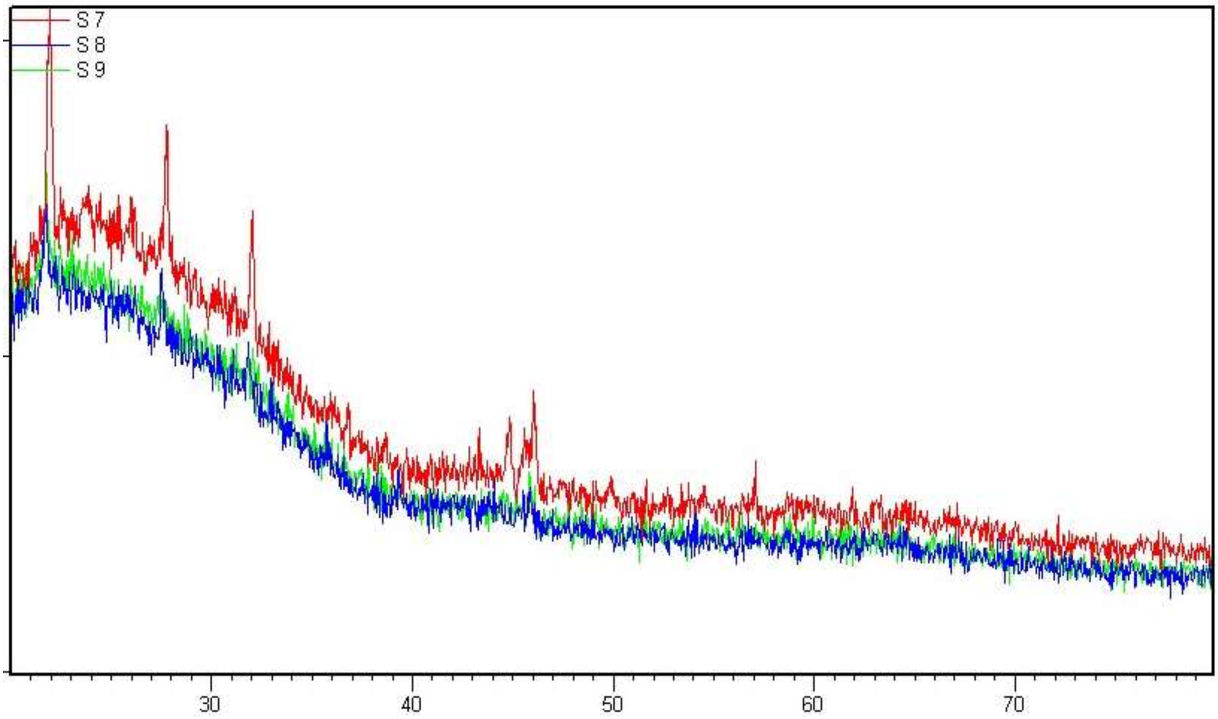

Figure 8: The XRD analysis at 900ºC Ratio 1:9

Table 6: The Leaching Concentration of Radionuclide after Vitrification of Ash Spent Resins

Temperature (°C)	Sample Ratio	Concentration of ^{60}Co in Leachate , Bq/mL
700	1:9	0.0042 $\pm$ 0.00079
	2:8	0.0069 $\pm$ 0.00085
	3:7	0.0082 $\pm$ 0.00085
800	1:9	0.0052 $\pm$ 0.00060
	2:8	0.0060 $\pm$ 0.006
	3:7	0.0032 $\pm$ 0.00079
900	1:9	Minimum Detectable Activity
	2:8	0.00055 $\pm$ 0.00077
	3:7	0.00058 $\pm$ 0.00072
1000	1:9	Minimum Detectable Activity
	2:8	Minimum Detectable Activity
	3:7	Minimum Detectable Activity

The leaching data are presented in **Table 6** for the glass form. The release of ^{60}Co from the slag shows that at 900ºC the concentration of radionuclide was minimum detectable activity that means no leachability of radionuclide at that temperature. The pH of leachate from slag formed at 700º, 800ºC, 900ºC and 1000ºC varied between 6.20 to 6.50.

CONCLUSIONS

The decision to reduce the volume of spent resins by incineration or other pre-treatment prior to immobilization depends on the precise requirements of the final repository. This study shows the effect of adding glass cullet powder (derived from clear glass bottles) as a matrix material that can be used as a flux to immobilize the radionuclides contained in the ash from spent resins, arising from the TRIGA Nuclear Malaysia reactor. Combustion is required prior to immobilization to form ash and glass, and the off-gas releases during vitrification are of concern. The optimum conditioning method of silicate glass were shown to be a combination of the maximum tested temperature of 900°C, the heating rate of furnace was setting at 20°C/min, and the resulting ash to be incorporated with glass at the maximum tested volume ratio of 1:9 (ash to glass cullet powder) in 45 minutes, including a cooling phase. The slag material was amorphous with minor amounts of crystalline phases.

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