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Application of Photothermal Driven Hydrogel in Waste water Desalination

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Abstract:

Water shortage is becoming one of the major social concerns. Evaporation and distillation of wastewater contribute to deal with this problem, while hydrogel, which is a macromolecular polymer, facilitates the evaporation and distillation. Acrylamide (AAm) can undergo addition polymerization whose product can be used to manufacture hydrogel with water and carbon nano tubes. Carbon nano tubes (CNT), which are very close to blackbodies, can increase the thermal absorption of hydrogels. It is the aim of this study to discuss and analyses the proportions of the ingredients, including AAm, CNT and water, and propose the proportions that have the strongest evaporation efficiency, swelling ability and water retention capacity. It was investigated that hydrogel with 0% of CNT, 20% of AAm and 80% of water has the strongest swelling ability, and hydrogel with 0.5% of CNT, 25% of AAm and 74.5% of water has the strongest water retention. It was also proved that adding carbon nano tubes can significantly improve the evaporation efficiency during the distillation process. This study has the significant potential for application to wastewater treatment. It is hoped that this study can reship and enrich wastewater treatment methods.

Keywords: Acrylamide hydrogel, Carbon materials, Solar-driven evaporation, γ-ray, Methyl blue

1 Introduction

As the rapid development of industrial, environmental pollution has become more serious. Industrial waste water and waste gas release causes a large number of harmful pollutants, such as heavy metal or microplastic, enter the water, threating the ecosystem and human health. At the same time, the global water source shortage problem is becoming increasingly sever. Thus, developing an efficient and low energy consumption technology to help produce freshwater from sea water or polluted water will be very meaningful. Even though traditional methods, for example distillation or chemical degradation, could effectively alleviate the water pollution issue, it still has limitations in many aspects, such as the total cost or efficiency, that limits its large-scale application. Therefore, it is still necessary to develop a more efficient, reliable method to remove pollution from sea water or polluted water.

Take acrylamide (AAm) which is the homomer used in this investigation as an example. There are mainly two mechanisms of the synthesis of hydrogels using AAm. AAm is dissolved in water. As shown in Figure 1, under γ-ray irradiation, AAm double bonds open to form free radicals and attack other double bonds, rapidly forming cross-linked PAAm hydrogels. [1]

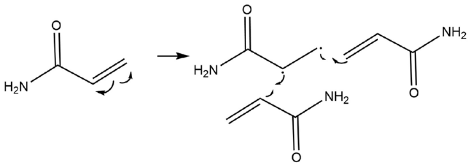

Figure 1. Mechanism diagram of acrylamide polymerization induced by γ-ray

As a kind of polymer material with three-dimensional network structure, hydrogel has high water content, versatility, responsiveness and other unique properties, which can be combined with various other materials to form composite materials, showing application potential in many fields. For example, hydrogels with TiO_2 catalysts were used to adsorb metal ions, dyes, and other aquatic pollutants due to their mechanical properties. Also, the TiO_2 catalysts in these hydrogels facilitate the photocatalytic degradation of pollutants. In addition, these hydrogels were very durable and stable, enables them to be reused several times.[2] Hydrogels with polyacrylamide, graphene and silver can increase water delivery efficiency in seawater treatments and have long-term antimicrobial performance, making them have the potential for wastewater treatments.[3] Additionally, many hydrogels can be applied to environmental management, flexible sensing, electrical engineering, and biomedicine due to their conductivity, mechanical flexibility, self-healing capabilities, environmental adaptability, and biocompatibility.[4] By combining poly (norepinephrine) with histidine, the photothermal efficiency and evaporation performance of hydrogels was enhanced significantly by 48%, which increases the efficiency of thermal energy conversion.[5]However, functionalization of existing polymers is mainly used to synthesize hydrogel from natural materials.[6] For instance, polysaccharides and proteins can be extracted from animals, plants, and seaweed, and these molecules can be applied to synthesized.

In our study, we analyzed the properties and applications of hydrogels in wastewater treatment by synthesising hydrogels with different proportions of ingredients, including carbon nano tube, water and acrylamide. We will compare the differences in the performance of different hydrogels with different components, and hope to find the most outstanding one.

2 Experimental Procedure

2.1 Materials

The AAm used in this experiment was purchased from Sinopharm Group Chemical World Co., LTD, and the carbon nano tube (CTN) used in this experiment was bought from Shenzhen Nanotech Port Co., Ltd. The 1-dodecyl-3-vinylimidazole chloride ([VC12m]Cl) and 1-butyl-3-vinylimidazolium chloride ([VBIm]Cl) were purchased from Lanzhou Yulu Fine Chemical Co., Ltd. The methyl blue was bought from Sinopharm Group Chemical World Co., LTD.

2.2 Preparation of hydrogel 0-0

12.5 g of AAm and 37.5 g of water were weighted using an electric balance and a measuring cylinder, respectively. AAm and water were mixed in a beaker, and the mixture was placed in an ultrasonic cleaner to dissolve the AAm and to make the solution even. The solution was transferred to vials. Nitrogen was delivered into the half-full vials to remove oxygen in the vials, and the vials were sealed. The solution was exposed under γ-ray to polymerize for 6 hours, so hydrogel 0-0 was prepared.

2.3 Preparation of hydrogel 1-1 to 1-3

Table 1. proportions of ingredients of different hydrogels

Number	Carbon nano tubes (CNT) /g	Acrylamide (Aam) /g	Water/g	1-dodecyl-3-vinylimidazole chloride ([VC12m]Cl) /g	1-butyl-3-vinylimidazolium chloride ([VBIm]Cl) /g
0-0	0	12.5	37.5	0	0
1-1	0.5	12.5	37.5	0.25	0.25
1-2	0.25	12.5	37.5	0.25	0.25
1-2-2	0.25	12.5	37.5	0.25	0
1-3	0.05	12.5	37.5	0.25	0.25
2-1	0	10	40	0	0
2-2	0	15	35	0	0
2-3	0	2.5	47.5	0	0

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12.5 g of AAm, 0.5 g of CNT, 37.5 g of water, 0.25 g of [VC12m]Cl and [VBIm]Cl were weighted. The chemicals were mixed in a beaker, and the mixture was placed in an ultrasonic cleaner to dissolve the AAm and CNT even in the solution. The solution was transferred to vials. Nitrogen was delivered into the half-full vials to remove oxygen in the vials, and the vials were sealed. The solution was exposed under γ-ray to polymerize for 6 hours, so hydrogel 1-1 was prepared. The previous steps were repeated without 0.25 g [VBIm]Cl, so hydrogel 1-2-2 was prepared. The previous steps were repeated with 0.25 g and 0.05 g of CNT, so hydrogels 1-2 and 1-3 were prepared. The detailed formula is shown in Table 1.

2.4 Preparation of hydrogel 2-1 to 2-3

10 g, 15 g and 2.5 g of AAm and 40 g, 35 g and 47.5 g of water were weighted, and the chemicals were mixed in a beaker. The mixture was placed in an ultrasonic cleaner to dissolve the AAm. The solution was transferred to vials. Nitrogen was delivered into the half-full vials to remove oxygen in the vials, and the vials were sealed. The solution was exposed under γ-ray to polymerize for 6 hours, so hydrogels 2-1 to 2-3 were prepared. The detailed formula is shown in Table 1.

2.5 Characterizations

The chemical composition of composite hydrogels was analyzed by a Fourier infrared spectrophotometer (FT-IR, Bruker Vertex 70). Thermal properties were analyzed using differential scanning calorimetry (DSC, Mettler Toledo) under a nitrogen atmosphere. A UV-vis-NIR spectrophotometer (Shimadzu, SOLID3700) equipped with an integrated sphere was used to measure the solar transmittance of the sample in the range of 200-2500 nm corrected.

2.6 Solar-driven interfacial evaporation

Hydrogel 1-2 was cut into a short cylinder with a diameter of 3.5 cm and a height of 1 cm. A foam board was cut into a ring with an external diameter that was equal to the diameter of a beaker and internal diameter of 3.5 cm, and the hydrogel cylinder was placed on the foam board. Then, the foam board with hydrogel was placed with a beaker that contained 100 ml of methyl blue solution with concentration of 10 mg/L. Half of the foam board was submerged into the methyl blue solution, and the beaker was placed on a culture dish and was covered by a large beaker. The culture dish with the two beakers was exposed under lighst generated by a solar simulator. After 5 hours when the solution evaporated under the light, water was condensed on the wall of the larger beaker. The wall of the large beaker was washed using distilled water, and the distilled water with water on the wall was collected into a test tube. 2 ml of water was collected and analyzed using an ultraviolet spectrograph.

2.7 Swelling experiment

The hydrogels (1-1, 1-2, 1-2-2, 2-1, 2-2) were cut into a cylinder about 1cm high and then divided into quarters, then they were put into the dryer in order to dry out the moisture. After the hydrogels were completely dry, their mass was recorded. Then they were separately moved into different bags, and deionized water was added into the bags until the hydrogels were completely submerged. In the first hour, the hydrogels were removed from the water and had been weighed for every 20 minutes, and they have been moved back to the water in the bag immediately after being weighted. During the second and third hour, the steps were repeated every 30 minutes. And after three hours, the repetition was done once per hour. Note that all of the weighted data has been recorded carefully.

2.8 Water retention experiment

All hydrogel samples (1-1, 1-2, 1-2-2, 2-1, 2-2) were cut into 1/4 cylinder with about 1 cm high, and they were separately put into bags. Deionized water was added into the bags until the hydrogels were completely submerged. After the hydrogels absorbed enough water, they were placed into a dryer with a standard temperature (323 K). The timer was set for every 20 minutes in the first hour, every 30 minutes in the second and third hour, and every hour in the rest of the time. The hydrogels were taken out of the dryer and were weighed when the timer reminded us to do so, and all weighting data were recorded carefully. The experiment is finished when the mass change of the hydrogel is approaching zero.

The change of the water retention capacity in a certain period of time is how we compare the hydrogen's ability of water retention. The water retention was determined by the rate of change of the hydrogel's mass after a specific period of time. The water retention capacity (%) could be expressed by the equation:

Water retention capacity = $(m_0\text{-}m_1)/m_0\times100\%$

Where m_1 (g) is the loss of water quality during the sample dehydration process, and m_0 (g) is the initial water content of the sample (Total gel weight multiplied by gel water content mass fraction). The experiment is finished when the mass change of the hydrogel is approaching zero.

3 Results

3.1 Design and characterization of hydrogels

Figure 2. Hydrogel photo of 0-0, 1-1, 1-2, 1-2-2, 1-3 (from left to right)

All vials filled with solution are solidified into colloidal solid after UV radiation (Figure 2). Among them, hydrogel 0-0, 2-1 to 2-3 is a transparent solids with some bubbles because of the absence of CNT. 1-1, 1-2, 1-2-2 and 1-3 had a certain amount of black sediment at the bottom of the bottle, and the upper part was approximately translucent. However, since 2-3 only involved 2.5 g AAm, so it does not have enough cohesion to form a stable hydrogel. Since 1-3 only involved 0.05 g CNT, there is only a very little amount of black sediment at the bottom of the bottle, which is not able to be applied in waste water desalination.

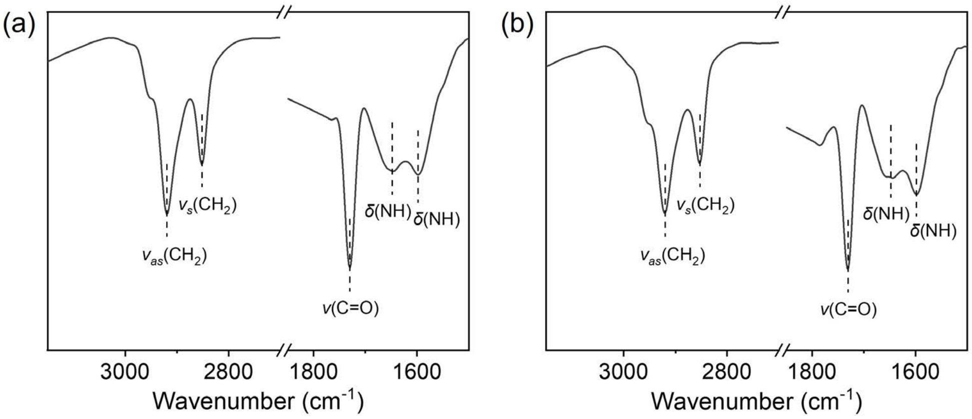

Figure 3. FTIR spectra of 0-0 (a) and 1-3 (b)

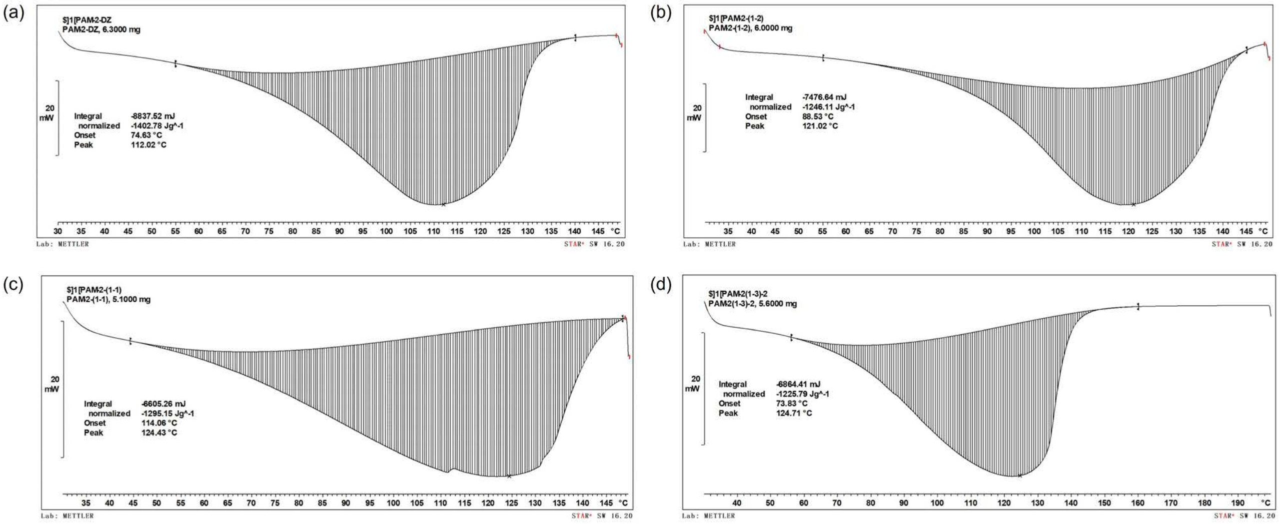

Figure 4. DSC curve of 0-0 (a), 1-1 (b), 1-2 (c) and 1-3(d)

In order to further verify the preparation of hydrogels, we conducted FTIR tests on the samples. As shown in Figure 3a and 3b, it can be seen that 0-0 and 1-3 have consistent peaks, which is due to the almost no signal of CNT in FTIR testing. The characteristic peaks of -CH3 groups appeared at 2922 cm⁻¹ and 2853 cm⁻¹, the peak at 1730 cm⁻¹ represented C=O bonds, the peaks at 1647 cm⁻¹ and 1599 cm⁻¹ were belonged N-H bonds of amide groups. It can be concluded that hydrogels with a polyacrylamide backbone were successfully prepared via γ-ray irradiation.

To explore the evaporation behavior of water in hydrogels, the vaporization enthalpy of water in 0-0, 1-1, 1-2 and 1-3 was investigated by means of a differential scanning calorimeter (DSC). As shown in Figure 4a-d, the enthalpy of vaporization of interfacial water in 0-0, 1-1, 1-2 and 1-3 was 1402.78 J/g, 1246.11 J/g, 1295.18 J/g and 1225.79 J/g. This result shows that after the addition of CNT, the enthalpy of vaporization of interfacial water in gel decreased significantly. That is, the polymer network we constructed realized efficient water activation.

3.2 Solar-driven interfacial evaporation

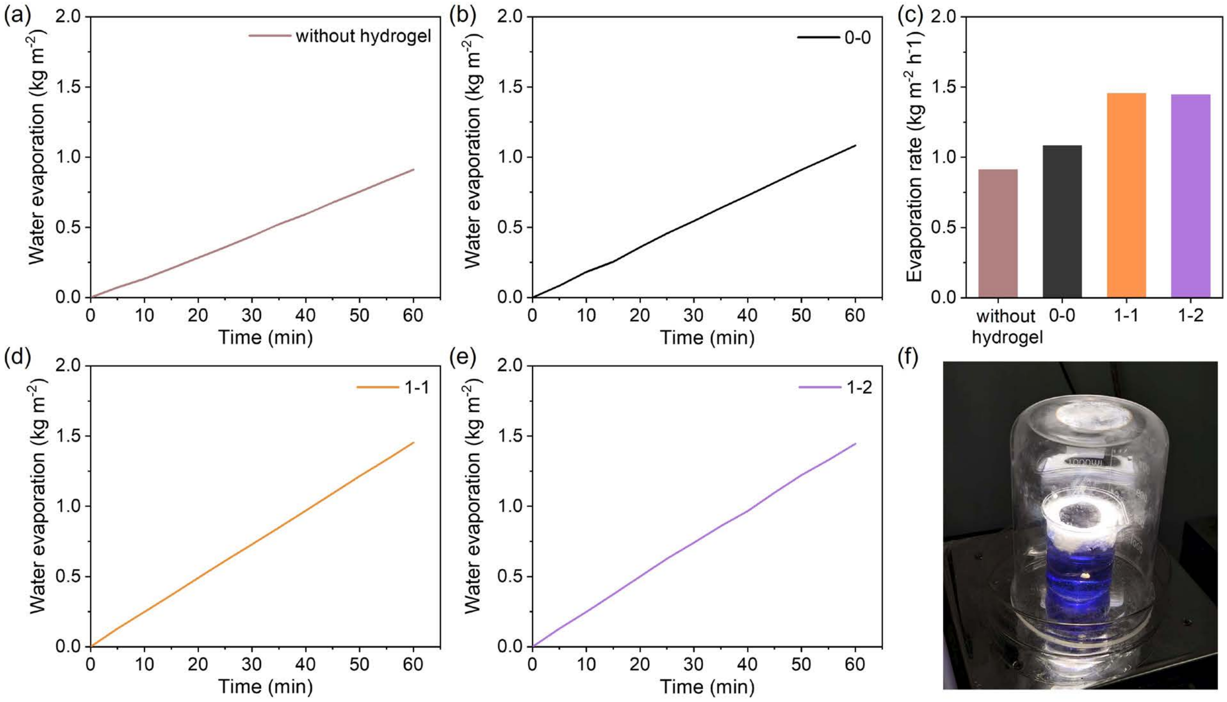

Figure 5. Water evaporation in 1 h of 10 mg/L methyl blue solution without hydrogel (a), under 0-0 (b), under 1-1 (d) and under 1-2 (e). (c) Evaporation rate of 10 mg/L methyl blue solution without hydrogel, under 0-0, under 1-1 and under 1-2. (f) Methyl blue solution under simulated sunlight xenon lamp light source

The solar evaporation system simulated in the laboratory is shown in Figure 5f. In order to measure the evaporation rate driven by solar energy, three groups of hydrogels fixed in foam and a control group are exposed to solar radiation (the sunlight simulated by a xenon lamp light source) for evaporation experiments. The methylene blue solution is used to simulate sewage, and the air humidity is controlled at about 45%. Under the irradiation of a certain intensity of a xenon lamp, the hydrogel was put into a beaker containing methylene blue solution. Water molecules constantly escape from the evaporator surface to the air. After preheating for about half an hour, when the surface temperature of the hydrogel is stable, the mass loss of the hydrogel is monitored for one hour.

We used 0-0, 1-1, 1-2 and directly sunlight methyl blue solution as the control group to evaluate the purification ability of different hydrogels. Within one hour, water evaporation in each group of photothermal evaporation experiments increased at a nearly constant rate (Figure 5a, 5b, 5d, 5e), which proved that the hydrogels had good stability under light conditions. The evaporation rate of water under two groups of hydrogels added with CNT reached approximately 1.5 kg m^{-2} h^{-1}, while the evaporation water without hydrogel and water under 0-0 was 0.91 kg m^{-2} h^{-1} and 1.08 kg m^{-2} h^{-1} (Figure 5c), which showed that the black body hydrogel had better photothermal performance than the ordinary hydrogel.

Comparing the UV absorption of the initial methyl blue solution and the photothermal evaporation product at 600 nm, it is not difficult to find that the methyl blue solution has an obvious absorption peak at 600 nm, while the distilled water obtained by photothermal evaporation of 1-1 hydrogel has no absorption of 600 nm UV light (Figure 6). As a result, the water was proven to be pure, which means the evaporation using hydrogel was justified to be applicable to wastewater treatments.

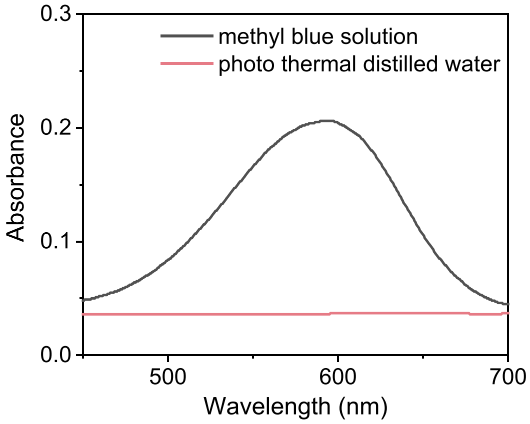

Figure 6. UV–vis absorption of initial methyl blue solution and photo thermal evaporation products

3.3 Swelling experiment

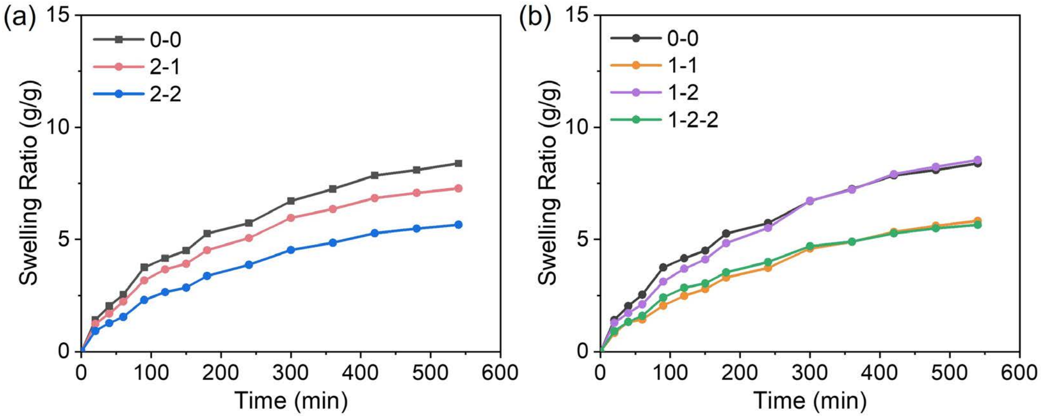

Figure 7. (a) Water absorption swelling test of 0-0, 2-1 and 2-2. (b) Water absorption swelling test of 0-0, 1-1, 1-2 and 1-2-2

In order to explore the continuous water transport capacity of hydrogels as an important role in the distillation process, we compared the water absorbability of different hydrogels by water swelling experiment, and the hydrogel's water absorbability was compared by its mass after water absorption in the same period of time. According to Figure 7, before about 300 minutes, 0-0 had a larger swelling ratio than 1-2, and 2-2 had a larger swelling ratio than 1-1, which means that adding CNT in hydrogels can inhibit the rapid swelling of hydrogels when encountering water. After 300 minutes, 1-2 had a larger swelling ratio than 0-0, and 1-1 had a larger swelling ratio than 2-2, which means that adding CNT to hydrogels can improve the total amount of water absorbed by the hydrogels. In conclusion, adding CNT to hydrogels can improve the water swelling ratio.

3.4 Water retention experiment

We compared the water retention capacity of different hydrogels by a water retention experiment (Figure 8), in order to find out the capacity of water retention during the drying process. The hydrogel's water retention capacity was compared by mass after water drying for the same period of time. According to Figure 8, before about 180 minutes, 2-1 and 2-2 had a larger weight retention than 1-2, and 0-0 had a larger weight retention than 1-1 which means that the CNT caused the hydrogel to lose more water during the first 180 minutes. After about 270 minutes, 1-2 had a larger weight retention than 2-1 and 2-2, and after about 180 minutes, 1-1 had a larger weight retention than 0-0. The two groups of comparisons showed that after a specific time, the CNT in the hydrogels helps to prevent water from evaporating from the hydrogels. In conclusion, adding CNT to hydrogels can improve the water retention capacity.

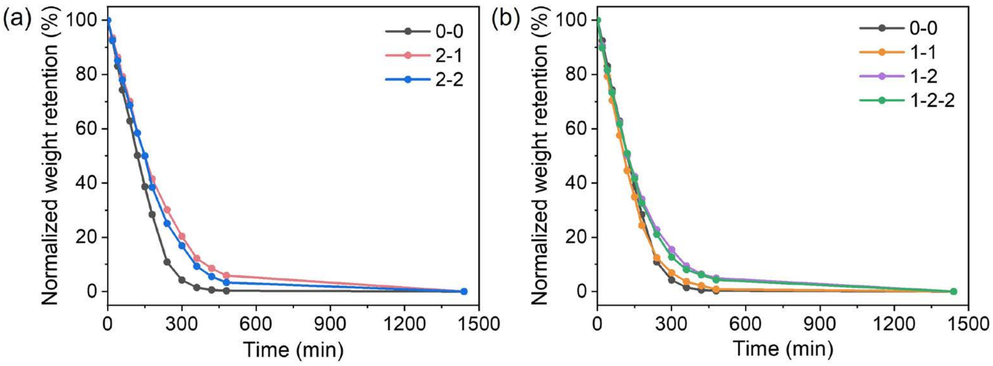

Figure 8. (a) Normalized weight ratios (normalized remaining mass) of 0-0, 2-1 and 2-2. (b) Normalized weight ratios (normalized remaining mass) of 0-0, 1-1, 1-2 and 1-2-2

4 Conclusion

In this study, we designed a black body hydrogel evaporator formed by adding CNT, which has the characteristics of high evaporation efficiency, stability, water retention and antifouling. Its three-dimensional porous structure makes the hydrogel achieve efficient water transport, and the hydrogel evaporator has shown an excellent evaporation rate. At the same time, through the comparison of multiple groups of experiments, we explored the effect of different doses of CNT on drying, water retention and evaporation rate. The efficiency and applicability in production were explored, which provided a basis for the application of black body hydrogel. In addition, CNT added in the black body hydrogel not only has advantages in increasing the evaporation rate in the distillation process, but also can change the swelling capacity of the hydrogel and increase the stability and strength of the hydrogel. It shows excellent versatility, which greatly solves the efficiency problem of sewage purification. The black body hydrogel evaporator developed in this study has advantages in production and cost, and has brought great potential for subsequent applications. Through low-cost fresh water production, it provides an efficient and economical solution to the global water shortage crisis and the global marine water pollution problem.

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