

## DEHYDRATION OF DECOMMISSIONING GELS FOR WASTE REDUCTION

G. Harvel<sup>1</sup>, V. Senka<sup>1</sup>, R. Khurmi<sup>1</sup>, Y. Aoki<sup>2</sup> and T. Sunagawa<sup>2</sup>

<sup>1</sup> Ontario Tech University, Oshawa, Ontario, Canada

<sup>2</sup> Fukui University of Technology, Fukui, Japan

[Glenn.Harvel@ontariotechu.ca](mailto:Glenn.Harvel@ontariotechu.ca)

### Abstract

One method of decontamination or capture of loose contaminants during the decommissioning process is the use of gels. Gels have some advantages over water based solutions in that spills are much easier to clean up while a water based solution may penetrate the concrete or soil and spread contaminants further. One disadvantage of gels is that they include other constituents that increase the overall waste volume. Thus, while they can do an excellent job of capturing and securing loose contaminants, they will increase the total volume shipped to a waste site for disposal. Regardless of the hydrocarbon compound used in the gel, there is usually also some water present. In this work, we examine two methods for the reduction of the gel volume by eliminating the water content. For gels that use water as part of their creation, this can result in waste mass reduction by greater than 80% and possible volume reduction greater than 85%. The first method assessed is natural evaporation processes and the second method assessed is a distillation process. The first process is much slower but is more reliable in ensuring that the water is evaporated from the gel leaving a dry puck for easy disposal. The second process is able to obtain higher volume reductions faster and is also able to breakdown some of the hydrocarbons releasing other non-contaminants however the second method is more challenging to ensure there is no carry-over of the waste product. The paper will discuss the nature of the gels tested, both techniques, and the volume reduction results obtained to date.

### 1. Introduction

During decommissioning activities there will be the generation of aerosols, particulates, and loose contamination. These forms are secondary contamination that can be spread throughout the environment. While ultimately there is an environmental impact concern, the more immediate concerns are related to worker dose and the increase in waste volume sent to either a Low Level Waste site or an Intermediate Level Waste site dependent upon the waste characterization. Methods to reduce the generated secondary waste need to be developed.

There are several methods that already exist such as steam cleaning or washing which can result in contaminated water or contaminated surfaces. Other methods such as dry ice or sand blast can result in a particulate form that would need clean-up.

It is important to facilitate efficient and cost-effective storage, and some desirable qualities for a liquid radwaste storage solution include transforming the waste into an insoluble and compacted solid [1]. One method to achieve this is through vitrification. This process involves transforming the waste into a glass, generally borosilicate, in order to enable the long-term storage of an insoluble solid with a relatively small volume increase [2]. In order to do this, the waste must be heated and mixed with glass-forming chemicals at around 1000°C, where the mixture is poured into a vessel which is welded shut and the mixture is left to cool and cure [2]. Once that glass cools, it forms a solid matrix which immobilizes any potentially mobile nuclides in whichever waste forms are contained in the glass for

several thousands of years [2]. Vitrification is currently in use in France, Japan, Russia, United Kingdom, and the USA, albeit it is currently used exclusively for separated high-level waste which arises from reprocessing [1].

Another solution involves converting the liquid into some sort of gel-like or highly viscous material. This highly viscous compound will then be stored in some sort of container either made of a metal or polymer. This design potentially can solve the following issues:

- 1) Keep the liquid more contained and prevent splashing and spreading of contaminated liquids;
- 2) Easier to visually detect leakage from the containers as compared to a liquid which may dissipate or penetrate the floor;
- 3) Easier clean-up in accident scenarios (i.e. spills); and
- 4) Potential reduction in overall volume compared to a solid solution.

There are a large number of types of gels available depending on the purpose or application. Some gels can be used to hold a chemical reaction so that it can detect radiation [4,5]. Others are based upon paste type materials which are discussed in another paper [6].

One type of gel (hydrogel) is the mixture of a polymer that contains many entangled chains that are super absorbent and can hold their weight in water hundreds and thousands of times in magnitude. Hydrogels are usually used in medicine and are currently being researched for tissue replacements. PHEMA (Poly (2-hydroxyethyl methacrylate)) is the main polymer used in medical hydrogels [7]. It usually requires ultraviolet light to obtain its form. Currently, PHEMA is used for contact lenses. Some commercial hydrogel polymers include Sodium Polyacrylate, a polymer that is currently used as a gardening additive to control the moisture soil by absorbing excess water. Other uses of Sodium Polyacrylate include the use in the primary absorbent layer of diapers and dewatering sludge [7,8].

Most hydrogel polymers are made of a Carboxylic acid that loses a Hydrogen creating a free negative ion at the ends of each monomer when introduced with water. The negative ions in a chain are the reason for the initial polymer to untangle. The Hydrogen in the water is then attached to the negative ion of each chain creating the gel due to the slight positive charge Hydrogen atoms have in water [7].

Another type of gel is a PolyVinyl Acetate (PVA) based gel. This type of gel uses a long polymer chain (PVA) and cross-linking to create a gel. The PVA gel can be adjusted to have different levels of viscosity by changing the constituent contents.

Hydrogels and PolyVinyl Acetate (PVA) based gels both appear to show promise for decommissioning related activities. In this work, hydrogels and a PVA based gel were assessed for their ability to be reduced in volume due to dehydration as one step in the waste packaging process.

## **2. Experimental Approach**

Two types of experiments are conducted in this work. The majority are dehydration experiments using natural processes for the removal of the water. The paper concentrates on these experimental results.

For comparison purposes, there is discussion related to distillation experiments where heating of the gel is used to change the evaporation rate of the water.

## 2.1 Dehydration Experiments

Two sets of dehydration experiments are performed. The first set of experiments are dehydration of the hydrogel. In these experiments, natural evaporation processes are studied where a gel is manufactured and hydrated with water and then evaporation rates are determined in both the open lab environment and in an enclosed environment chamber. The results are also compared to evaporation rates for water under similar conditions.

In the second set of experiments, natural evaporation is studied for the PVA type gel. The same process is used as for the hydrogel except the study is only performed in the environment chamber to eliminate wind effects (air motion in the lab).

The hydrogel chosen for the first set of experiments is Sodium Polyacrylate. This gel is extremely absorbent and is able to capture large volumes of liquids. Figure 1 shows a small amount of gel swelled with water and the same gel after several days of dehydration. The significant volume reduction is excellent but the rate is obviously high which may impact the handling of the gel during the decommissioning processes.

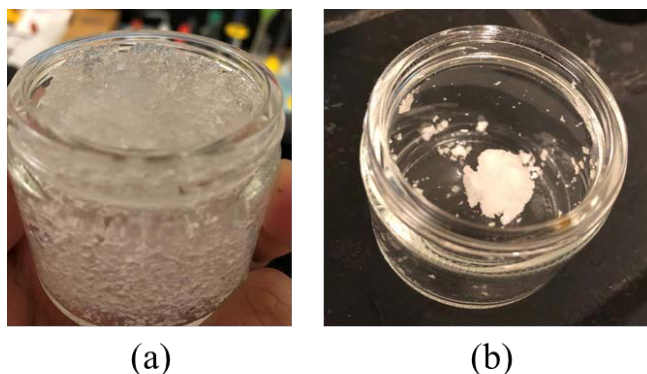


Figure 1 Sodium PolyAcrylate hydrogel (a) saturated with water, (b) dehydrated.

A simple experiment was performed to determine the evaporation rates using water as the absorbent. The gels were mixed in fixed quantities to a set volume of water. Samples were prepared as per Table 1. The intent was to observe the evaporation rate of the hydrogel. The reference sample is tap water and is expected to act as a reference since the majority of the hydrogel is water.

The first set of samples were tested in the open laboratory environment. There is a modest air flow exchanging the air keeping the humidity relatively low. The trends of the first set of samples are similar to all other sample sets but have higher evaporation rates for the open-air cases. This is likely due to the air flowing over the samples assisting the evaporation rate.

The second set of samples were tested in an environmental chamber which was sealed from the rest of the laboratory. The environmental chamber ensured there was no air flow rate affecting the samples. However, the net volume of free air is much smaller affecting the capacity for water transfer to the air phase.

Similarly, the PVA gel samples were prepared as per Table 2 and the same experimental process was followed as for the hydrogel. The PVA gel was studied in the environmental chamber for direct comparison to the hydrogel experiments.

The evaporation rate of water can be determined by many different models but generally depends upon the saturation vapour pressure, humidity, temperature, and the air flow rate above the water [9,10]. Additionally heating sources such as solar heating or fluid temperature also affect the evaporation rate [9,10]. For the dehydration experiments performed in this work, no heating sources are applied. Humidity is the most important factor in these experiments.

The reference model for evaporation used is the Dalton Model [10] shown as follows:

$$E = \Delta e \sqrt{(A + BW^2)} \quad (1)$$

,where E is the evaporation rate in mm/day,  $\Delta e$  is the deficit in vapour pressure, W is the wind speed 1.5 m above the liquid and A and B are constants. For this work, A is set to 0.2664 and B is set to 0.021 taken from a reference related to desert evaporation as the lab in winter is relatively dry [10]. Note that similar numbers are found from other sources for constants A and B. Figure 2 shows the theoretical relationship of the evaporation rate as a function of temperature, humidity, and covering wind speed. There is significant sensitivity to all three parameters.

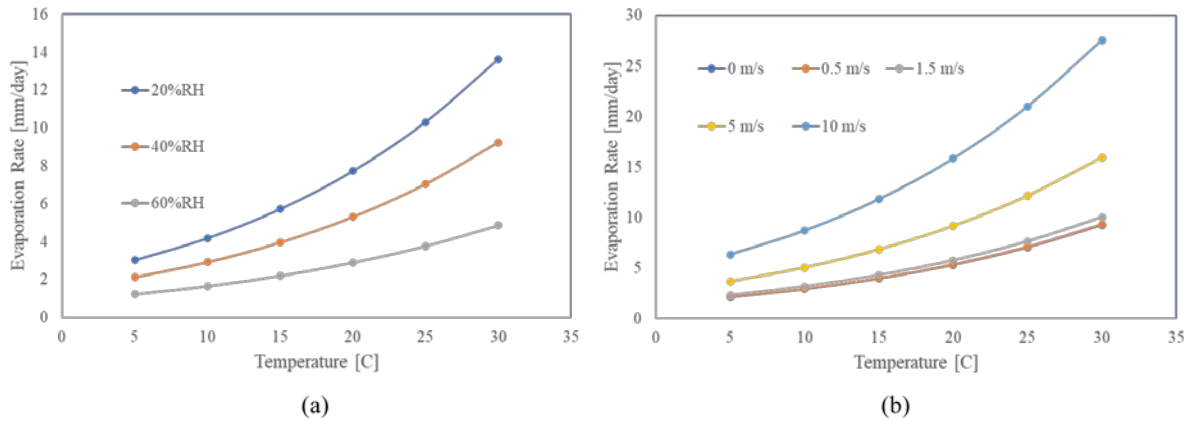


Figure 2 Effect of Environment on Evaporation Rate (a) temperature and humidity, (b) temperature and wind speed.

## 2.2 Distillation Experiments

The third set of experiments studies the application of solution heating to the PVA type gel. Gels are made and then heated at different rates in an Erlenmeyer flask connected to a distillation cooling jacket. The vapour produced from the heating process is condensed and recovered in a separate beaker. The recovered liquid is tested for pH and conductivity as well as visually inspected for clarity. The purpose of these experiments is to qualitatively determine the extent to which heating or boiling may accelerate the dehydration process.

Table 1 Experimental Hydrogel Samples for Evaporation Studies

| Sample # | Ingredients                       | Environmental Conditions |
|----------|-----------------------------------|--------------------------|
| 1        | Water Only                        | Open                     |
| 2        | 0.25g Sodium Polyacrylate + water | Closed                   |
| 3        | 0.25g Sodium Polyacrylate + water | 9 penetrations           |
| 4        | 1g Sodium Polyacrylate + water    | Open                     |
| 5        | 1g Sodium Polyacrylate + water    | Closed                   |
| 6        | 1g Sodium Polyacrylate + water    | 9 penetrations           |

Table 2 Experimental PVA Gel Samples for Evaporation Studies

| Sample # | Ingredients                       | Environmental Conditions |
|----------|-----------------------------------|--------------------------|
| 1        | Water Only                        | Open                     |
| 2        | 0.25g Sodium Polyacrylate + water | Closed                   |
| 3        | 0.25g Sodium Polyacrylate + water | 9 penetrations           |
| 4        | 1g Sodium Polyacrylate + water    | Open                     |
| 5        | 1g Sodium Polyacrylate + water    | Closed                   |
| 6        | 1g Sodium Polyacrylate + water    | 9 penetrations           |

### 3. Experimental Results

#### 3.1 Hydrogel Experiments

The experiment using the hydrogels in the laboratory is shown in Figure 3 where the sample number refers to the test case in Table 1. The samples were maintained at the same location and thus experience the same lab temperature, pressure and humidity.

The evaporation rate for the water only beaker was measured and compared to the Dalton Model. The wind speed was set to zero. There is an airflow in the lab due to the HVAC system but it is a slow airflow. The vapour pressure deficit was determined based upon a room temperature of 25 °C and a relative humidity of 50% and the vapour pressure determined using the Antoine model. The experiments were performed in January where the humidity is very low in the lab. The results showed an evaporation rate of 4.9 mm/day.

Figure 4 shows the comparison of the evaporation of the water beaker in comparison to the Dalton model for both the open-air lab case and the case where the samples are in the environment chamber. In both scenarios, adjusting for the humidity and temperature of the environments shows excellent agreement for both cases. The humidity is higher in the environment chamber as the air is not being replaced. There are some deviations and these are expected as the day to day humidity and lab temperature are not constant during the days of the measurement and the model assumes the average value.

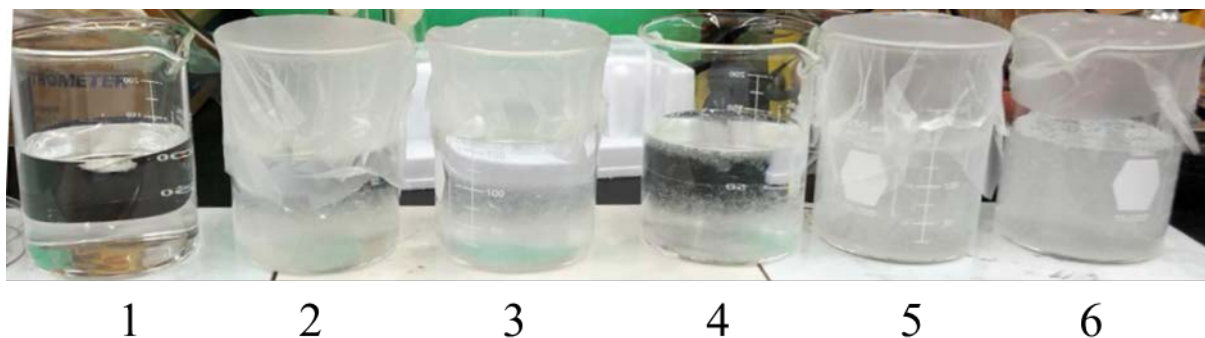


Figure 3 Samples of Sodium Polyacrylate gels with sample number as per Table 1.

The results imply that the evaporation rate can be controlled by using a controlled environment and adjusting the humidity.

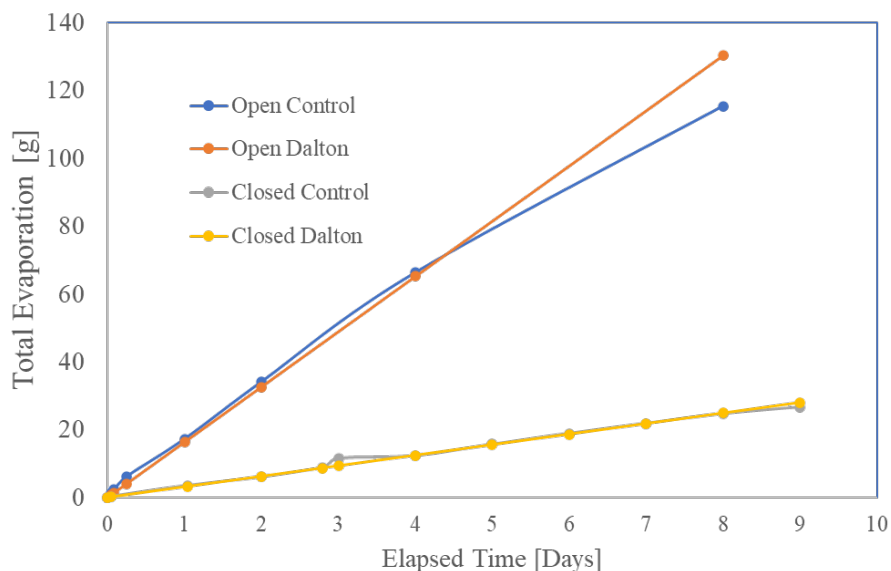


Figure 4 Evaporation of water reference test cases in open air lab experiment and enclosed environmental chamber with comparison to Dalton Model estimation

The evaporation rates for the remaining 5 samples are shown in Figures 5 and 6. One sample was found to have similar evaporation as the water reference case and the Dalton model. This sample was fully exposed to the laboratory atmosphere in the same manner as the reference sample. Thus, it is not surprising that the evaporation rates are similar. The rate is decreased slightly as time increases and this is likely due to the change in effective surface area as the gel components that have less water content are beginning to be exposed.

Two samples have no or little evaporation. These are the fully sealed samples and represent that it is possible to minimize evaporation of a hydrogel if the container is sufficiently sealed.

The remaining two samples are also sealed except that 9 holes of 6 mm diameter are punctured in the seal. One sample contained 0.25g of Sodium Polyacrylate hydrogel and the other contained 1g of Sodium Polyacrylate hydrogel. Both samples produced the same evaporation rate indicating that the amount of gel constituents does not affect the results in the early stages of evaporation where the water component is dominant and the environment is controlled.

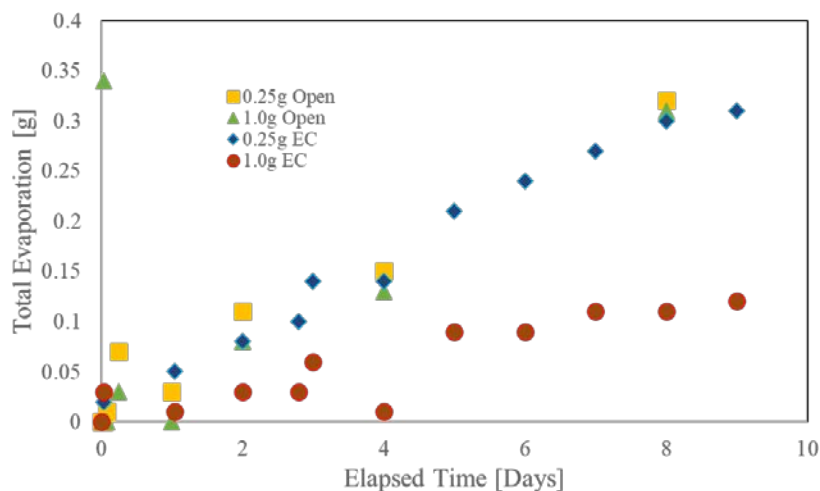


Figure 5 Comparison of the evaporation rate for the sealed hydrogel in the open lab air and the enclosed environment chamber with different hydrogel concentrations.

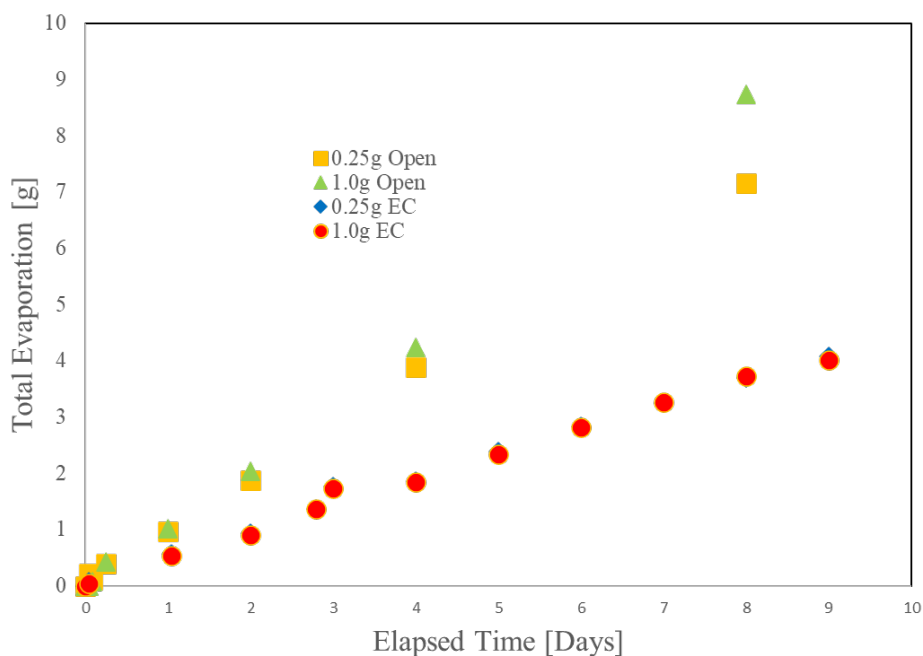


Figure 6 Comparison of the evaporation rate for the sealed hydrogel in the open lab air and the enclosed environment chamber with different hydrogel concentrations.

### 3.2 PVA based Gel experiments

The other gel studied is a PolyVinyl Acetate-water based gel. This gel was also observed to dehydrate naturally but the dehydration rate is much slower than the sodium polyacrylate gel. This type of gel has longer hydrocarbon chains and a stronger cross-linking resulting in a gel that is more difficult to break down. The same dehydration experiment is done as for the sodium polyacrylate gel.

In this case, a plastic container was used instead of open beakers. The plastic container is meant to have less evaporative losses through leaks in the seal. The container includes a plastic lid which is then sealed with electrical tape. 5 samples were prepared as per Table 2 with a typical sample shown in Figure 7. One sample was completely sealed while another was left fully open.



Figure 7 Typical PVA gel sample being measured. This sample is nearly fully evaporated.

Figure 8 shows the evaporation rates for the PVA gel. The gel samples are smaller in volume than in the hydrogel experiments and thus reach fully dehydrated states sooner. Figure 8(a) show the amount evaporated for the fully open container plateaus at 9 days. The rate was compliant with the Dalton model in the early days when significant water content remained. Figure 8 (b) shows that once the open surface area is accounted for in all of the samples, then the evaporation rate of the PVA type gel is consistent. Thus, it is possible to use the Dalton model, modified for the PVA gel constituency, for prediction of gel dehydration rates as long as the gel retains water content. Once the gel has fully evaporated, then of course, the model will deviate as it assumes an infinite supply of water is present.



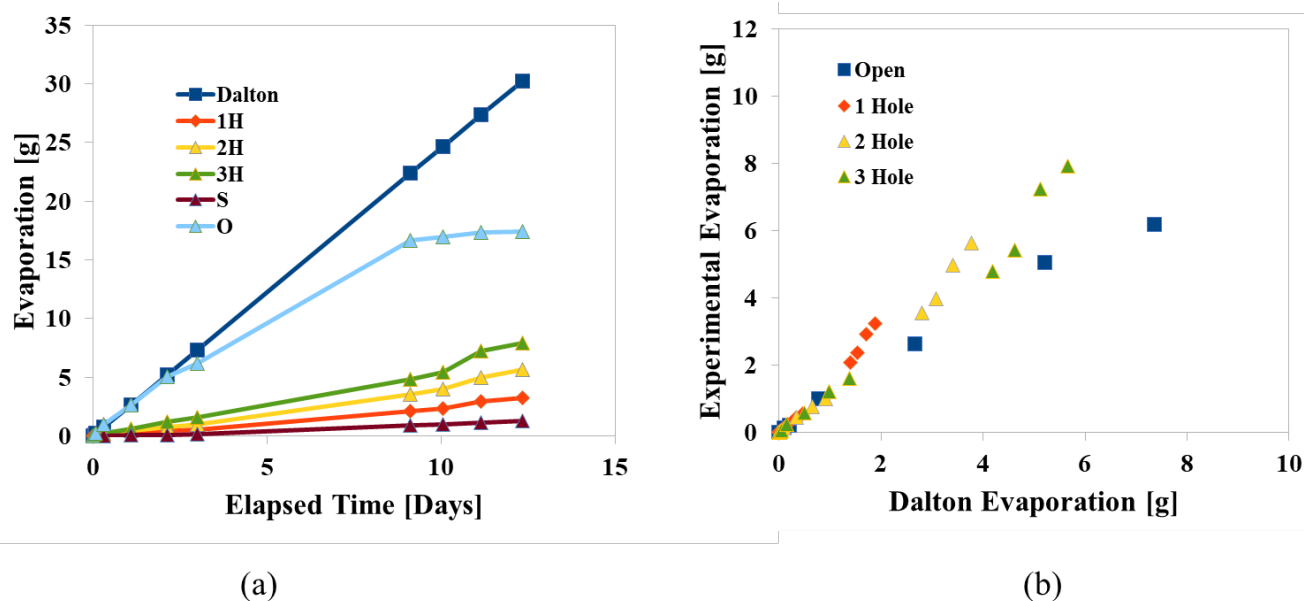


Figure 8 Evaporation of PVA type gel with comparison to standard Dalton model: (a) Evaporation Rate, (b) after adjustment for surface area; S – sealed, O – fully open, 1H – 1 penetration, 2H – 2 penetrations, 3H – 3 penetrations.

### 3.3 Gel Distillation Experiments

The results of the natural evaporation process clearly indicates that the rate can be controlled by the amount that the gel is exposed to an atmosphere. For the initial stages of decommissioning where the gel is used to collect contamination and to store it, keeping the container properly sealed will be an important step in the process to control the waste. When it is time to reduce the volume of the waste, natural evaporation can be used but will require a large open surface area to remove the air in a reasonable time frame. Small samples can be dehydrated within 1 week but larger containers would be limited. Assuming that the gel and waste were stored in a 208 L drum, and was filled to the full height of 88 cm (34.8”) then the approximate number of days required for evaporation can be determined. At a reference room temperature of 20 C and a humidity of 50%, the drum would release water vapour at a rate of 4.1 mm/day which corresponds to approximately 215 days for full evaporation.

The humidity and temperature could be adjusted to increase the rate but it is unlikely to be a practical solution to allow for natural dehydration of the PVA type gel. Air flow rate through a vented filter system is another possibility for increasing and controlling the evaporation rate although it is suspected that the exposed surface area will be extremely important.

Heating the gel will of course increase the vapour release rate. An experiment was performed where the gel solution was placed in an Erlenmeyer flask and heated near boiling. The water vapour was released and passed through a condenser column and recovered in a separate beaker. The purpose of this experiment was to determine if the rate of evaporation (or boiling in this case) could be increased. The gels were completely boiled within 2 hours which is much faster than the natural evaporation processes. However, some barriers were observed. Applying too much heat to the gel results in

breakdown of the gel constituents and the release of gel into a wet vapour. This phenomenon is a type of carryover event where the steam is now transporting droplets of gel. This results in the recovery beaker being contaminated and defeating the purpose of the test.

Experiments at a lower heating rate did not produce any carryover and essentially a distilled water was obtained. The success at lower heating rates suggests that it would be possible to dehydrate the gel. Further experiments are necessary to determine the optimal conditions to ensure good dewatering of the gel without carryover of any non-water elements.

#### **4. Concluding Remarks**

In this work we have studied the possibility of dehydrating gels to reduce their ultimate waste volume. Two gels were studied: a hydrogel and a PVA based gel. In both cases dehydration is possible with volume reductions greater than 80%. The optimal volume reduction will require further study to quantify.

Natural evaporation processes are faster for the hydrogel than for the PVA based gel which is expected due to the stronger cross-linking with the PVA based gel. The hydrogel would evaporate very close to the same rate as water. This means that while natural evaporation process is effective in reducing volume it may be too fast and not allow the hydrogel to process the clean-up properly.

The PVA gel natural evaporation rate is 5 times slower than the hydrogel. This results in sufficient work time to allow the PVA gel to be set and to process any form of contamination be it liquid or solid in nature. However, the slow rate of evaporation means that the PVA gel would need approximately 215 days for a 200L drum to completely evaporate. Thus, it may be useful to apply heat or increase the air flow through the drum to increase the evaporation rate.

Simple boiling and condensing experiments showed that the PVA gel can be reduced very quickly and small volumes can be done in one to two hours. At a slow heating rate, the vapour produced appears to be equivalent to distilled water although tests are needed to confirm. However, if the heating rate should increase significantly, this can result in liquid carryover and significant frothing of the gel. In this case, the distillate becomes contaminated.

This work is a proof in principle that gel evaporation can be achieved and that large volume reductions can also be achieved. The main residual concern is ensuring that only water is liberated during the dehydration processes. Future work will aim to quantify the volume reduction and test the evaporated product to confirm that only water is present.

#### **5. References**

- [1] J. R. Grover, "The Solidification of High-Level Radioactive Wastes," International Atomic Energy Agency.
- [2] J.L. Thompson, "Vitrification of Nuclear Waste," Stanford University, Stanford, 2010.
- [3] World Nuclear Organization, "Treatment and Conditioning of Nuclear Waste," World Nuclear Organization, June 2017.
- [4] J.B. Davies and C. Baldock, "Sensitivity and stability of the Fricke-gelatin-xylenol orange gel dosimeter", *Radiation Physics and Chemistry*, **77**: 690-696. 2008.
- [5] Y. Aoki, G. Harvel, and T. Sunagawa, "Research and Development of a New Type Gel Dosimeter" PVA-KI Gel Dosimeter", PBNC 2018, San Francisco, USA, October 2018.

- [6] G. Harvel, V. Sarvendran, and R. Khurmi, “Development of Gels for Decommissioning Applications”, 5<sup>th</sup> Canadian Conference on Nuclear Waste Management, Decommissioning, and Environmental Restoration, Niagara Falls, Canada, August 27-31, 2023.
- [7] Ashkan Bigham, Erfan Rezvani Ghomi, Hongrong Luo, Seeram Ramakrishna, Mina Zare, Mohamad Zare, “Phema: An overview for biomedical applications,” International journal of molecular sciences, 22(120): 6376, June 2021.
- [8] O. Asafu-Adjaye, B. Via, B. Sastri, and S. Banerjee, “Continuous dewatering of sludge and aqueous slurries by sodium polyacrylate,” Case Studies in Chemical and Environmental Engineering, December 2022
- [9] Z. li, N. Pan, Y. He and Q. Zhang, “Evaluating the best evaporation estimate model for free water surface evaporation in hyper-arid regions: a case study in the Ejina basin, northwest China”, Environ Earth Sci. 75:295, 2016. DOI 10.1007/s12665-015-5090-3.
- [10] S. Ali, N.C. Ghosh, and R. Singh, “Evaluating best evaporation estimate model for water surface evaporation in semi-arid region, India”, Hydrological Processes 22: 1093-1106, 2008. DOI 10.1002/hyp.