# WASTE SOLIDIFICATION USING CRT GLASS FOR

# RETARDATION OF HAZARDOUS ELEMENT IN

# SPENT RESIN

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**Abstract**

Spent ion exchange resins (spent resins) were generated from TRIGA PUSPATI Reactor (RTP). The RTP produces approximately 50 kg per year of spent resins. Thus, a treatment method is needed due to continuous generation of spent resins and limited storage capacity. According to IAEA, immobilisation is one of the effective ways to overcome this problem. Thus, in this research, vitrification technology is applied. Spent resins were mixed with glass to produce glass waste form. From the result, the optimum composition of spent resins mixed with CRT glass were between 10-20%. Then, the waste glass form was tested with XRD to identify crystal in the sample. The waste glass form is amorphous phase. For chemical durability, product consistency test (PCT) had been conducted. The normalized released for B, Na and Si is in range of 0.001 - 0.006 g/m2, 0.010 - 0.037 g/m2, and 0.024 - 0.039 g/m2, respectively. For conclusion, despite minor difference of additive effect, temperature effect and waste loading, the optimized composition for the glass containing spent resin can accommodate up to 20% waste loading with tin slag as additive and melted at 1,200°C.

## INTRODUCTION

The development of the nuclear industry has been started in 1985 and becomes skyrocketing over the centuries, which causes more and more radioactive waste has been produced. In the nuclear industry, spent ion exchange resins (spent resins) contribute to the significant proportions of solid radioactive wastes. According to Atomic Energy Licensing Act (Act 304) in Malaysia [1], spent resins have been classified as low-level radioactive waste, and intermediate-level radioactive wastes depend on the radioactivity level as loaded by Co-60, Cs-137, Sr-90, U-235. Thus, the treatment and disposal of the spent resins have become an urgent problem in developing nuclear power plants as spent resins are considered problematic waste [2].

In Malaysia, these spent resins are generated from PUSPATI TRIGA Reactor (RTP), a TRIGA MARK II. TRIGA stands for Training, Research, Isotope Production and General Atomic. It is used for training, research and isotope production. Generally, these ion exchange resins are used 10 -15 times in a nuclear reactor, depending on the exchange capacity and regeneration efficiency. A new ion exchange resin will replace the resin contaminated with radioactive [3]. Every year approximately 50 kg per year of spent resins [4]. A treatment and disposal method are needed due to the continuous generation of the spent resins and the limited storage capacity through immobilization [5].

Therefore, the proper precautions have to be taken to solidify the radioactive waste to protect the ecological environment and human health and ensure our future generation is not a burden.

## MATERIALS AND METHOD

### Materials

The spent resins were provided by WasTeC, which is one of the departments in Malaysia Nuclear Agency. WasTeC was responsible for collecting all radioactive waste produced from various applications in Malaysia for processing, treatment, and storage. The type of spent resins used in Malaysia RTP is Amberlite IRN 150. The cathode-ray tube (CRT) glass and steel slag were supplied by local industrial company. Microstructure characterization of both CRT glass and steel slag have been studied previously [6].

### Vitrification of spent resins

To remove the excess moisture in the sample, the spent resins was dried at 90°C in the laboratory oven for 24-48 hours until it achieved constant weight. The CRT and tin slag were crushed into small pieces and mixed with spent resins in desired composition shown in Table 1. Then, 20 g of mixed composition was put in the 50 mL alumina crucible with lid in high temperature furnace. The mixed composition was melted at 1,150°C and 1,200°C for 1-2 hrs and let to cooled down until ambient temperature. The solidified glass sample was crushed for further analyses.

TABLE 1. SAMPLE COMPOSITIONS

|  |  |  |  |
| --- | --- | --- | --- |
| Item | Spent resins (%) | CRT glass (%) | Steel slag (%) |
| Sample 1 | 10 | 75 | 15 |
| Sample 2 | 15 | 75 | 10 |
| Sample 3 | 15 | 85 | - |
| Sample 4 | 20 | 80 | - |

### Physical evaluation

The sample was observed after melting through naked eyes. Subsequently, the sample was 500 um sieved prior for x-ray diffraction (XRD) analysis. The sample was analyzed in Centre for Research and Instrumentation Management (CRIM), Universiti Kebangsaan Malaysia, using XRD instrument (model Bruker D8 Advanced) for presence of crystal phase during glass fabrication. The spectrum obtained was compared with International Center for Diffraction Data (ICDD) for identification of crystal phase in the sample.

### Chemical evaluation

For durability testing, the samples were prepared and measured using the product consistency test (PCT) designated ASTM C1285-14 [7]. The vitrified spent resins were crushed and sieved to the particle size of 100 - 200 mesh based on the procedure. The sieved vitrified was ultrasonically washed with deionized water three times. Then, the deionized water was replaced with absolute ethanol, and the material was rewashed three times. The samples were then dried in the oven overnight at 90 ± 10°C. The glasses were retrieved and placed into Teflon vessels filled with deionized water for a recommended ratio of the volume of solution-to-mass of solid of 10 ± 0.5 mL. The test was repeated in duplicate for each sample. Blanks also were prepared with deionized water at a leachate volume similar to that of the sample. All samples and blanks were placed together in an oven at 90 ± 2°C for 7 days.

After the leaching period, the samples were retrieved and cooled to room temperature. The leachates were removed from the vessels, pH was measured and recorded, and remaining solutions were filtered with a 0.45 um syringe filter and stored in a clean container. Nitric acid (1% from the leachate volume) was spiked into the filtered leachate to prevent possible hydrolysis of heavy cations. The solutions were analyzed using inductively coupled plasma-mass spectrometry (ICP-MS) for glass components. Normalized releases of elements were then calculated using equation (1) as reported previously [8],

*NC = C/F* (1)

where *NC* is normalized releases for elements, g/m2; *C* is the concentration of elements in solution, g/m2, and *F* is the fractional weight of elements in glass.

## RESULTS AND DISCUSSION

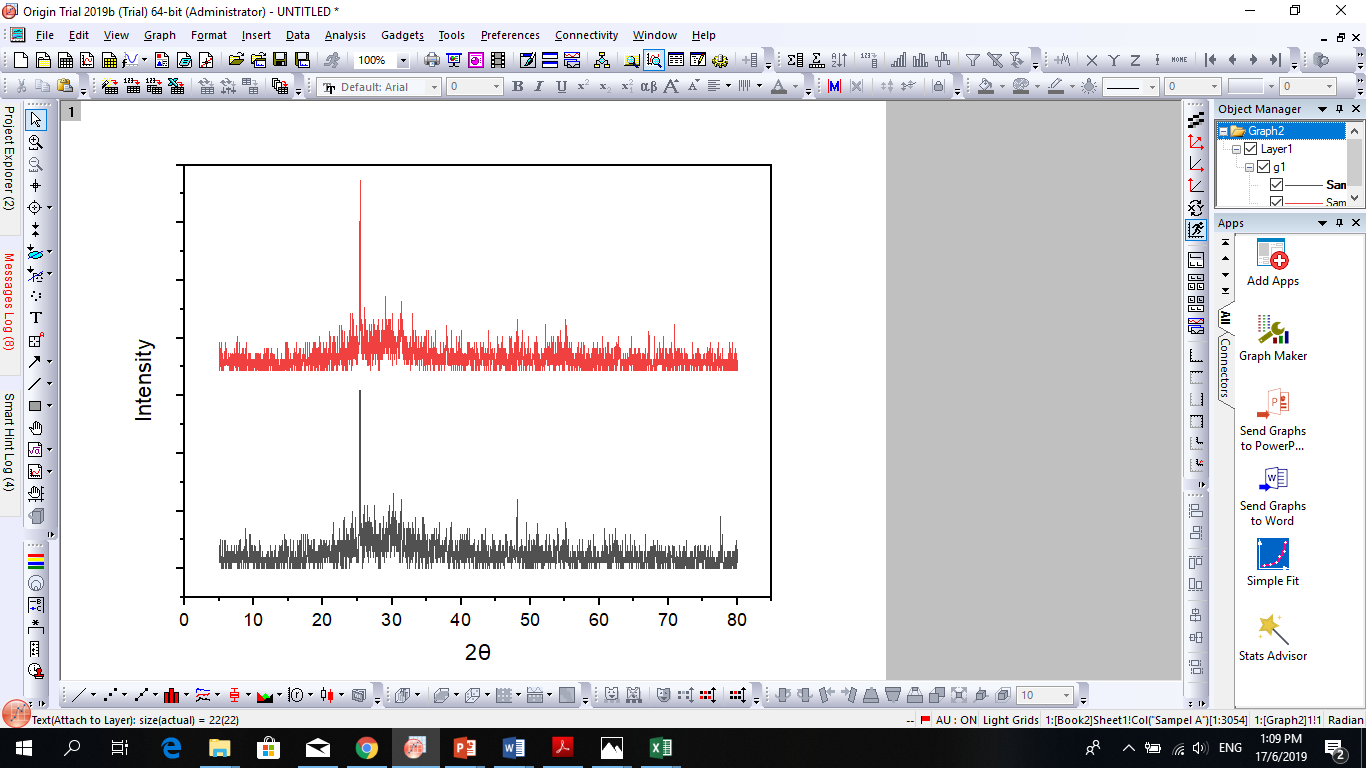
### Physical evaluation

The observation after melting process was done and summarized in Table 2. Based on International Atomic Energy Agency (IAEA) [9], the maximum temperature recommended for spent resin vitrification is around 1,200°C, due to volatile radionuclide beyond 1,200°C such as Cs.

TABLE 2 GLASS WASTE FORM OBSERVATION

|  |  |  |
| --- | --- | --- |
| Item | 1,150°C | 1,200°C |
| Sample 1 |  |  |
|  | The glass formed look shiny and intact. There are few bubbles that be seen inside glass structure. | |
| Sample 2 | https://lh6.googleusercontent.com/IUu5MVoKOO4CyhNlLgKEcI9OTqBdnvvy-VkcLXsuTds1owwd9WD2CL3UB-C-JeGIjdRKajiw4-hTw64KxMExntzCL1KX-RXaGX5cvrPZGUHAurVNzQzR99q3oyLyYpnM_UnFmV0SXRU82wHX9Q |  |
|  | The glass formed look shiny and intact. Small bubbles were seen inside the glass structure. | |
| Sample 3 | https://lh6.googleusercontent.com/D79tDzrnk3e_F67FZblmUkdL5WVYF1pHw8w4acSrqlNqN1D6WG6YOzNN28kVbUL3JQfGngm_KbvnsNGUk-8_B_tXicTl_wAibWsMm3WH4qZ72Vt6b90_lglbZCsySZ6JzuUTKwtBQMpCuC8Ptw |  |
|  | There are few undissolved powders that can be seen inside the glass | The glass formed is intact and shining with small bubbles observed inside the glass. |
| Sample 4 | https://lh4.googleusercontent.com/8XvmihOOP2pSD0cJlhFKSzyc0-vu7jeRiNMU_Yy-SD_19xoidDOhWMfpmDzXkIHP8O1yTHqSsrfn9usiCmo4E0MWUf_eo2-Vw72WLGou9PNlwK5XHwfSuEYY32dxKf6bRs3vu09aj3_XjGKuDw | https://lh3.googleusercontent.com/yndeWpnT9xE9pqLhzqO2e_LAyJdzljLFbs8-q7PMCKRFcPmGhLLiN5XRCR-t_gqAMZd7nDf0wtM4XLnHpB3dHoHexvXRg6zVAGb-vnXEjiycEahvNJwaPMGtvU9-QfILDrRl79fQIe32vnjcgTtZ6FboudbmlCjLVAAbFGRNpw0eSVVe3nmDyjY3lg66i9JFLjqqw65a8sKFi0S3ON6reSGrpgL7RLB6wKizcZp6eyxVe7cbdNXzb7RjYbgwErUuA-tUjtaxbjXBt81srWO0VN2_3HS4LC84P08SVtDNpihGKBqXQ7CIbCBpI_JYHyH9iI09eTBsyaD8PMvQloVFFnan3lc9VAHRzqM8N3fMI5Box04ovkYxVwEbEqEB4_VI1EyP3r3Abbmea34CftsPG4H3PFmbq-kcoqiuyeM0mL3UtXfuALiwXMsJxXsiUt7lWXAxFg-somGWsSpKA4kl2z0wCi95hYG7J4Inac6SmJ-TSQN19N-iO1x1KQ3cjy98zsMwDxcie-9YZzjKs0vsT8A8uMnEsm1oU24m_T6VDVcx59-VVETmHRJd-Kw7Dn9StEiSpBnxW2tVCyu4DSFCQk_f82_DHhLAe-AjIvHL6HLGxvD3A44uL-uXokGMJULaVpv9_v0PW6camPsJifPtObMYRbEWHCmJ=w493-h657-no |
|  | There are more undissolved powders that can be seen, the glass is not intact nicely. | The glass formed is intact with small bubbles observed inside the glass. |

Based on XRD analysis, several samples were compared to see the difference and effect of tin slag as additive, effect of melting temperature and also effect of waste loading which shown in Figure 1, Figure 2 and Figure 3, respectively. The analysis was carried out by incorporated standard anatase (TiO2) in the glass powder for comparison with visible peak, if observed. Based on the figures, the effect is minor to be compared by physical evaluation.



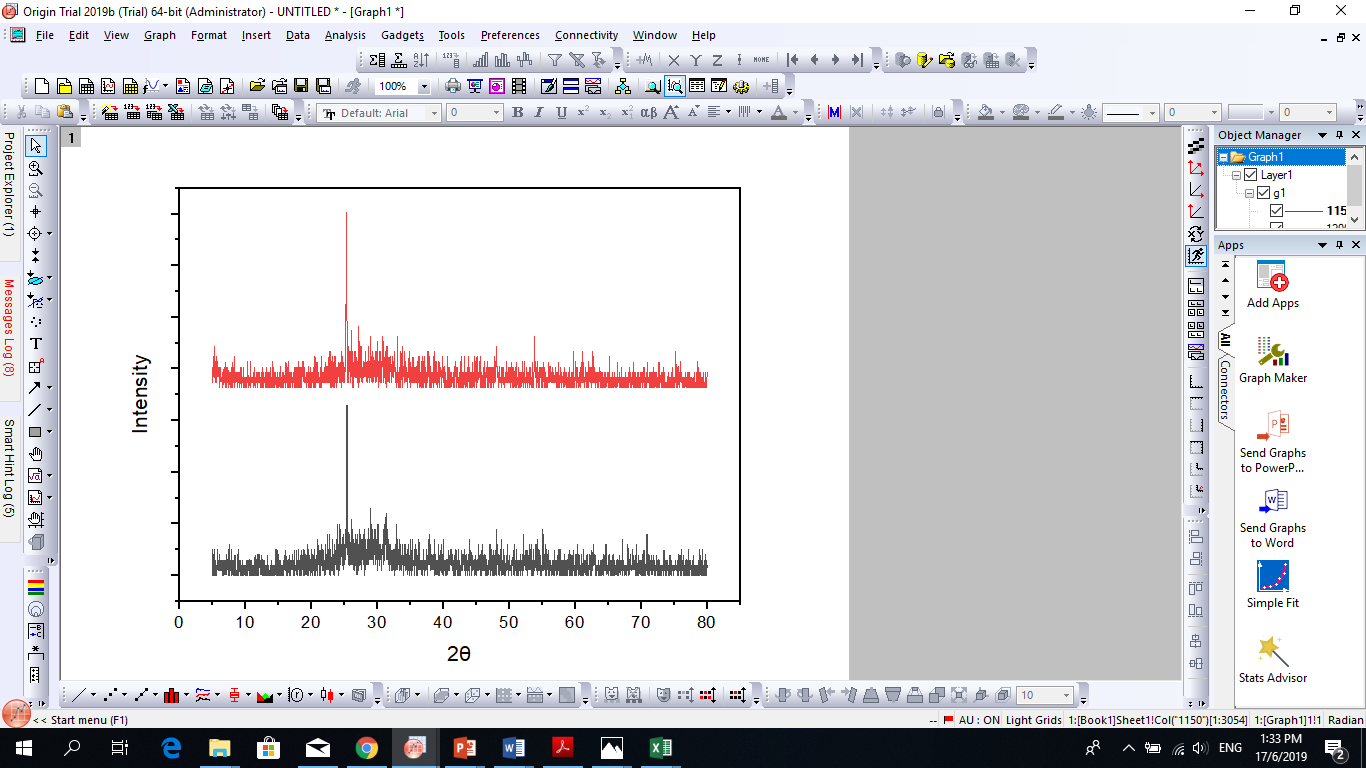
Sample 3

1,200°C

Sample 2

1,200°C

FIGURE 1. EFFECT OF TIN SLAG IN THE COMPOSITION AT SIMILAR TEMPERATURE



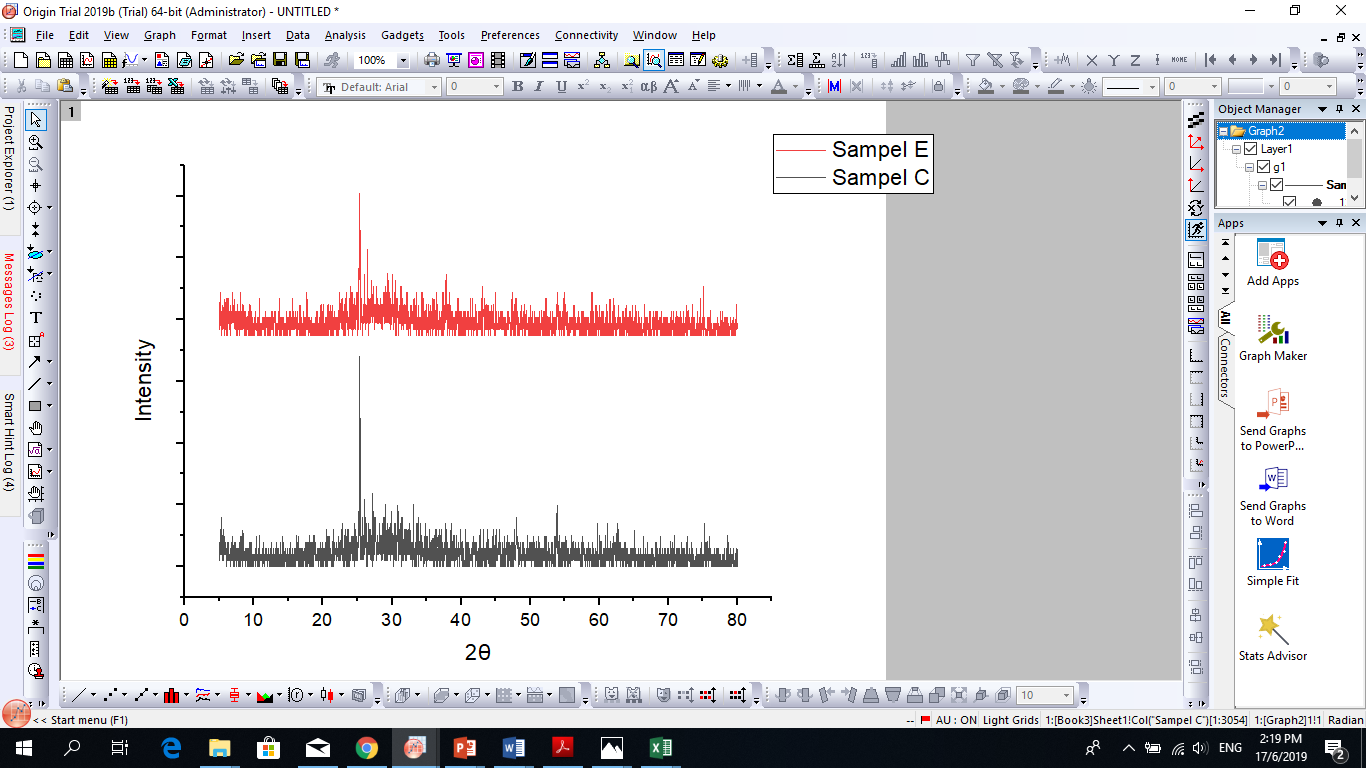
Sample 2

1,150°C

Sample 2

1,200°C

FIGURE 2. EFFECT OF TEMPERATURE FOR SIMILAR SAMPLE COMPOSITION



Sample 1

1,150°C

Sample 2

1,150°C

FIGURE 3. EFFECT OF WASTE LOADING FOR SAMPLE AT SIMILAR TEMPERATURE

### Chemical evaluation

The pH and total of normalize released for B, Si and Na from vitrified spent resins were assessed by PCT test [7]. Chemical evaluation involved the glass reaction in the deionized solvent. The process involved in glass dissolution includes ion exchange, water diffusion, hydrolysis and precipitation. The pH before the assessment is 7 and had been increased after 7 days. Similar comparison was done for chemical evaluation to see the effect of tin slag as additive, effect of melting temperature and effect of waste loading which summarized in Table 3, Table 4 and Table 5, respectively.

Based on Table 3, the effect of additional tin slag might lower the release of glass components in solution and it shows the effect of additive is prominent. Although the difference in Table 4 is not significant, the glass waste-formed at temperature 1,200°C release more Si and B but less Na than the same composition at 1,150°C. This was also reflected to higher pH of solution for Sample 2 melted at 1,150°C (pH 8.2) which might affect the durability of glass. On the other hand, increasing waste loading showed less of B and Na releases but more Si release. Thus, the optimized composition for the glass containing spent resin can accommodate up to 20% waste loading with tin slag as additive and melted at 1,200°C.

TABLE 3. EFFECT OF TIN SLAG IN THE COMPOSITION AT SIMILAR TEMPERATURE

|  |  |  |
| --- | --- | --- |
| Elements | Normalized release (g/m2) | |
| Sample 2  1,200°C | Sample 3  1,200°C |
| B | 0.002 | 0.006 |
| Si | 0.035 | 0.039 |
| Na | 0.010 | 0.019 |

TABLE 4. EFFECT OF MELTING TEMPERATURE FOR SIMILAR SAMPLE COMPOSITION

|  |  |  |
| --- | --- | --- |
| Elements | Normalized release (g/m2) | |
| Sample 2  1,150°C | Sample 2  1,200°C |
| B | 0.001 | 0.002 |
| Si | 0.031 | 0.035 |
| Na | 0.021 | 0.010 |

TABLE 5. EFFECT OF WASTE LOADING FOR SAMPLE AT SIMILAR TEMPERATURE

|  |  |  |
| --- | --- | --- |
| Elements | Normalized release (g/m2) | |
| Sample 1  1,150°C | Sample 2  1,150°C |
| B | 0.005 | 0.001 |
| Si | 0.024 | 0.031 |
| Na | 0.037 | 0.021 |

## CONCLUSION

The physical and chemical evaluation of spent resins in the CRT glass as host showed that the characteristics of good glass can be achieved without no phase separation or presence of crystals with minor difference of glass components release in the solution; thus, we can conclude that the durability of glass is good.

ACKNOWLEDGEMENTS

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