Hydrogel Membranes Based on Gum Tragacanth with Tunable Structures and Properties. II. Comprehensive Characterization of the Swelling Behavior

A. Kiani, H. Asempour

Polymer Engineering Department, Amirkabir University of Technology, Tehran 1591634311, Iran

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ABSTRACT: Hydrogel membranes based on gum tragacanth (GT), with a wide range of crosslinking densities (0.83–23.87 $\times$ 10^{-6} mol/cm^3) and swelling capacities (120–1580 g/g), were prepared with the aid of heat alone and four crosslinking reagents according to a procedure reported in our previous article. In this article, we report on the comprehensive evaluation of the swelling behavior of the prepared hydrogel membranes. These include the swelling kinetics, swelling behavior in terms of the pH and ionic strength of the swelling media, water absorbency under load (AUL), swelling as a function of sample size, deswelling behavior, and successive swelling and deswelling cycles. The swelling kinetics of the hydrogel membranes with higher crosslinking densities and lower swelling equilibria were closer to those of the case II transport mechanism; this is desirable in the field of drug-delivery systems (DDSs). Furthermore, higher crosslinking densities, and hence stronger structures, of the membranes led to higher AUL values and more stable performances in the successive swelling and deswelling cycles. Moreover, because of the anionic nature of the gum, the prepared GT hydrogels showed pH- and ionic-strength-dependent swelling behavior. All of these results reveal that GT could be a promising material for the preparation of hydrogel membranes for applications in the fields of DDS, tissue engineering, and the separation of small molecules. © 2012 Wiley Periodicals, Inc. J. Appl. Polym. Sci. 000: 000–000, 2012

Key words: biopolymers; films; hydrogels; membranes; polysaccharides

INTRODUCTION

According to Peppas et al.,[1] *hydrogels* are defined as "three-dimensional, hydrophilic, polymeric networks capable of imbibing large amounts of water or biological fluids" in the range of 10–1000 times their weights.[2] These materials have porous structures with pore sizes (ξ's) from nanometers to micrometers.[3] Hydrogels with neutral or ionic natures[3] can be derived from naturally occurring or synthesized polymers.[4] Nowadays, hydrogels with diverse microstructures are prepared for special applications in the fields of agriculture,[5] food and cosmetics,[3,6] drug-delivery systems (DDSs),[4,7–9] and tissue engineering.[4,10,11]

The most important parameters specifying the structure and characteristics of a hydrogel, and hence its applications, are the polymer volume fraction in the swollen state, effective molecular weight of the polymeric chains between crosslinks (M_c), and its network mesh or ξ. These parameters represent the swelling capacity, degree of crosslinking, and network porosity of the hydrogel, respectively.[3,4] Therefore, precise measurement and adjustment of these characteristics are of crucial importance to the recognition of a hydrogel structure for its subsequent applications.

The effective molecular weight of the polymeric chains between crosslinks can be obtained with the aid of rheomechanical measurements.[12] ξ can be determined indirectly with rubber-elasticity measurements and equilibrium swelling (Q_∞) experiments.[3] This parameter controls the physical properties of a hydrogel, especially its mechanical strength, degradability, and diffusivity behavior.[4] The polymer volume fraction in the swollen state is usually determined through Q_∞ measurements and is influenced by the hydrogel structure, its particle size, and environmental factors, such as temperature, pH, and the ionic strength of the swelling media.[3,13–15] However, to examine the suitability of a hydrogel for a specific application, one should consider its kinetics of absorption,[16,17] deswelling behavior,[18,19] swelling and deswelling behavior,[18,20] and its water absorbency under load (AUL).[21,22]

Gum tragacanth (GT) is a natural polysaccharide produced as a dried exudation from the stems and

Correspondence to: H. Asempour (asempour@aut.ac.ir).

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TABLE I
Q_∞, Diffusion Constant, and Crosslinking Density (n_x) Values of Hydrogels Belonging to the H–C, Gl–C, EG–C, and TEG–C Series Obtained under the Specified Experimental Design Conditions

	% w/v	T (°C)	t (h)	Q_∞ (g/g)	$n_x \times 10^6$ 2(mol/cm^3)	n	$K \times 100$	AUL (g/g)
H–C sample	[GT]							
H–C1	0.5	57	2	606.9	4.1	0.901	11.35	—
H–C2	0.5	77	6	403.8	8.09	0.923	12.42	—
H–C3	0.5	97	24	368.2	9.45	0.968	16.73	—
H–C4	0.75	57	24	321.5	11.85	0.946	15.59	—
H–C5	0.75	77	2	544.7	4.91	0.845	8.06	—
H–C6	0.75	97	6	441.8	6.97	0.989	18.24	—
H–C7	1	57	6	1582.1	0.83	0.619	3.57	—
H–C8	1	77	24	1054.8	1.63	0.739	4.94	—
H–C9	1	97	2	1390.4	1.03	0.653	4.91	—
Gl–C sample	$[OH]_{Gl}/[COOH]_{GT}$							
Gl–C1	0.33	57	2	967.6	3.01	0.671	6.92	30.2
Gl–C2	0.33	77	6	528.2	8.27	0.775	8.68	51.9
Gl–C3	0.33	97	24	307.8	20.36	0.951	14.19	60.9
Gl–C4	0.5	57	24	313.9	19.7	0.985	15.18	58.9
Gl–C5	0.5	77	2	602.1	6.64	0.775	8.11	39.0
Gl–C6	0.5	97	6	550.7	7.71	0.758	8.45	42.0
Gl–C7	1.0	57	6	434.9	11.43	0.883	10.37	57.4
Gl–C8	1.0	77	24	302.1	21.01	0.924	12.92	67.0
Gl–C9	1.0	97	2	666.7	5.6	0.827	9.89	35.3
EG–C sample	$[OH]_{EG}/[COOH]_{GT}$							
EG–C1	0.25	57	2	657.1	3.59	0.746	9.48	—
EG–C2	0.25	77	6	423.4	7.48	0.951	17.94	—
EG–C3	0.25	97	24	368.5	9.43	0.931	13.48	—
EG–C4	0.5	57	24	409	7.93	0.988	20.22	—
EG–C5	0.5	77	2	622.5	3.93	0.817	6.98	—
EG–C6	0.5	97	6	540.2	4.98	0.861	11.84	—
EG–C7	1.0	57	6	495	5.76	0.973	17.16	—
EG–C8	1.0	77	24	326.4	11.55	0.962	17.45	—
EG–C9	1.0	97	2	543.4	4.93	0.924	11.99	—
TEG–C sample	$[OH]_{TEG}/[COOH]_{GT}$							
TEG–C1	0.5	77	6	649.1	3.67	0.801	8.46	—
TEG–C2	0.5	97	24	443.4	6.93	0.933	11.69	—
TEG–C3	1.0	77	24	935.4	1.99	0.706	5.97	—
TEG–C4	1.0	97	6	380.2	8.96	0.982	12.46	—

branches of *Astragalus gummifer* and other Asiatic species of Astragalus. The gum is obtained in two different forms, ribbons and flakes.[23] GT is composed of a complex mixture of heterogeneous polysaccharides and occurs as a slightly acidic calcium, magnesium, and potassium salt.[24] This biopolymer consists of two major fractions: a water-soluble fraction called *tragacanthin* and an insoluble but swellable fraction called *bassorin*.[23,25] The physical, rheological, and solution properties of GT with unknown and known botanical sources have also been the subject of a number of studies.[23–27]

In our previous article,[28] we reported the preparation of hydrogel membranes based on GT with different swelling capacities (120–1580 g/g) and crosslinking densities (0.83–23.87 $\times$ 10^{-6} mol/cm^3). The hydrogel samples were made with the aid of heat alone and with four crosslinking reagents, including glycerin (Gl), ethylene glycol (EG), triethylene glycol (TEG), and glutaraldehyde (Glt). The microstructures and Q_∞ values of the prepared hydrogel membranes based on either the Taguchi method of experimental design or full factorial design were also reported. The initial concentration of the GT solution is denoted by [GT], the molar ratio of the functional groups of the crosslinking reagents to that of GT is denoted by $[X]_Y/[X']_{GT}$, and the crosslinking reaction temperature and time are denoted by T and t, respectively; these were taken as variables in our previous research work. In Tables I and II, a brief description of the variables and their levels as well as the Q_∞ values and crosslinking densities obtained for the whole samples are provided.

This work is a continuation of our previous work[28] and describes comprehensive studies on the swelling behavior of the prepared hydrogels, including the swelling kinetics in distilled water and different aqueous media with different pH values and ionic strengths, AUL, deswelling behavior, and successive swelling and deswelling cycles of some of the hydrogel samples.

TABLE II
Q_∞, Diffusion Constant, and Crosslinking Density (n_x) Values of Hydrogels Belonging to the Glt–C Series Obtained under the Specified Experimental Design Conditions

Glt–C sample	[Glt] (μmol/100 mL of 1% w/v GT solution)	T (°C)	Q_∞ (g/g)	$n_x \times 10^6$ (mol/cm^3)	n	$K \times 100$
Glt–C1	3.72	75	186.2	12.57	0.836	9.68
Glt–C2	3.72	80	179.2	12.98	0.974	13.04
Glt–C3	3.72	85	128.9	22.85	0.981	13.41
Glt–C4	3.72	90	160	15.19	0.989	12.01
Glt–C5	7.44	75	172.2	13.31	0.922	10.28
Glt–C6	7.44	80	125.6	23.62	0.953	12.04
Glt–C7	7.44	85	131.1	21.59	0.915	10.61
Glt–C8	7.44	90	120.3	26.41	0.867	9.72
Glt–C9	18.6	75	183.8	12.86	0.763	8.79
Glt–C10	18.6	80	165.5	14.33	0.892	10.38
Glt–C11	18.6	85	129.3	22.6	0.953	11.58
Glt–C12	18.6	90	143.2	17.74	0.931	11.04
Glt–C13	37.2	75	185.1	12.64	0.793	8.75
Glt–C14	37.2	80	189.9	12.36	0.731	7.32
Glt–C15	37.2	85	124	23.87	0.752	9.51
Glt–C16	37.2	90	176.8	13.06	0.842	9.39
Glt–C17	55.8	75	262.5	6.91	0.685	6.18
Glt–C18	55.8	80	233.4	8.44	0.656	6.58
Glt–C19	55.8	85	213.2	9.72	0.706	7.29
Glt–C20	55.8	90	195	11.38	0.635	7.41

EXPERIMENTAL

Thin films of GT hydrogels were prepared according to the methods described in our previous article[28] by heat treatment of the gum (H–C series) and by reaction with different crosslinking reagents (Gl, EG, TEG, and Glt) to obtain Gl–C, EG–C, TEG–C, and Glt–C series, respectively. Analytical grades of HCl, KOH, NaCl, $CaCl_2$, and Na_2SO_4 and all of the other chemicals used in this study were obtained from Merck Co. (Germany).

The swelling behavior of each of the hydrogel films in distilled water was studied with the well-known tea-bag method,[28] and the kinetics of swelling of the gel was obtained with the following equation:[29]

$$Q_t = \frac{M_t - M_o}{M_o} \tag{1}$$

where M_0 is the initial weight of the dry specimen, M_t is the weight of swollen specimen, and Q_t is the swelling ratio of the specimen at time t. The Q_∞ value of each hydrogel sample was taken as the value of Q_t when it reached a constant value. All reported values are the average of three measurements on a sample.

To measure the swelling behavior of the hydrogels in some electrolyte solutions, a number of aqueous solutions of NaCl, $CaCl_2$, and Na_2SO_4 at different concentrations (0.00005, 0.001, 0.01, and 0.1M) were prepared with distilled water.

To assess the pH dependency of Q_∞ of the hydrogel samples, solutions with different pH values were prepared by the dissolution of HCl and KOH at various concentrations in distilled water; these solutions were used as the swelling media. The swelling values were reported after equilibrium was reached, at around 2 h.

The swelling behavior of the hydrogel samples under load (AUL) was examined with the aid of the simple apparatus depicted in Figure 1.

To perform this experiment, about 0.5 g of the film specimen (2×2 mm^2) was spread uniformly on the surface of the pervious sample holder. The Teflon-made piston was placed over the samples in the sample holder, and the load on the samples was adjusted to 1.5 kPa. The AUL value of the specimen was measured from the displacement of the Teflon-made piston caused by the swollen hydrogel.

To evaluate the efficiency of each swollen hydrogel sample in releasing its imbibed water under ambient conditions, the weighed sample ($\sim$ 0.5 g) was first inserted in distilled water (1000 mL) and allowed to swell and reach to its Q_∞ point. The swollen sample was then spread on the surface of a clean polyethylene film and weighed at specific time intervals. The water release rate of the sample was calculated with the following equation:[19]

$$\%\mathrm{RW} = \frac{M_t - M_o}{M_\infty - M_o} \times 100 \tag{2}$$

where %RW represents the percentage of water remaining within the hydrogel at time t, M_t is the hydrogel weight at time t, M_∞ is the hydrogel weight at the equilibrium swelling point, and M_0 is the weight of the initial dry gel specimen.

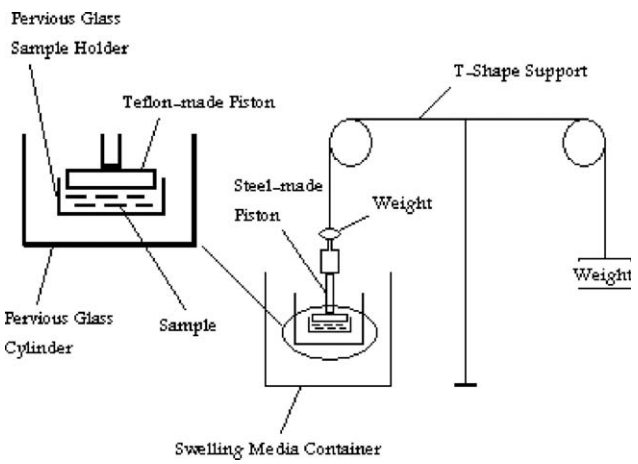

Figure 1 Schematic of the setup used for AUL measurements.

To investigate the behavior of the hydrogel samples on repeating swelling and deswelling cycles, the dry gel specimen was first allowed to reach its Q_∞ in distilled water and then dried in a vacuum oven (37°C, 100 mbar) to a constant weight. This procedure was successively repeated three times for the same gel specimen, and the changes in the Q_∞ and swelling kinetics values are reported.

RESULTS AND DISCUSSION

One of the main characteristics of each hydrogel system for a specific application is its water absorption rate (kinetics).[30] In general, the penetration of each small molecule (permeant), such as water, into a glassy polymeric network can occur with different mechanisms. These are classified into three categories on the basis of the relative importance of the diffusion to the chain relaxation: Fickian diffusion ($n = 0.5$), anomalous or non-Fickian diffusion ($0.5 < n < 1$), and case II transport ($n = 1$).[31,32] To determine whether the absorption is diffusion-controlled, relaxation-controlled, or a combination of these two, one can exploit the absorption kinetics data together with the correlation proposed by Ritger and Peppas[33] as follows:

$$\frac{M_t}{M_\infty} = Kt^n \tag{3}$$

where n and K are constants indicative of the nature of the absorption mechanism, M_t is the amount of water absorbed at time t, and M_∞ is the amount of absorbed water at equilibrium swelling. The values of n and K and Q_∞ data[28] determined for all series of hydrogel samples prepared in this study are presented in Tables I and II. It was observed that the n values for all of the series of hydrogel samples were in the range 0.5–1; this indicated an anomalous or non-Fickian mechanism as the result of the simultaneous occurrence of Fickian diffusion and relaxation of polymeric chains. However, for the most of the hydrogel samples, the n values are very close to unity, that is, case II transport. Soppirnath and Aminabhavi[34] found that the water transport mechanism in Glt-crosslinked hydrogels of guar-*g*-acrylamide shifted from Fickian to non-Fickian with an increase in the crosslinking densities of the hydrogels. They also found that the crosslinking density increased and the water uptake decreased as a function of the Glt level in the prepared hydrogels. On the other hand, a comparison of the Q_∞ values and the corresponding n values in Tables I and II indicated that in all of series of hydrogel samples, the n values became closer to unity for those hydrogels with lower values of Q_∞. This was due to the lower mobility and greater relaxation times of the hydrogel chains with denser networks.

The other finding from the swelling experiments was that the rate of water absorption by each hydrogel sample was directly dependent on its Q_∞ value so that the samples with higher absorption capacities absorbed water at a higher rate. Thus, each parameter that caused an increase in the crosslinking density or a decrease in the Q_∞ value of a hydrogel sample led to a reduction in its water absorption rate. Table II shows that the hydrogel samples belonging to the Glt–C series imbibed the least amount of water because they contained fewer ionic groups than the other hydrogel series[28] and they had higher crosslinking densities.

The GT molecule contains a large number of carboxylic acid and carboxylate groups. Therefore, the water absorptivity of the hydrogel samples prepared based on this natural gum not only depended on the thermodynamic affinity with the permeant molecule and elastic retractive forces but also on the effect of Donnan equilibrium.[3] This effect reflects the difference in ionic contents within the hydrogel network and the external medium; that is, the ionic strength of the external medium can affect the Donnan equilibrium. Therefore, it is expected that increasing the concentration of ions and their valences in a swelling medium would cause a decrease in the chemical potential difference of two phases. Therefore, water diffusion into the hydrogel network is decreased.[13–15]

The results obtained from determination of Q_∞ in aqueous solutions of NaCl and $CaCl_2$ showed the Q_∞ values of all of the investigated samples (H–C9, Gl–C5, EG–C5, and TEG–C3 samples) decreased exponentially with increasing concentration of the salt. The variation of Q_∞ in terms of the NaCl and $CaCl_2$ concentrations in water for the H–C9 sample are depicted in Figure 2. The swelling capacities of the hydrogel samples in NaCl solutions were much higher than those in $CaCl_2$ solutions having similar concentrations. This behavior was attributed to the greater number of ions created by $CaCl_2$ dissociation in the media and also to the ability of Ca^{2+} to replace the monovalence ions previously existing in the hydrogel and, thus, establish physical crosslinks and hydrogel collapse.[24,35] Mohammadnia et al.[36] prepared a semisynthetic hydrogel from GT by polyacrylonitrile grafting onto GT chains and evaluated its swelling behavior as a function of the ionic strength of the swelling media. Q_∞ of the synthesized hydrogel in distilled water was 307 g/g, whereas in 0.15*M* NaCl and $CaCl_2$, it was equal to 65 and 3.5 g/g, respectively; this was in accordance with our obtained results.

The Q_∞ values of the studied hydrogel samples in 0.001*M* solutions of NaCl, $CaCl_2$, and Na_2SO_4 are demonstrated in Figure 3.

As it was observed, the Q_∞ values for all of the hydrogel samples in the NaCl solution were the

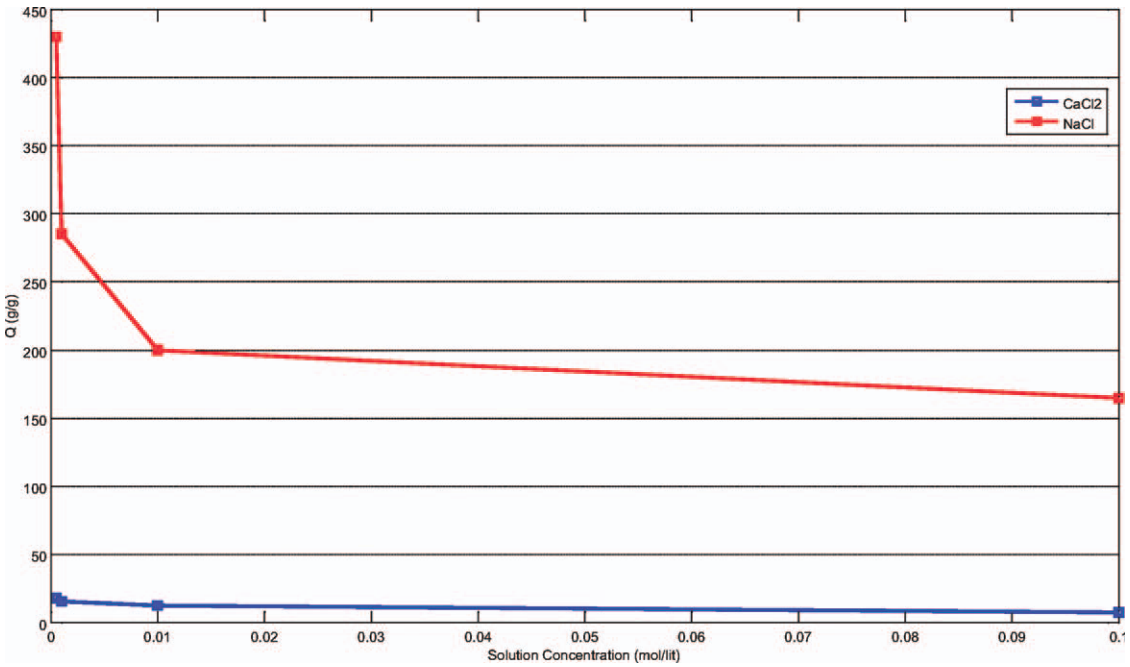

Figure 2 Variation of the Q_∞ values of the H–C9 sample in terms of the concentrations of the NaCl and $CaCl_2$ solutions. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

greatest, and those in the $CaCl_2$ solution were the lowest. The lower swelling capacities of the hydrogels in Na_2SO_4 solution compared to those in the NaCl solution were attributed to the higher number of Na^+ ions created by the Na_2SO_4 salt and also the higher valence of SO_4^{2-} ions compared to Cl^- ions, which hindered the water absorption by the hydrogel samples.

The swelling of the hydrogel samples in 0.001*M* solutions of NaCl and Na_2SO_4 versus time initially increased rapidly and then reached a constant value and became independent of time. However, the swelling behavior of the samples in $CaCl_2$ solution was found to pass through a maximum with the extension of time. This behavior was attributed to the replacement of monovalence ions previously existing in the hydrogel samples by bivalent Ca^{2+} present in the external solution. The swelling behavior trends for the Gl–C5 sample are shown in Figure 4.

The results obtained from the measurement of the reswelling capacities (Q_∞^{re}'s) of the hydrogel samples in 0.001*M* NaCl solution revealed that the Q_∞^{re} values of the samples increased a little bit in comparison

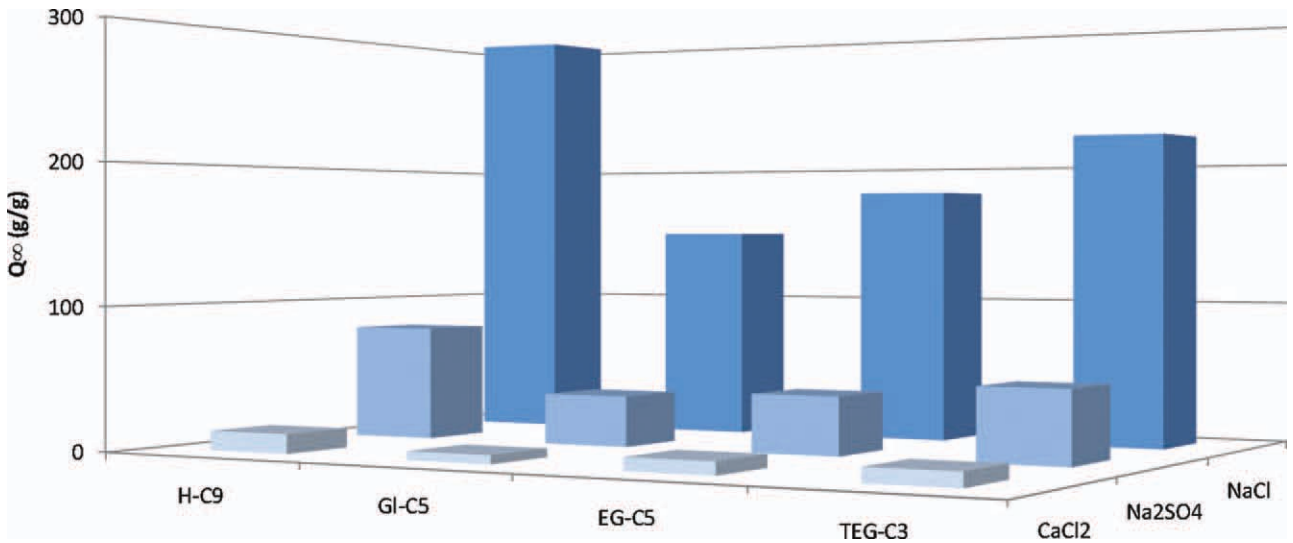

Figure 3 Effect of 0.001 molar solutions of NaCl, $CaCl_2$, and Na_2SO_4 on the swelling capacities of some of the hydrogel samples. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

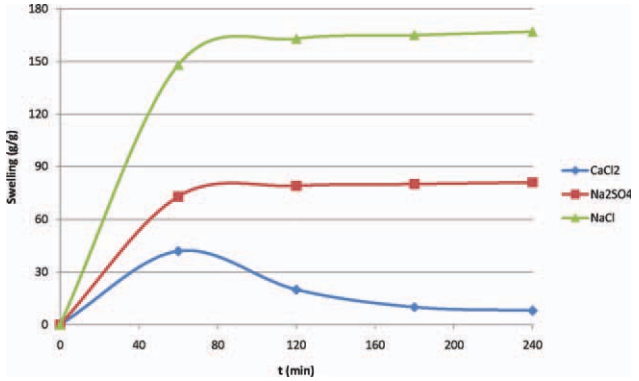

Figure 4 Swelling kinetics of the Gl–C5 sample hydrogel in solutions of NaCl, Na_2SO_4, and $CaCl_2$ at a concentration of 0.001*M*. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

with their initial swelling capacities, most probably because of the breakage (hydrolysis) of the weak bonds present in the hydrogels. However, the behavior of the samples in $CaCl_2$ solution was quite different so that the Q_∞^{re} value became lower than the Q_∞ value. This observation was attributed to the aforementioned ion-exchange phenomenon occurring in the first run of the swelling and, hence, establishment of new physical crosslinks in the samples, which dramatically reduced their Q_∞^{re} values.

It was expected that the swelling behavior of the GT anionic hydrogels would be pH dependent.[3,37] Thereby, the swelling behavior of a number of the hydrogel samples was investigated within the range $1.5 < pH < 12.30$. The results obtained from this experiment are shown in Figure 5.

It was seen that the swelling capacities of the hydrogel samples at low pH values were quite low, most probably because of the decreasing ionic dissociation degree of the carboxylate groups present in the samples.[15,27,38] By increasing the pH of the external media to a nominal value of 3.5, the swelling capacities of the hydrogel samples increased because of the increase in the ionic dissociation of the carboxylic acid groups. At pH values between 3.5 and 9, the swelling capacities of the hydrogel samples almost remained constant, and at a pH value of 9.5, a rather sharp increase in the swelling capacity of the hydrogels was observed. This sharp increase could be attributed to the partial hydrolysis of the polymeric main chains and the crosslink bonds, which, hence, decreased the crosslinking densities of the samples' networks. It is noteworthy that at pH values higher than 10, the crosslinked GT samples degraded rapidly, and their degradation products became soluble in the media. The swelling kinetics of the hydrogel samples at various pH values were found to be the highest at pH 10 and the lowest at pH 2.65. George and Abraham[39] observed the same pH-dependent behavior in alginate–guar systems crosslinked with Glt. They found that the anionic nature of alginate led to an increase in swelling of the hydrogel systems with pH increases in the swelling medium from 1.2 to 7.4. It was also seen that alginate dissolved easily in basic media.

The swelling capacity of a hydrogel under a load is a measure of its mechanical strength and stability.[40] In many applications, a hydrogel is applied under a specific pressure or load.[41] Therefore, the swelling behaviors of the hydrogel samples belonging to the Gl–C series were studied under a load (1.5 KPa). The results obtained from this study are given in Table I. It was observed that the Q_∞ values of the samples under the load (AUL) were much lower than the corresponding values under load-free conditions. Furthermore, the hydrogel samples with higher crosslinking densities showed higher AUL values. This result was exactly the opposite of those of the swelling behavior of the hydrogel samples under load-free conditions. Therefore, each parameter that increased the crosslinking density of the hydrogel positively affected its swelling capacity under load. Thus, the strength of the GT hydrogel sample was of crucial importance to its AUL behavior, and the crosslinking density of the GT hydrogel needed to be so optimized to create a gel with an appropriate modulus to tolerate an external pressure without having a negative effect on its swelling capacity.

To examine the effect of the hydrogel sample size on its swelling capacity, three specimens of TEG–C2 hydrogels with similar thicknesses but different dimensions (2×2, 5×5, and 15×15 mm^2) were prepared and subjected to swelling tests in distilled water. The result obtained is shown in Figure 6. As shown, the Q_∞ value of the gel increased with decreasing specimen dimensions. However, the gel-blocking phenomenon occurring with the reduction

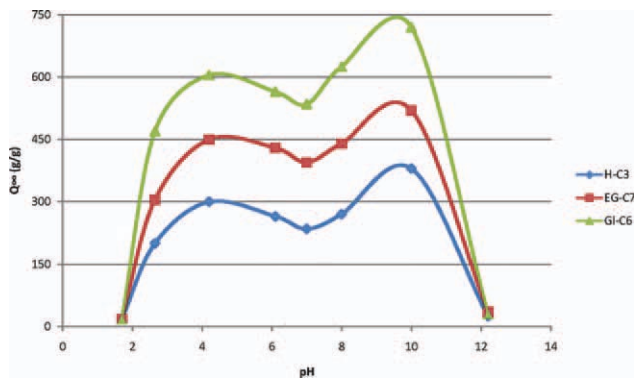

Figure 5 Variation of the swelling capacities of some of the hydrogel samples as a function of the pH value of the swelling medium. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

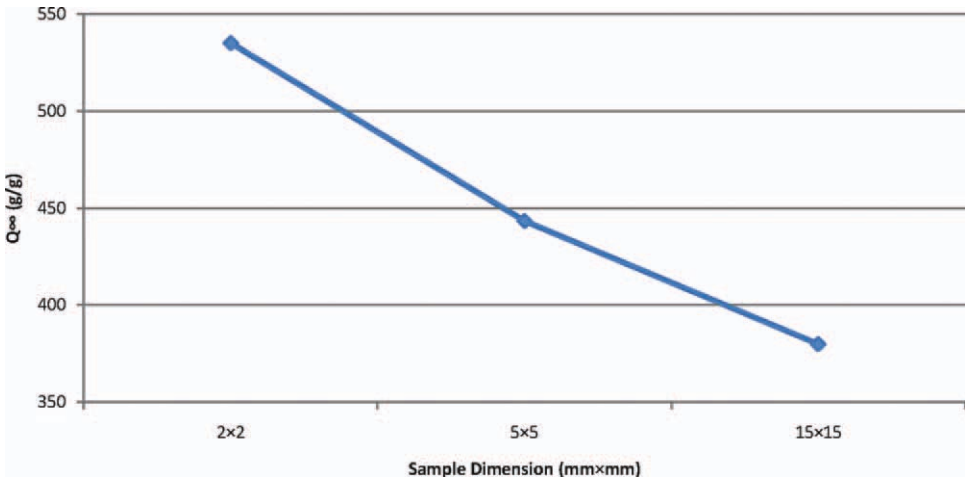

Figure 6 Effect of the sample size on the swelling capacity of the TEG–C2 hydrogel. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

of the sample size had to be taken into consideration as it reduced the swelling rate and capacity.[42]

Another characteristic of hydrogels is their ability to retain water in their three-dimensional networks for a rather extended time. In this study, the water-release behavior of different series of the hydrogel samples, except the Glt–C series, were examined. It was observed that the higher crosslinking densities of the samples caused acceleration of the water release rate from the swollen hydrogel because of the higher retractive elastic forces. Therefore, in these series of hydrogels, each parameter that increased the crosslinking density of the hydrogel practically increased its water release rate.

The behavior of a hydrogel with multiple swelling and deswelling cycles is very important for some applications, especially in the case of superabsorbent materials.[43] Therefore, all of the hydrogel samples of the Gl–C and Glt–C series were subjected to three successive swelling and deswelling cycles. The results show that the swelling capacities in each cycle were higher compared to the previous cycle; this was attributed to the breakdown of a number of chemical bonds through hydrolysis in each cycle. Moreover, we observed that the difference in the swelling values of the hydrogel samples in two consecutive cycles was higher for the hydrogel with a lower crosslinking density. Therefore, we concluded that in any application where durability and steady performance of a GT hydrogel in successive cycles of swelling and deswelling is required, the choice of a hydrogel with a low crosslinking density is not recommended.

CONCLUSIONS

The hydrogels prepared based on GT were found to have anomalous or non-Fickian swelling behavior. When Q_∞ of the samples decreased, their swelling kinetics approached the case II transport mechanism. The water absorption rate of the hydrogel samples was found to be dependent on their Q_∞ values so that the swelling rate of the samples increased with increasing Q_∞ value.

The prepared GT hydrogels also showed that their swelling behavior was completely dependent on the pH and ionic strength of the swelling media because of the ionic nature of the initial gum. Furthermore, the size and valence of the ions present in the swelling media were determining parameters in the swelling behavior of the investigated hydrogels.

The results obtained from the measurement of the swelling capacities of the hydrogel samples under a load (AUL) revealed that the AUL values were quite lower in comparison with the corresponding Q_∞ values in the load-free state. The AUL values were found to be strongly dependent on the gel strength in a way that with increasing crosslinking density, the strength of the gel increased, and a higher AUL value was achieved. This behavior was exactly the opposite that of load-free swelling.

We also perceived that the swelling capacity of the prepared hydrogels increased with decreasing sample dimensions.

The deswelling rate of the swollen hydrogel samples were improved with increases in their crosslinking densities. Furthermore, a study of multiple swelling and deswelling cycles of the hydrogel samples showed that those samples possessing higher crosslinking densities would function more satisfactorily in applications where a steady performance of the material in successive cycles of swelling and deswelling is required.

As a whole, GT seems to be a promising natural material for the preparation of hydrogel membranes with diverse microstructures and properties, despite

its complex structure. This diversity of properties would be useful for practical applications of these hydrogels in the fields such as DDSs, tissue engineering, and separating membranes.

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