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Synthesis and Study on Swelling Properties of Polyaniline/Polyacrylamide Hydrogel Composites

Sayantika Adhikari

Central Institute of Plastics Eng. & Technology, Lucknow-226008
adhikarisayan90@gmail.com

Abstract—*The study presents a scalable and versatile synthesis of Conducting Polymer Hydrogel composites, having Polyaniline (PAni) as conducting filler and Polyacrylamide (PAAm) as matrix. Polyaniline is dispersed by chemical oxidative polymerization technique inside the hydrogel network (in situ). The effect of swelling and dehydration properties of the composites were evaluated by change in weight at ambient temperature with time and equivalent water content was calculated. The results indicate that the basic swelling properties are least affected by incorporation of PAni in hydrogel network. These composites represents a simple and efficient method for development of electro-active hydrogels for various applications such as for electrochemically controlled drug delivery devices, bioelectronics and energy storage devices.*

Keywords*: Conducting polymers, Hydrogels, Polyaniline composite, Swelling property*

1. INTRODUCTION

Hydrogels are three dimensional networks of cross-linked hydrophilic polymers that typically retain around 30% weight of water. These water-containing gels have received a lot of attention globally due to their scope for different applications, such as controlled drug release, ocular devices, soil additive to conserve water, wound dressings, food-thickening agents, implants or other applications that require the utilization of biocompatible materials. Polyacrylamide hydrogels (PAAm) can absorb relatively high amounts of water and their swelling capacity is not very sensitive to pH or electrolytes. Conducting polymers have been incorporated into Hydrogels to form conducting hydrogel composite interpenetrating networks. A conducting hydrogel could be used in fuel cells, super capacitor, dye sensitive solar cell and rechargeable lithium batteries due to its better conductivity property, colloid stability, low cost and simple preparation. The incorporation of the electro active polymer into a highly permeable hydrogel matrix has been pursued as a route to the development of a novel class of potentially biocompatible, smart hydrogel composites that can find varied applications in bio systems. The incorporation of conducting polymers inside the Hydrogel network has been widely studied. Electro-polymerization of Polyaniline inside the Hydrogel matrix was reported for electro controlled release of safranin. Similarly, chemical in-situ polymerization of aniline inside the Hydrogel matrix has been reported for Poly Vinyl alcohol based hydrogels.

Formation of Nano-dimensional PAni inside the hydrogel network is yet to find a mention in literature. In the current study, Hydrogel composites have been developed by incorporating Polyaniline into hydrogel network through chemical synthesis after network formation of Hydrogel. This process is relatively simple and economical.

2. EXPERIMENTAL

2.2. Materials used

Acrylamide (AAm), N, N′-methylene-bis-acrylamide (MBA), Potassium persulfate (KPS), N,N,N′,N′-tetramethylethylenediamine (TEMED), and Aniline (Ani) were purchased from Sisco Research Lab, Mumbai. Aniline was double distilled before use. Ammonium per oxy disulphate (APS) and Hydrochloride (HCl) were supplied by Loba chemie Ltd, Mumbai, India.

2.3. Synthesis of polyacrylamide hydrogels

Table 2.1 shows the compositions of 3 physically different polyacrylamide hydrogels prepared as per the procedure reported by Wallace et al. Different amounts of both acrylamide (AAm) and N,N′-methylene-bis-acrylamide (MBA) were dissolved in 10ml of distilled water at ambient temperature and stirred under nitrogen atmosphere for 10 minutes. Then the initiator (KPS, 0.01g) and TEMED, 100µl were added with vigorous stirring. TEMED was employed to provide a more stable media for the polymerization of acrylamide. Before gelification, this pre-gel solution was poured in pre-marked cylindrical glass tubes to obtain cylindrical hydrogels. After gelification, hydrogels were extracted by breaking the tubes and immersed in distilled water for 24 hours for stabilization and removal of impurities.

Table 2.1: Polyacrylamide Hydrogels composition

Hydrogels	1	2	3
AAm/g	1.00	0.50	2.00
MBA/g	0.030	0.026	0.270

Amounts of monomers added to 10mL distilled water + 100µL TEMED + 0.010 g of Potassium peroxosulfate.

2.3. Impregnation of Polyaniline

Hydrogels were dried and left for 24 hours before impregnation of Aniline. A given amount of Aniline (2.32gms) was dissolved in 1M HCl and the gel prepared was allowed to soak in the 0.5 M Aniline solution for 24 hours. This aniline containing gel was washed with distilled water and allowed to dry. The dried gels were dipped in a 0.5M APS solution (5.71gms) and allowed to soak. As soon as the gel swells in APS solution, the APS initiates polymerization of Aniline. As the polymerization reaction proceeds, the semi-transparent gel turns green in color. The PAni impregnated gel is repeatedly washed with distilled water and then dried at room temperature.

3. RESULTS AND DISCUSSION

3.1 Dehydration of hydrogels at ambient temperature with time

Pure hydrogels and PAni/PAAm hydrogels were dried and the rates of dehydration are shown in table 3.1. The rate of dehydration was decreased with increased amount of PAni.

The initial dehydration rate for pure hydrogels was slightly higher compared to the PAni/PAAm hydrogels. For the PAni/PAAm hydrogel composites there may be retardation in diffusion of water through the 3-D network due to the presence of PAni in the porous hydrogel network.

Table 3.1: Dehydration studies of pure hydrogels and PAni/PAAm hydrogels

Sl. No.	Time(hours)	Dry wgt. Of pure hydrogels			Dry wgt. Of PANi/PAAm hydrogels		
		Sample 1	Sample 2	Sample 3	Sample 4	Sample 5	Sample 6
1	0.0	1.14	1.32	0.97	1.35	1.02	0.95
2	0.5	1.13	1.29	0.92	1.30	0.97	0.89
3	1.0	1.07	1.26	0.89	1.23	0.92	0.83
4	1.5	0.97	1.20	0.84	1.13	0.88	0.79
5	2.0	0.95	1.16	0.79	1.06	0.85	0.73
6	2.5	0.92	1.13	0.74	1.00	0.80	0.67
7	3.0	0.90	1.07	0.69	0.96	0.76	0.64
8	3.5	0.87	1.03	0.65	0.87	0.72	0.58
9	4.0	0.83	1.00	0.60	0.86	0.70	0.56
10	4.5	0.80	0.97	0.57	0.83	0.69	0.52
11	5.0	0.77	0.92	0.53	0.74	0.68	0.48
12	5.5	0.73	0.89	0.50	0.70	0.67	0.47
13	6.0	0.71	0.84	0.49	0.69	0.66	0.46
14	6.5	0.68	0.80	0.45	0.65	0.64	0.42
15	7.0	0.65	0.77	0.41	0.63	0.62	0.41
16	7.5	0.61	0.73	0.39	0.61	0.60	0.40
17	8.0	0.59	0.70	0.35	0.58	0.58	0.39
18	8.5	0.54	0.66	0.33	0.57	0.56	0.38
19	9.0	0.49	0.64	0.30	0.55	0.54	0.38
20	9.5	0.48	0.62	0.30	0.53	0.52	0.37
21	10.0	0.47	0.60	0.30	0.52	0.50	0.37
22	10.5	0.44	0.57	0.26	0.50	0.48	0.36
23	11.0	0.40	0.55	0.26	0.48	0.46	0.36
24	11.5	0.37	0.52	0.26	0.46	0.44	0.36
25	12.0	0.35	0.49	0.26	0.45	0.43	0.35
26	12.5	0.33	0.47	0.25	0.43	0.42	0.35
27	13.0	0.31	0.45	0.25	0.42	0.41	0.35
28	13.5	0.29	0.43	0.25	0.40	0.40	0.35
29	14.0	0.27	0.41	0.25	0.38	0.39	0.34
30	14.5	0.25	0.39	0.24	0.37	0.38	0.34
31	15.0	0.23	0.37	0.24	0.37	0.37	0.34
32	15.5	0.21	0.35	0.24	0.37	0.36	0.34
33	16.0	0.19	0.33	0.24	0.37	0.35	0.34
34	16.5	0.18	0.31	0.24	0.36	0.34	0.34
35	17.0	0.17	0.29	0.23	0.36	0.33	0.34
36	17.5	0.16	0.27	0.23	0.36	0.32	0.33
37	18.0	0.15	0.25	0.23	0.36	0.31	0.33
38	18.5	0.14	0.24	0.23	0.36	0.30	0.33
39	19.0	0.13	0.23	0.23	0.35	0.29	0.33
40	19.5	0.13	0.22	0.23	0.34	0.28	0.33
41	20.0	0.12	0.21	0.23	0.33	0.27	0.33
42	20.5	0.12	0.20	0.22	0.33	0.27	0.33
43	21.0	0.11	0.19	0.22	0.33	0.26	0.32
44	21.5	0.11	0.19	0.22	0.33	0.26	0.32
45	22.0	0.11	0.18	0.22	0.33	0.25	0.32
46	22.5	0.10	0.18	0.22	0.32	0.25	0.32
47	23.0	0.10	0.17	0.22	0.32	0.24	0.32
48	23.5	0.10	0.16	0.22	0.32	0.24	0.32
49	24.0	0.10	0.15	0.22	0.32	0.24	0.32

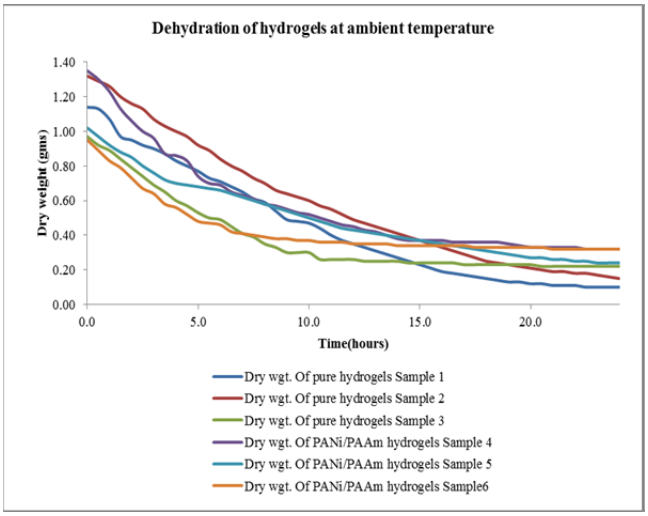

Fig. 3.2: Dehydration of hydrogels at ambient temperature

3.2 Swelling behavior of hydrogels

The swelling behavior of Hydrogel composites were studied over 24 hours till the weight increase became constant. Samples were weighed after every 30 minutes and the change in weight was plotted against time in Fig. 3.2. It was observed that the hydrogels obtained equivalent weight after about 24 hours. The swelling behavior of the hydrogels was not affected significantly by the presence of PAni. Hence the hydrogel composites can be used as and where hydrogels are used.

Table 3.2: Swelling studies of pure hydrogels and PAni/PAAm hydrogels

Sl. No.	Time(hours)	Swollen Weight of Pure Hydrogels			Swollen weight of PANi/PAAm Hydrogel		
		Sample 1	Sample 2	Sample 3	Sample 4	Sample 5	Sample 6
1	0.0	0.11	0.10	0.22	0.11	0.11	0.22
2	0.5	0.37	0.40	0.38	0.22	0.29	0.35
3	1.0	0.46	0.55	0.44	0.28	0.33	0.42
4	1.5	0.55	0.67	0.50	0.33	0.37	0.49
5	2.0	0.63	0.76	0.56	0.38	0.42	0.53
6	2.5	0.69	0.80	0.62	0.45	0.46	0.57
7	3.0	0.76	0.83	0.65	0.48	0.50	0.61
8	3.5	0.80	0.86	0.68	0.55	0.53	0.65
9	4.0	0.87	0.88	0.70	0.61	0.57	0.67
10	4.5	0.94	0.90	0.73	0.64	0.61	0.69
11	5.0	1.00	0.92	0.76	0.68	0.63	0.71
12	5.5	1.03	0.95	0.78	0.70	0.66	0.73
13	6.0	1.07	0.97	0.80	0.72	0.70	0.75
14	6.5	1.10	0.99	0.82	0.74	0.74	0.77
15	7.0	1.12	1.02	0.85	0.76	0.76	0.79
16	7.5	1.15	1.04	0.87	0.78	0.80	0.79
17	8.0	1.17	1.08	0.88	0.80	0.83	0.80
18	8.5	1.19	1.10	0.89	0.83	0.87	0.80
19	9.0	1.22	1.14	0.90	0.85	0.90	0.81
20	9.5	1.25	1.16	0.90	0.87	0.94	0.81
21	10.0	1.26	1.18	0.91	0.90	0.96	0.82
22	10.5	1.27	1.20	0.91	0.93	1.00	0.83
23	11.0	1.28	1.23	0.92	0.95	1.03	0.84
24	11.5	1.29	1.24	0.92	0.97	1.05	0.85
25	12.0	1.30	1.25	0.93	1.00	1.07	0.84
26	12.5	1.31	1.27	0.93	1.03	1.11	0.84
27	13.0	1.34	1.29	0.94	1.05	1.14	0.85
28	13.5	1.36	1.31	0.94	1.07	1.16	0.85
29	14.0	1.37	1.35	0.94	1.10	1.18	0.85
30	14.5	1.37	1.37	0.94	1.12	1.20	0.86
31	15.0	1.37	1.39	0.94	1.14	1.22	0.86
32	15.5	1.38	1.41	0.94	1.16	1.24	0.86
33	16.0	1.38	1.43	0.94	1.18	1.26	0.87
34	16.5	1.38	1.45	0.94	1.20	1.28	0.87
35	17.0	1.38	1.47	0.94	1.22	1.30	0.87
36	17.5	1.39	1.48	0.94	1.23	1.31	0.88
37	18.0	1.39	1.49	0.94	1.25	1.32	0.88
38	18.5	1.39	1.50	0.94	1.26	1.34	0.88
39	19.0	1.40	1.51	0.94	1.27	1.35	0.88
40	19.5	1.40	1.51	0.94	1.28	1.37	0.89
41	20.0	1.40	1.52	0.94	1.28	1.38	0.89
42	20.5	1.41	1.52	0.94	1.29	1.39	0.89
43	21.0	1.41	1.53	0.94	1.29	1.40	0.89
44	21.5	1.41	1.53	0.94	1.30	1.41	0.89
45	22.0	1.42	1.54	0.94	1.30	1.41	0.90
46	22.5	1.42	1.54	0.94	1.30	1.42	0.90
47	23.0	1.42	1.54	0.94	1.31	1.42	0.90
48	23.5	1.42	1.55	0.94	1.31	1.43	0.90
49	24.0	1.42	1.55	0.94	1.31	1.43	0.90

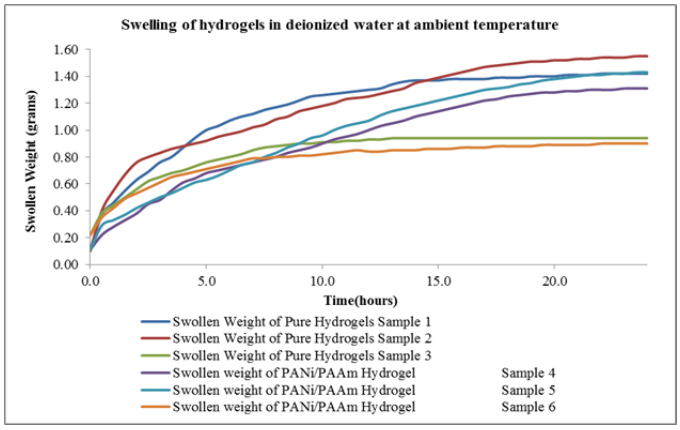

Fig. 3.2: Swelling of hydrogels in deionized water at ambient temperature

3.3 The percentage swelling of hydrogel composites.

The percentage of swelling of hydrogel composites was calculated from the following equation:

$$\%S = \frac{Mt - Mo}{Mo} \times 100$$

Where Mt is the mass of swollen hydrogel at time t and Mo is the mass of dry hydrogel at time, t= 0.

Table 3.3: Equilibrium swelling (%S) of hydrogel composites

Samples	Equilibrium Swelling Value(ESV)%
Pure Hydrogel 1	1190
Pure Hydrogel 2	1450
Pure Hydrogel 3	328
PAni/PAAm Hydrogel 4	1090
PAni/PAAm Hydrogel 5	1200
PAni/PAAm Hydrogel 6	310

3.4 Equilibrium water content of hydrogel composites

The equilibrium water content (EWC) was determined using the following procedure. Soluble hydrogel composites were dried at room temperature. The dried samples were weighed to constant weight (Ms). The samples were then immersed in a large amount of distilled water and allowed to swell till the weight becomes constant (Mo). The EWC of the hydrogel composites was determined from following equation:

$$EWC\% = \frac{Ms - Mo}{Ms} \times 100$$

Where Ms is the weight of the swollen gel at time t (equilibrium) and Mo is the weight of the dry gel at time, t=0.

Table 3.4: Equilibrium water content of hydrogel composites

Samples	Equilibrium water content (EWC)%
Pure Hydrogel 1	92.25
Pure Hydrogel 2	93.54
Pure Hydrogel 3	76.59
PAni/PAAm Hydrogel 4	91.60
PAni/PAAm Hydrogel 5	92.30
PAni/PAAm Hydrogel 6	75.56

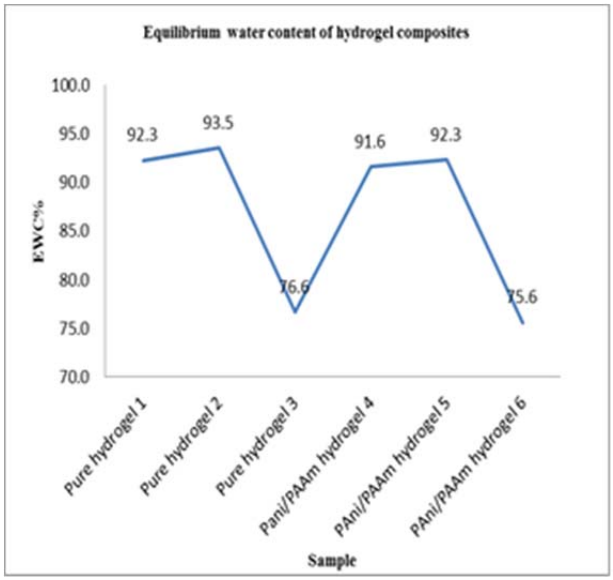

Fig. 3.3: Equilibrium water content of hydrogels

Equivalent water content is a measure of the swelling capacity of the hydrogels. Since polymerization of Aniline occurs in the already established network of Polyacrylamide, the presence of Polyaniline does not hamper the basic swelling characteristics of the hydrogel as reflected by the EWC percent. The presence of PAni will decrease the flexibility but the overall porosity of the matrix network is unaffected. The essential characteristic of this technique for producing hydrogel composite lies in the fact that Aniline and dopant enter the hydrogel network against concentration gradient and hence the dispersion is uniform.

4. CONCLUSION

Conducting hydrogel composites of PAni/Polyacrylamide were prepared by synthesizing Polyaniline inside the hydrogel matrix. From the interpretation of the results it may be concluded that the basic swelling properties are not hampered by the incorporation of conducting polymer PAni into PAAm hydrogel. Similarly, water retention properties of the hydrogel are not significantly affected. The presence of PAni in the PAAm network increases its stability. Hence such composites are an effective substance for application where conductivity

and swelling properties are required like drug release, tissue engineering biosensors.

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