Journal of Polymer Science and Technology 6(2) 2021:27-38
ISSN: 2550-1917

EFFECT OF CALCIUM CHLORIDE WEIGHT PERCENTAGE ON THE DEVELOPMENT OF *IOTA*-CARRAGEENAN BASED HYDROGEL

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ABSTRACT

Hydrogels are three-dimensional networks of hydrophilic polymers that absorb water but are not soluble in it. In this work, calcium chloride ($CaCl_2$) was used as a crosslinking agent to enhance the mechanical and swelling characteristics of *iota*-carrageenan (*i*-carrageenan) based hydrogels. The influence of $CaCl_2$ weight percentage, the effect of pH, and time were all used to determine the swelling effectiveness of the hydrogel film. It was found that the hydrogel film with 9 wt.% of $CaCl_2$ (ICC9) had the highest swelling degree with 127.89 g/g. In comparison to other hydrogel films, ICC9 was highly stable in deionized (DI) water for the longest duration (27 hours) before completely being dissolved. After all, the highest degree of swelling for all the samples occurs at pH 6.01. This showed that the presence of $CaCl_2$ in the *i*-carrageenan hydrogel film enhanced stability in water.

Keywords: Calcium chloride; degree of swelling; hydrogel; *iota*-carrageenan.

INTRODUCTION

Hydrogels are polymeric polymers that can absorb a lot of water without easily dissolved in it. The presence of functional groups such as hydroxyl, carboxyl, and sulphate on polymer chains affects the water absorption rate of hydrogels [1]. Most hydrogels are now manufactured from natural sources, and *iota*-carrageenan (*i*-carrageenan) is one of the primary components in hydrogel manufacturing, with applications in biology, agriculture, hygiene goods, and medicines [2]. The hydrogel made of *i*-carrageenan absorbs a lot of water and is biocompatible and non-toxic [3]. *i*-carrageenan is an endless natural polymer that may be derived from the *Eucheuma Spinosum* seaweed species.

The capacity of *i*-carrageenan to absorb water was demonstrated in a study done by Li et al. [4]. The *i*-carrageenan/chitosan/gelatine (CCG) system is the subject of their research. The researchers discovered that using *i*-carrageenan in the CCG system reduced swelling from 10.56 ± 0.07 to 19.46 ± 0.68. Electrostatic repulsion between sulphate groups in the *i*-carrageenan structure can assist to increase the space between the polymers and therefore the degree of swelling. Hydrogels based on poly(vinyl alcohol)/*i*-carrageenan (PC) have been utilised for drug release vehicles, according to Croitoru et al. [5]. They chose *i*-carrageenan over *kappa*-carrageenan (*k*-carrageenan) because *i*-carrageenan has two sulphate groups, OSO_3^- per galactan structural unit, which improves physical crosslinking and offers more active binding sites in the hydrogel matrix.

Although hydrogels have been widely used in a variety of fields, there have been few investigations into their stability in aqueous solutions. Nur'Ain Nabiela [6] managed to prepare *k*-carrageenan-based hydrogel at a temperature of 30 °C and a weight of 1.0 g that could endure for up to 475 minutes. The hydrogel then entirely dissolved in water after 475 minutes. When compared to the findings of Al-Mubaddel et al. [7], this capability was deemed low. Hydrogel films based on chitosan/polyacrylonitrile blends were employed in their investigation, which can endure up to 2880 minutes in water.

As a result, the influence of calcium chloride ($CaCl_2$) concentration on the mechanical characteristics and degree of swelling of *i*-carrageenan-based hydrogels was investigated in this work. Afifah Iswara Aji et al. [8] conducted a study that led to this suggestion. They observed that adding calcium ions (Ca^{2+}) to the *i*-carrageenan chain improves mechanical characteristics and hydrogel durability in DI water. Their research focuses on the application of *i*-carrageenan in the food packaging industry. The development of an *i*-carrageenan/$CaCl_2$ hydrogel film and the influence of weight percentage of $CaCl_2$, pH, and time on its capacity to swell were the focus of this study.

EXPERIMENTAL

Preparation of i-carrageenan based hydrogels

i-carrageenan based hydrogel was prepared according to the report by Nur'Ain Nabiela [6]. However, in this study, *k*-carrageenan was replaced with *i*-carrageenan. For 8 hours at 80 °C, 1 g of *i*-carrageenan was dissolved in 45 mL of DI water. The solution was then placed onto a separate petri dish and allowed to evaporate for 2 days in a fume hood. After that, it was dried on a hot plate for 1 day at 30 °C to obtain the *i*-carrageenan hydrogel film. I0 was assigned to the hydrogel film samples.

Preparation of i-carrageenan/$CaCl_2$ based hydrogels

According to the report by Nur' Ain Nabiela [6], *i*-carrageenan with various concentrations of $CaCl_2$ based hydrogel was also produced. 1 g of *i*-carrageenan was dissolved in 45 mL of DI for 8 hours at 80 °C. $CaCl_2$ solutions of various concentrations (1 wt.%, 3 wt.%, 5 wt.%, 7 wt.%, and 9 wt.%) were produced simultaneously for 30 minutes with stirring. After that, the $CaCl_2$ solution was added to the i-carrageenan solution, and it was stirred. Each subsequent solution was placed into a separate petri dish and allowed to evaporate for 2 days in a fume hood. After that, it was dried for 1 day on a hot plate at 30 °C to form an *i*-carrageenan/$CaCl_2$ hydrogel film. For 1 wt.%, 3 wt.%, 5 wt.%, 7 wt.%, and 9 wt.% of $CaCl_2$, the hydrogel film samples were assigned as ICC1, ICC3, ICC5, ICC7, and ICC9.

Determination of the point of zero charge (pH_{pzc}) for i-carrageenan/$CaCl_2$ hydrogel

The pH value at which the charge at the surface of the hydrogel film is zero was calculated using the value of the point of zero charge (pzc). pH_{pzc} was determined using the solid addition method. In a beaker, 120 mL of KNO_3 (0.1 M) solution was prepared. To achieve the required acidic and alkaline pH levels, 0.1 M HCl or 0.1 M NaOH solutions were added to the 0.1 M KNO_3 solution. The hydrogel films were then submerged in a pH solution that had been prepared. Allow 30 minutes for the mixture to interact before stirring at 200 rpm. A pH meter was used to test and record the final pH after removing the hydrogel film from the solution. pH_i was plotted against the difference between initial and final pH ($pH = pH_i - pH_f$). The pH_{pzc} value was determined by observing the point on the x-axis where ΔpH is 0.

Swelling test

a) Effect of weight percentage of calcium chloride

According to the Nur' Ain Nabiela [6] study, a 1 cm x 1 cm film of *i*-carrageenan hydrogel was

Journal of Polymer Science and Technology 6(2) 2021:27-38
ISSN: 2550-1917
submerged in DI water for 7 hours 55 minutes in the presence of various weight percentages of $CaCl_2$. The hydrogel films were then weighed after being removed from the DI water. The following equation was used to determine the degree of swelling:

$$Degree\ of\ Swelling\ (\frac{g}{g}) = \frac{W_a - W_b}{W_b} \quad (1)$$

where W_a and W_b are the weight of hydrogel before and after the swelling test.

b) Effect of pH
A film of *i*-carrageenan hydrogel approximately 1 cm x 1 cm was prepared with various weight percentages of $CaCl_2$ and submerged in a different pH solution at the same time. 2.85, 5.45, 6.01, 10.64, and 11.83 were the pH values used. The solution was made by diluting DI water with 0.1 M HCl or 0.1 M NaOH solution. For the swelling test, each type of hydrogel film was submerged in five different pH solutions for 7 hours and 55 minutes. After the swelling test was completed, the hydrogel film was removed from the pH solution and weighed. The degree of swelling was calculated by using equation (1).

c) Effect of time
A 1 cm × 1 cm sheet of *i*-carrageenan hydrogel with various weight percentages of $CaCl_2$ was submerged in DI water. If the hydrogel film did not disintegrate after 5 hours, the time increment was raised every 5 hours until it dissolved. 12 hours, 17 hours, 22 hours, 27 hours, and 32 hours of immersion were used. The films were removed and weighed. The degree of swelling was calculated by equation (1).

RESULTS AND DISCUSSION

Physical observation
Fig. 1 shows the state of the hydrogel film before and after swelling for 7 hours and 55 minutes. The hydrogel film I0 degraded entirely in DI water, as can be seen. Moreover, as the percentage of $CaCl_2$ in the hydrogel film increased, so does the hydrogel film's ability to maintain its form. This proves that the addition of Ca^{2+} ions has formed a crosslink with the $–OSO_3^-$ group and this has produced a hard hydrogel with better mechanical properties [17].

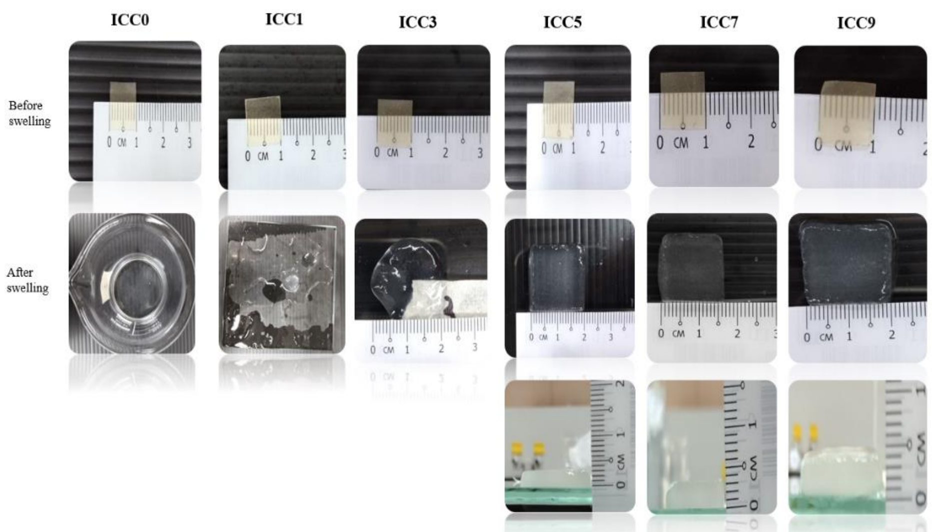


Figure 1 Physical examination of *i*-carrageenan-based hydrogel films at various $CaCl_2$ percentages before and after swelling

Determination of the point of zero charge (pH_{pzc})

The pH_{pzc} value for the I0 hydrogel film was 6.55, as shown in Fig. 2 (a). Furthermore, even though negatively charged *i*-carrageenan structures were provided by hydrogen ions, H^+, at a pH below the pH_{pzc}, the amount of H^+ ions given was insufficient to neutralise the hydrogel surface. This is demonstrated by the fact that the pH value below pH_{pzc} is negative [18]. Hydroxide ions, OH^- from sodium hydroxide (NaOH), established hydrogen bonds with a hydroxyl group on the *i*-carrageenan structure at pH > pH_{pzc}, as shown in Fig. 2 (b). The interactions that exist cause the surface of the hydrogel to become positive. This was confirmed as the value of ΔpH above pH_{pzc} was positive.

However, the pH_{pzc} values for the ICC1, ICC3, ICC5, ICC7, and ICC9 films differed, with values of 6.80, 6.78, 6.76, 6.35, and 6.56, respectively. The development of a crosslink between the Ca^{2+} ion and the $–OSO_3^-$ group is caused by the presence of $CaCl_2$ in the *i*-carrageenan chain. The number of Ca^{2+} ions per unit volume in solution increases as the $CaCl_2$ concentration increased [20]. Despite the crosslinking, there are still a few $–OSO_3^-$ groups that do not interact with Ca^{2+} ions but do interact with H^+ ions. However, the fact that pH remained negative suggests that there was still a free $–OSO_3^-$ group in the structure of *i*-carrageenan that was not bound to H^+ ions electrostatically. Furthermore, pH above pH_{pzc} had a positive value. The –OH group on the *i*-carrageenan structure will create hydrogen bonds with OH^- ions from NaOH, according to the same explanation. The interactions that occur lead the hydrogel's surface to become positive.

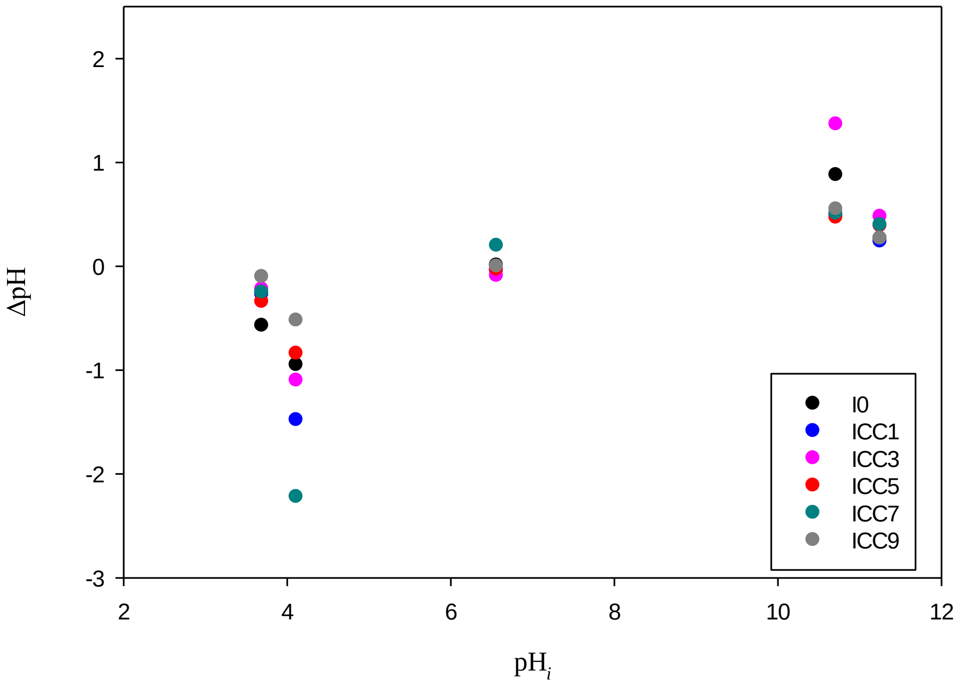


Figure 2 (a) Determination of pH_{pzc} for *i*-carrageenan/$CaCl_2$ hydrogel films

Effect of weight percentage of calcium chloride

With various $CaCl_2$ weight percentages, Fig. 3 (a) demonstrated the degree of swelling of the *i*-carrageenan hydrogel film. Because *i*-carrageenan hydrogel films disintegrate in DI water and cannot retain their form after being removed from the water, we cannot measure the degree of swelling for I0 and ICC1 hydrogel films. The swelling of hydrogel film *i*-carrageenan, on the other hand, increased from 3 wt. % to 5 wt. % to 7 wt. % to 9 wt. %. ICC3 hydrogel film has a swelling degree of 104.18 g/g, ICC5 hydrogel film has a swelling degree of 107.21 g/g, ICC7 hydrogel film has a swelling degree of 113.67 g/g, and ICC9 hydrogel film has a swelling degree of 127.89 g/g. The amount of crosslinking formed in the *i*-carrageenan hydrogel film increased as the weight percentages of $CaCl_2$ used increased [14].

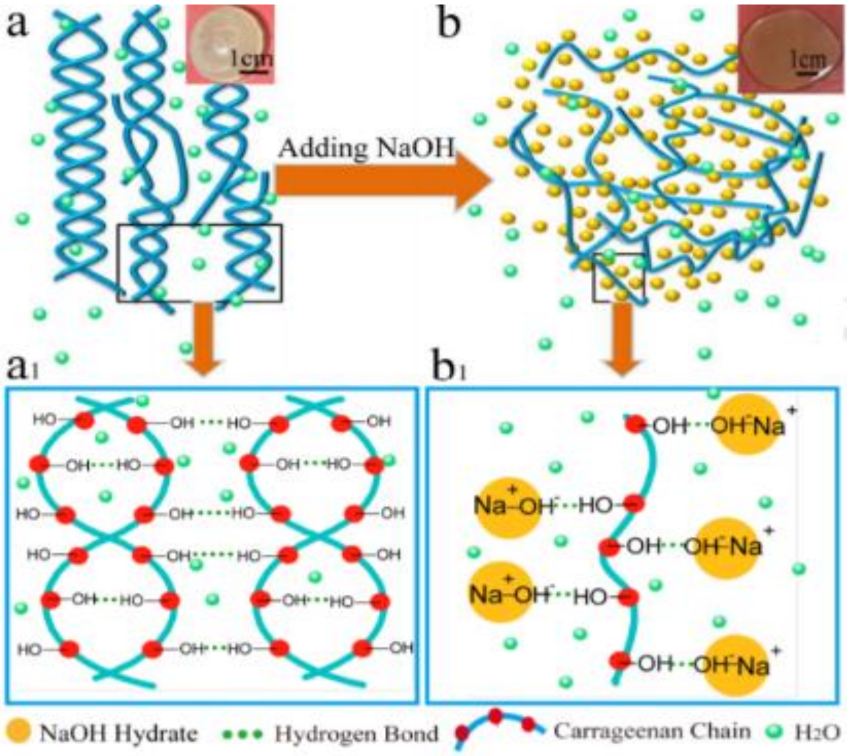


Figure 2 (b) Formation of hydrogen bonds between OH^- ions from NaOH with –OH group of *i*-carrageenan structure [18]

However, the ability of the ICC9 hydrogel film to absorb a considerable amount of water suggests that crosslinking does not occur at maximal levels in the *i*-carrageenan hydrogel film. The proposed interaction is shown in Fig. 3 (b). We predicted ionic bonding to form crosslinks between the $–OSO_3^-$ ions in the *i*-carrageenan structure and Ca^{2+} ions. In addition, an electrical attraction exists between Ca^{2+} ions and oxygen atoms on the $–OSO_3^-$ group, results in the formation of hydrogen bonds between the molecules [15-16].

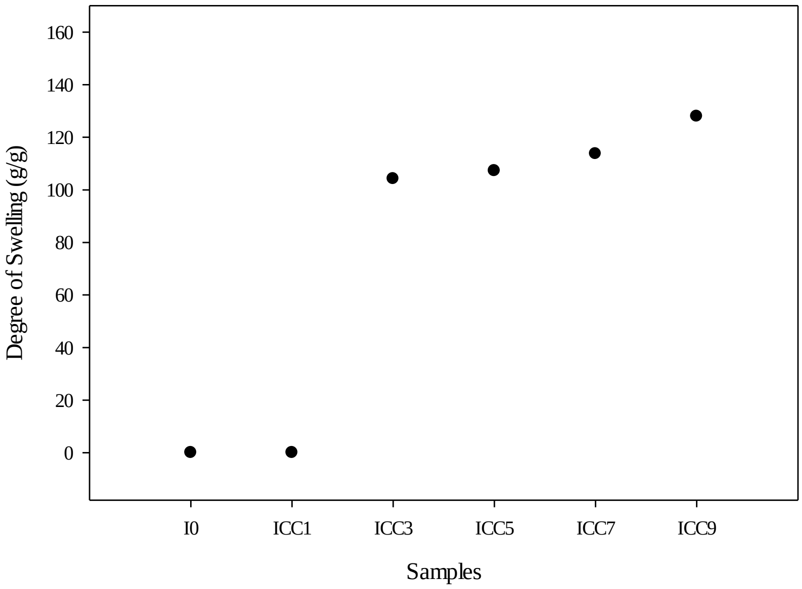


Figure 3 (a) Degree of swelling for *i*-carrageenan/$CaCl_2$ hydrogel films

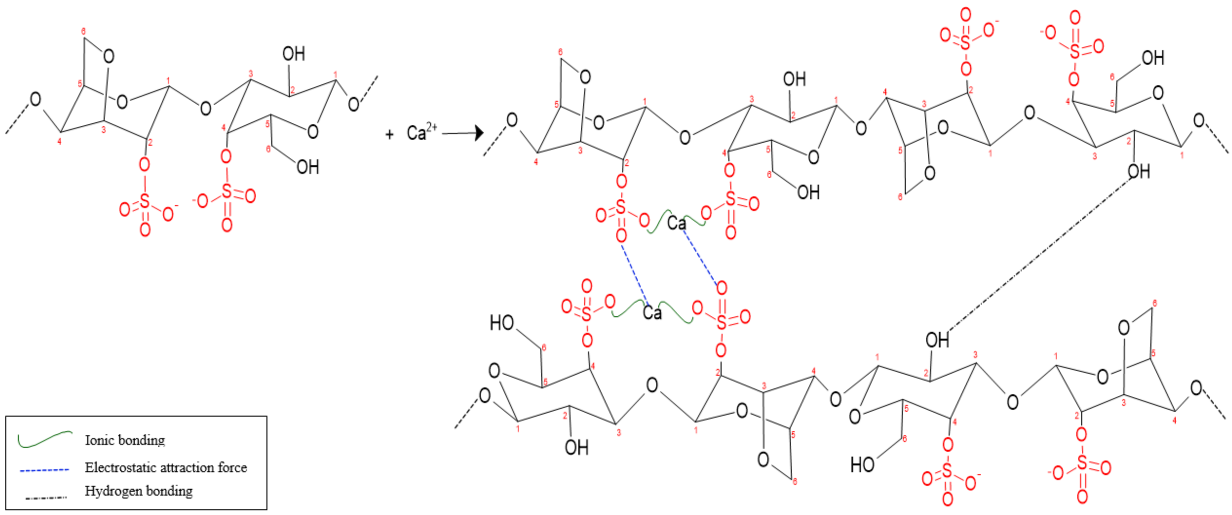


Figure 3 (b) Gelation mechanism of *i*-carrageenan with Ca^{2+} ions [15-16]

Effect of pH

The degree of swelling for ICC3, ICC5, ICC7, and ICC9 hydrogel films differs in all pH solutions, as shown in Fig. 4 (a), and a scheme for the *i*-carrageenan network at various pH levels is shown in Fig. 4 (b). Most of $–OSO_3^-$ groups in *i*-carrageenan chains that are not engaged in crosslink formation with Ca^{2+} ions will be protonated by H^+ ions under acidic environment. The H^+ ion concentration is the highest at pH 2.85 (1.41 x 10-3 mol/L). More $–OSO_3^-$ groups will be protonated by H^+ ions as the quantity of H^+ ions in solution increases, and the repulsive interaction between $–OSO_3^-$ and $–OSO_3^-$ will be weak [21]. As a result, at pH 2.85, the hydrogel films ICC3, ICC5, ICC7, and ICC9 do not swell. At pH 5.45 and pH 6.01, the H^+ ion concentrations are 3.55 x 10^{-6} mol/L and 9.77 x 10^{-7} mol/L, respectively. This implies that at pH 5.45 and 6.01, the concentration of H^+ ions are lower than at pH 2.85. The amount of $–OSO_3^-$ groups increased in accordance with the increase in pH, resulting in a greater rejection force. As a result of increase in pH from 2.85 to 6.01, the swelling degree increased.

The degree of swelling for hydrogel films varies depending on the ionic radius at the initial pH and pH alkaline [22]. The electrostatic interaction between Ca^{2+} ions and $–OSO_3^-$ groups was greater than that between Na^+ ions and $–OSO_3^-$ groups because the ionic radius of Ca^{2+} ions is smaller than that of Na^+ ions [23]. The greater the electrostatic attraction, the more stable the structure of the hydrogel film was. As a result, the degree of swelling at the initial pH was greater than at the alkaline pH. The hydrogel film cannot swell properly in alkaline environment because the screening action of Na^+ ions weaken the rejection of anionic ions. Due to the higher NaOH concentration at pH 11.83, 6.76 x 10^{-3} mol/L, compared to pH 10.64, which is 4.37 x 10^{-4} mol/L, the degree of swelling was lower at pH 11.83 than at pH 10.64. This is owing to the increased concentration of NaOH, which results in more Na^+ ion screening effects.

Effect of time

The capacity to swell decreased with time as the immersion of the hydrogel film in aqueous solution increased, as seen in Fig. 5. Table 1 showed that even at a time durability of 7 hours 55 minutes, I0 and ICC1 hydrogel films are not able to endure. The ICC3 hydrogel film can only provide a reading for the degree of swelling after 12 hours since ICC3 degrades in DI water after 17 hours. ICC5, ICC7, and ICC9 hydrogel films may persist for 17 hours, 22 hours, and 27 hours, respectively, before fully dissolving in DI water in the following hours.

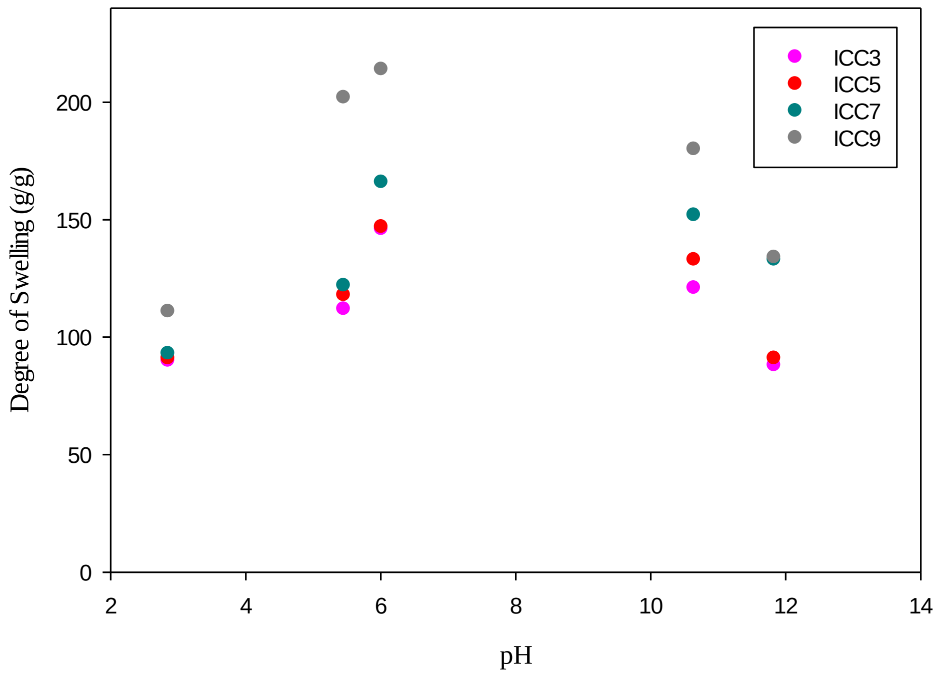


Figure 4 (a). Effect of pH on degree of swelling of *i*-carrageenan/$CaCl_2$ hydrogel films

The degree of swelling of ICC3, ICC5, ICC7, and ICC9 hydrogel films decreased with time as the structure of the hydrogel film became more compact. The development of hydrogen bonding would constrain the mobility of the gel network chains, according to Bennour and Lourzi et al. [24]. As a result, a strong hydrogel network formed, resulting in lower swelling. When the hydrogel can no longer absorb water, it becomes brittle and dissolves in aqueous solution [25].

The goal of this work was to enhance the mechanical characteristics of hydrogels by forming a crosslink between $–OSO_3^-$ groups in the *iota*-structure and Ca^{2+} ions. The mechanical characteristics of the material will improve when the cross-linker concentration is raised [26]. The ICC9 hydrogel film has the best mechanical characteristics when compared to other hydrogel

ISSN: 2550-1917

films, as it can survive in DI water for the longest duration, 27 hours, and can be totally dissolved for 32 hours. Overall, ICC9 films have a 27-hour maintenance time as compared to studies by Saidin and Mobarak [29] and Nur'Ain Nabiela [6] employing *kappa*-based films, which had 60-minute and 7-hour and 55-minute maintenance times, respectively. This suggests that the presence of $CaCl_2$ in *i*-carrageenan has improved the stability of the hydrogel film in water.

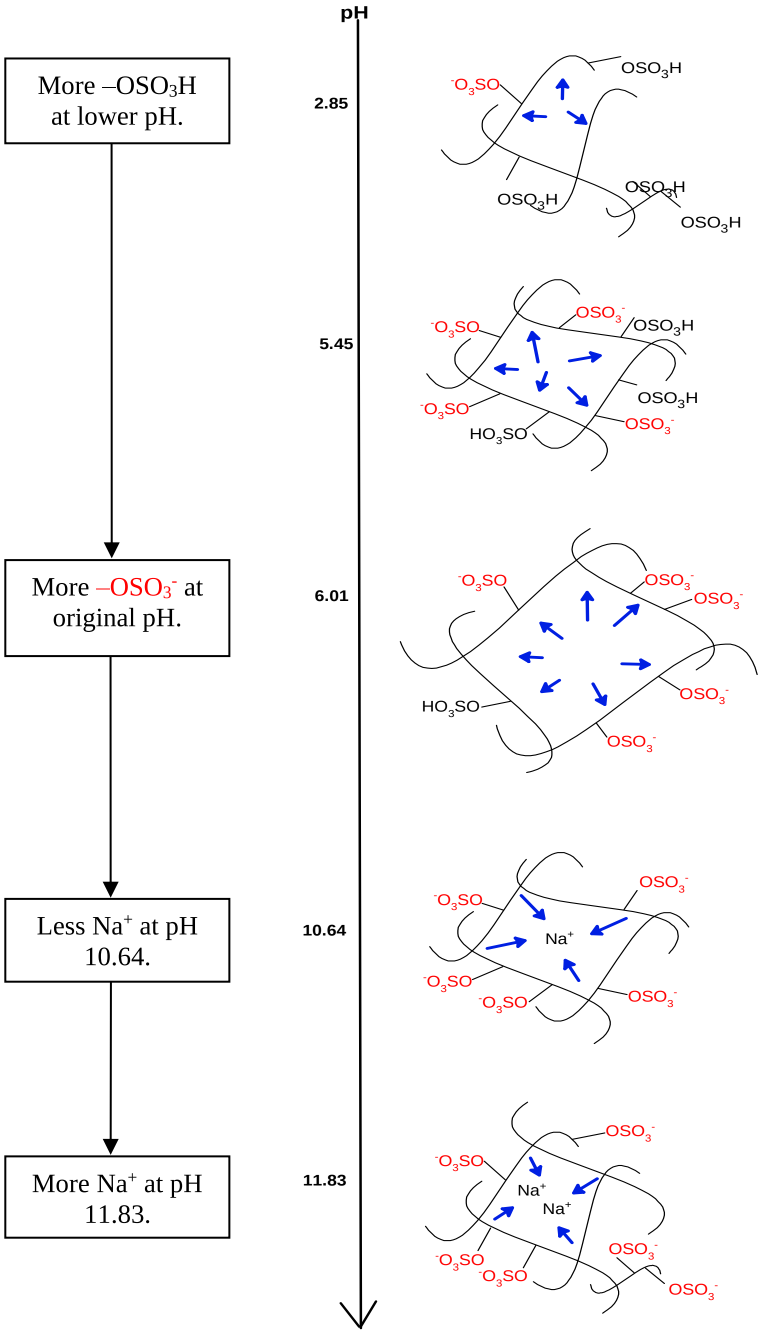


Figure 4 (b) A scheme for *i*-carrageenan network at various pH levels [30]

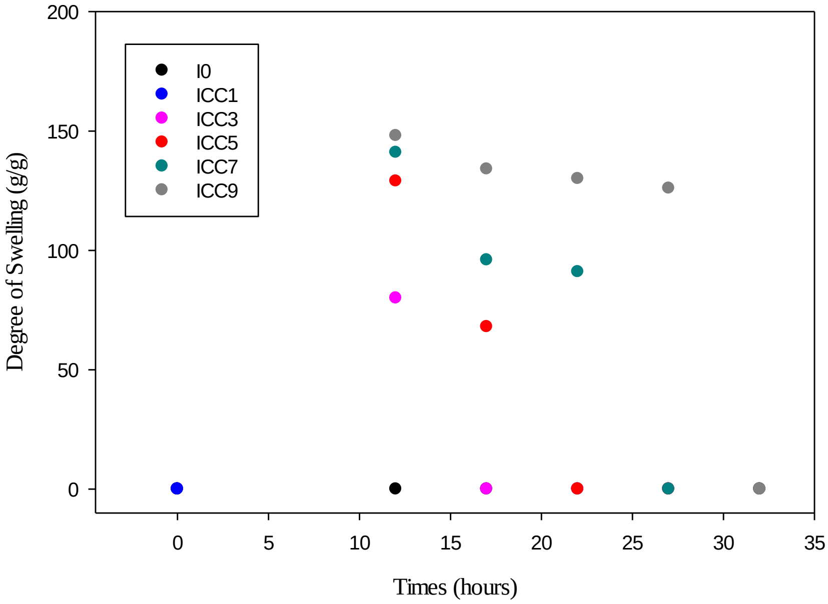
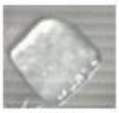


Figure 5 Effect of time on the degree of swelling of *i*-carrageenan/$CaCl_2$ hydrogel film

Table 1 Condition of hydrogel films at different time of immersion

Time	Hydrogel films condition	
	Dissolved hydrogel films	Insoluble hydrogel films
12 hours	 I0  ICC1	 ICC3  ICC5  ICC7  ICC9
17 hours	 ICC3	 ICC5  ICC7  ICC9
22 hours	 ICC5	 ICC7  ICC9
27 hours	 ICC7	 ICC9
32 hours	 ICC9	

CONCLUSION

A hydrogel based on *iota*-carrageenan with various weight percentages of calcium chloride ($CaCl_2$) has been successfully developed. It shows that the ICC9 has a high swelling capacity and high mechanical properties compared to other films when the effects of time, pH, and different weight percentage of $CaCl_2$ are applied to the film. ICC9 was able to hold out in DI water for 27 hours compared to I0, ICC3, ICC5 and ICC7. This proves that the presence of $CaCl_2$ has enhanced the ability of hydrogel based on *iota*-carrageenan as adsorbent.

ACKNOWLEDGEMENTS

We gratefully thank our colleagues from Faculty Science and Technology who provided insight and expertise that greatly assisted the research.

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