Synthesis and Characterization of Hybrid Alginate/ Cellulose as Drug Delivery System

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Abstract— The technology behind drug delivery system has expanded vastly nowadays. By searching the best ingredient in order to treat people, researcher has come up with one of the best materials. The materials here are alginate and cellulose. Alginate that come from green algae and cellulose that is derived from plant. The new things here that will be developed are hybrid or combinations of alginate and cellulose aerogel. Aerogel in the other hand is solid like material consists of 99% of air. In this study, the hybrid alginate/ cellulose aerogel was prepared using the sol-gel method. Sol means solution and gel is when the mixture of solvent which is distilled water, alginate, cellulose, calcium carbonate (CaCO₃), paracetamol, and glucono-δ-lactone (GDL) is combined and when it is complete it the mixture turned into gel and that is called sol-gel. Then, these hybrid alginate/ cellulose aerogel were further drying with air drying and supercritical drying of Carbon Dioxide (CO₂). Hybrid alginate/ cellulose aerogel was loaded with paracetamol to investigate the drug release characteristics. This hybrid alginate/ cellulose were analyzed using Infrared Spectroscopy (FTIR), Scanning Electron Microscope (SEM), and thermogravimetric analysis (TGA). Release results indicate that the hybrid aerogels loaded with paracetamol with 3wt% alginate+ 1wt% Carboxymethyl Cellulose (CMC) show better drug absorption for drug delivery because the surface area covered with paracetamol is bigger compared to blank hybrid alginate/ cellulose aerogel.

Keywords: Sodium alginate (Na-Alg), Aerogel, Sodium carboxymethyl cellulose (CMC), drug delivery system (DDS).

I. INTRODUCTION

A substance that include certain materials with rare geometry and design, not correlate with a particular mineral or material with specific formula that is called aerogels. Aerogel is an extraordinary class of nanoporous solids with complex interconnectivity and branched shape of several nanometers. Besides that, it comes in different designs, colours, and structure from monolithic to powders. In addition, aerogels have very few amount of solid component because it mostly composed of 99.8% of air which makes the substance a nearly invisible look [1]. Other than that, aerogels are made from sol-gel method, where the liquid in a gel is discharge above its critical pressure and temperature and changed with air, then producing a solid [1]. There is an increasing attraction in using biopolymers for aerogel manufacture and one of them is alginate [2].

Extracted from brown seaweed, alginate polymer composed of straight, unbranched polysaccharides with acid debris of 1,4-linked- β -D-mannuronic acid and α -L-gluronic acid leftover [3]. Besides that this natural biomaterial has been approved in various field as it is environmentally friendly, cheaper price, safe for health

and steady [4].One of them is in the field of drug delivery as is it a naturally producing biopolymer that has a lot of potential utilisation [3]. It is one of the most biopolymeric models applied in delivery systems because it is biodegradable, biocompatible, simple shape, and strength to form gels like in the existence of metal cations especially calcium, in aqueous solutions [5].

Cellulose is the most plentiful, economical and easily accessible carbohydrate polymer in the whole planet, normally extracted from plants or their debris [6]. To get the genuine product, this polymer usually branches with hemicellulose and lignin has to go through harmful chemical process with nasty alkali and acid procedure [6].

Water soluble ionic cellulose ether that produces precipitate or system which emulsions in aqueous derivate from carboxymethylation of the hydroxyl type of cellulose is a type of cellulose named Sodium carboxymethyl cellulose (CMC) [7]. Besides that, it has some advantages that can be considered if need to use it which includes greater transparency, affordable and environmentally friendly, plus this Sodium carboxymethyl cellulose (CMC) will benefit in order to lessen the possibility for toxicity when they being used as coating substances [8]. In order to improve its properties, the Sodium carboxymethyl cellulose (CMC) can be cross linked using metal ions [7].

Different types of drug delivery systems, such as nanoparticles, polymeric micelles, carbon nanotubes and nanocapsules have been widely studied to obtain optimum drug release kinetics. During the last decade, the application of biopolymer aerogel as drug delivery systems (DDS) has gained interest since these structures have large surface area and approachable pores allowing for high drug loading. Aerogels can be obtained either by introducing alginate in a solution with free cations (diffusing setting) of by liberating cations from an insoluble compounds caused by pH decrease (internal setting). On the other hand, the widely used substance in food and pharmaceutical industries called Alginate is a natural linear anionic polysaccharide found in the cell wall of brown algae. Furthermore, if used as a portion of formulation, Alginate has a good mucoadhesive feature that will make the increase in drugs absorbance. Recently, hybrid alginate-based aerogels have gained interest for multiple applications in life science. The method includes mixing a second component in sodium alginate solution and crosslinking with carbonates of metals. The decrease, increase or retention of the surface area of the hybrid system influence by the surface area contribution of the separation phase strongly. However, there is no or very little studied about hybrid of alginate/cellulose aerogels as drug delivery system nowadays. Due to interesting property changes, cellulose in the form of nanocomposite and polymer can give very good drug delivery systems applications. Therefore, by introducing the synthesis and characterization of hybrid alginate/cellulose aerogels as drug delivery system, many potential for treatment in pharmaceutical industry can be introduce.

II. METHODOLOGY

2.1 Materials

Alginic acid sodium salt from brown algae, carboxymethyl

cellulose (CMC), calcium carbonate (CaCO₃) and glucono- δ -lactone (GDL) were purchased from Sigma Aldrich, paracetamol, ethanol, and distilled water.

2.2 Preparation of alcogels

2.2.1 Hybrid Alginate/ Cellulose alcogels synthesis

Using analytical balance, measure 1% (w/v) of sodium alginate and 1% (w/v) of CMC using the weighing boat. 1% (w/v) means that ratio of weight sodium alginate to volume of distilled water need to be used in this experiment is 0.162: 15 (g: mL) and for CMC also the same which is 0.162: 15 (g: mL). 2% of calcium carbonate (CaCO₃) is added to the hybrid alginate/ CMC solution. Calcium carbonate will help Ca²⁺ divalent cations to form crosslinker of alginate Ca²⁺ [4]. 3% of paracetamol that has been grounded with mortar and pestle was added after CaCO3 has been dissolved together with hybrid alginate/ CMC solution at this stage. To permit the solution to turn into a gel, 2% of glucono-δ-lactone (GDL) was added to free the Ca²⁺ and thus decrease the pH of the solution. The complete hybrid mixture was poured into the mold, then was enclosed with parafilm to avoid contamination and was kept in the cooler or refrigerator for 18 hour at temperature 4°C, to make it turn into solid [4]. Lastly to exchange the water capacity from the hydrogel with alcohol before supercritical drying, the hybrid (alginate/ CMC) hydrogels was submerged in ethanol-water mixture range from 30% v/v, 50% v/v, 70% v/v, 90% v/v and two times using purified or 100% of ethanol.

2.3 Drying of alcogels

2.3.1 Supercritical drying of alcogels

At pressure 120±5bar and temperature 40°C the hybrid (alginate/ CMC) alcogel will be deal with supercritical drying by CO₂[4]. Equipment name supercritical fluid extractor was used in order to perform supercritical drying. Alcogel monoliths will be put in drying compartment. At a speed of 4 bar/min which the desires pressure the system will be moderately pressurized [4]. A repeated circular motion of CO₂ starts to withdraw alcohol from the hybrid (alginate/ CMC) alcogels [4]. At times of 4 hour, the drying technique will be performing eight times. Finally in order to prevent shrinkage or harm of these aerogels, after eight cycle of CO₂ rotation will be finish, at a rate of 2 bar/min the aerogel will be obtained.

2.3.2 Air drying of alcogels

The alcogel of hybrid alginate/ cellulose was put on the petri dish covered with aluminum foil and many small hole was punching in order for air drying to take place. They were left for air dry for 24 hour in the room temperature until the size of alcogel of hybrid alginate/ cellulose had been reduced.

2.4 Characterization analysis

At pressure 70 bar and temperature 200°C, gas was removed from the hybrid alginate/ cellulose aerogels, respectively, under vacuum (<1 mPa) for 20 hour [4]. Using the method of Brunauer, Emmett, Teller (BET) the specific area was calculated while the pore size distribution was calculated from the desorption isotherm technique [4]. The surface morphology of the alginate aerogels will be determined by using Scanning Electron Microscope (SEM) [4]. The samples will be scan at voltage of 2–4 kV [4]. The aerogels (exterior and interior surfaces) will be qualitatively determined by Fourier Transform Infrared Spectroscopy (FTIR) in the range from 400 to 4400 cm⁻¹ of wavelength [4].

3 RESULTS AND DISCUSSION

3.1 Characterization of aerogel

3.1.1 Air Drying

It was observed that during the preparation and drying of hybrid alginate/ cellulose, the size of aerogel was reduced. During

the solvent exchange where the hybrid alginate/ cellulose alcogel was submerged in the mixture of ethanol-water solution for 24 hour. The different concentration of ethanol was used which ranging from 30% v/v, 50% v/v, 70% v/v, 90% v/v and 100% of ethanol. This method was done in order to remove the water from the alcogel to get the desired aerogel.

Figure 1 shows the shape of blank hybrid alginate/ cellulose aerogel after air drying at room temperature for 24 hour. The shape for each hybrid alginate/ cellulose aerogel is also different because of the different weight percentage. The shape of blank 1wt % alginate+1wt % CMC showed that it has quite circular shape that does not show the sign of shrinkage. This is because the circular shape is quite perfect and uniform eventhough it had undergo air drying process.

The shape of blank 1wt % alginate+2wt % CMC on the other hand showed that the reduction size of aerogel compared to the first one. This might be caused by the higher amount of CMC used in the experiment. Shape or morphology of hybrid alginate/ cellulose aerogel for blank 1wt % alginate+3wt % CMC showed that the reduction in size also. The shrinkage also happen at this stage because of greatest amount of CMC used compared to the other two previous alginate. 2wt % alginate+1wt % CMC of hybrid alginate/ cellulose aerogel displayed in figure 1, has bigger size. This might cause by the bigger amount of alginate used. The last hybrid alginate/ cellulose aerogel which contain 3wt % alginate+1wt % CMC also illustrate bigger morphology due to higher number of alginate used. It also has uniform circular shape.



Figure 1: Air drying of blank hybrid alginate/ cellulose aerogel: a) 1wt % alginate+1wt % CMC, b) 1wt % alginate+2wt % CMC, c) 1wt % alginate+3wt % CMC, d) 2wt % alginate+1wt % CMC, e) 3wt % alginate+1wt % CMC.

Air drying of hybrid alginate/ cellulose aerogel loaded with paracetamol was display in figure 2. The first one was hybrid alginate/ cellulose aerogel loaded with paracetamol containing 1wt % alginate+1wt % CMC+3% paracetamol. The shape here is uniform and in perfect circular shape because of the same amount of alginate and CMC used. Second image shows hybrid alginate/ cellulose aerogel loaded with paracetamol containing 1wt % alginate+2wt % CMC+3% paracetamol. The size has increased because of the higher number of CMC used in this aerogel. Unfortunately for hybrid alginate/ cellulose aerogel loaded with paracetamol containing 1wt % alginate+3wt % CMC+3% paracetamol, the size of aerogel has slightly become smaller and a little shrinkage can be seen. Same as hybrid alginate/ cellulose aerogel in figure 2, the aerogel with higher alginate weight percentage showed bigger size. The hybrid alginate/ cellulose aerogel were hybrid alginate/ cellulose aerogel loaded with paracetamol containing 2wt % alginate+1wt % CMC+3% paracetamol and 3wt % alginate+1wt % CMC+3% paracetamol.



Figure 2: Air drying of hybrid alginate/ cellulose aerogel loaded with paracetamol: a) 1wt % alginate+1wt % CMC+3% paracetamol, b) 1wt % alginate+2wt % CMC+3% paracetamol, c) 1wt % alginate+3wt % CMC+3% paracetamol, d) 2wt % alginate+1wt % CMC+3% paracetamol, e) 3wt % alginate+1wt % CMC+3% paracetamol.

3.1.2 Supercritical Drying

Figure 3 showed the shape of blank hybrid alginate/ cellulose aerogel after supercritical drying using supercritical fluid extractor (SFE). The shape for each hybrid alginate/ cellulose aerogel is also different because of the different weight percentage and drying time. The shape of blank 1wt % alginate+1wt % CMC showed that it has circular shape that does not show the sign of shrinkage. This is because the circular shape is quite perfect and uniform eventhough it had undergo air drying process.

The shape of blank 1wt % alginate+2wt % CMC on the other hand showed that the reduction size of aerogel compared to the first one but still perfect circular shape. This might be caused by the higher amount of CMC used in the experiment. Shape or morphology of hybrid alginate/ cellulose aerogel for blank 1wt % alginate+3wt % CMC showed that the reduction in size also. The shrinkage also happened at this stage because of greatest amount of CMC used compared to the other two previous alginate. 2wt % alginate+1wt % CMC of hybrid alginate/ cellulose aerogel displayed in figure 3, has bigger size. This might cause by the bigger amount of alginate used. The last hybrid alginate/ cellulose aerogel which contain 3wt % alginate+1wt % CMC also illustrate bigger morphology due to higher number of alginate used. Longer drying time made the aerogel puff a little bit.



Figure 3: Supercritical drying of blank hybrid alginate/ cellulose aerogel: a) 1wt% alginate+1wt% CMC+3%, b) 1wt % alginate+2wt % CMC+3%, c) 1wt % alginate+3wt % CMC+3%, d) 2wt % alginate+1wt % CMC+3%, e) 3wt % alginate+1wt % CMC+3%.

Supercritical drying of hybrid alginate/ cellulose aerogel loaded with paracetamol was display in figure 4. The first one was hybrid alginate/ cellulose aerogel loaded with paracetamol containing 1wt % alginate+1wt % CMC+3% paracetamol. The shape here is uniform and in perfect circular shape because of the same amount of alginate and CMC used. Second image showed hybrid alginate/ cellulose aerogel loaded with paracetamol containing 1wt % alginate+2wt % CMC+3% paracetamol. The size has increased because of the higher number of CMC used in this aerogel. Unfortunately for hybrid alginate/ cellulose aerogel loaded with paracetamol containing 1wt % alginate+3wt % CMC+3% paracetamol, the size of aerogel has slightly become smaller and a little shrinkage can be seen. Same as hybrid alginate/ cellulose aerogel in figure 4, the aerogel with higher alginate weight percentage shows bigger size. The hybrid alginate/ cellulose aerogel were hybrid alginate/ cellulose aerogel loaded with paracetamol containing 2wt % alginate+1wt % CMC+3% paracetamol and 3wt % alginate+1wt % CMC+3% paracetamol. This aerogel also puff up because the drying is longer in the SFE compared to other aerogel.



Figure 4: Supercritical drying of hybrid alginate/ cellulose aerogel loaded with paracetamol: a) 1wt % alginate+1wt % CMC+3% paracetamol, b) 1wt % alginate+2wt % CMC+3% paracetamol, c) 1wt % alginate+3wt % CMC+3% paracetamol, d) 2wt % alginate+1wt % CMC+3% paracetamol, e) 3wt % alginate+1wt % CMC+3% paracetamol.

3.2 IR spectra analysis

FTIR analysis was used in this experiment to study the component present in the hybrid alginate/ CMC aerogel. As shown in figure 5, several characteristics absorption bands can be observed at 3313 cm⁻¹ (O-H stretch), 2188 cm⁻¹ (C=C stretch), 1602.75 cm⁻¹ (C=C stretch), 1403.47 cm⁻¹ (-C-H stretch), 914.51 cm⁻¹ (=C-H stretch), 1015.97 cm⁻¹ (C-O stretch), and 872.62 cm⁻¹ (=C-H stretch), for FTIR spectrum of blank hybrid alginate/ cellulose aerogel of 1wt % alginate+1wt % CMC. In FTIR spectrum of blank hybrid alginate/ cellulose aerogel of 1wt % alginate+2wt % CMC the absorption bands present were 3352 cm⁻¹ (O-H stretch), 1793 cm⁻¹ (C=O stretch), 1641.59 cm⁻¹ (C=O stretch), 1391.91 cm⁻¹ (C-F stretch), 870.98 cm⁻¹ (=C-H stretch), and 710.98 cm⁻¹ (=C-H stretch). In addition at FTIR spectrum of blank hybrid alginate/ cellulose aerogel of 1wt % alginate+3wt % CMC the absorption bands present were 3352 cm⁻¹ (O-H stretch), 1793 cm⁻¹ (C=O stretch), 1641.59 cm⁻¹ (C=O stretch), 1391.91 cm⁻¹ (C-F stretch), 870.98 cm⁻¹ (=C-H stretch), and 710.98 cm⁻¹ (=C-H stretch). At FTIR spectrum of blank hybrid alginate/ cellulose aerogel of 2wt % alginate+1wt % CMC the absorption bands present were 3318.12 cm⁻¹ (O-H stretch), 2162.93 cm⁻¹ (C=C stretch), 1630.20 cm⁻¹ (C=C stretch), 1412.66 cm⁻¹ (S=O stretch), 1029.19 cm⁻¹ (C-O stretch), and 872.69 cm⁻¹ (C=C stretch). The FTIR spectrum of blank hybrid alginate/ cellulose aerogel of 3wt % alginate+1wt % CMC the absorption bands present were 3357.43 cm⁻¹ (N-H stretch), 1602.55 cm⁻¹ (C=C stretch), 1411.72cm⁻¹ (S=O stretch), 1025.53cm⁻¹ (C-O stretch), 875.85 cm⁻¹ (C=C stretch), 710.90 cm⁻¹ (C-H stretch), and 614.02 cm⁻¹ (C-Br stretch). Some of the absorption bands appear in FTIR spectrum of blank hybrid alginate/ cellulose aerogel for pure CMC were 3256.82 cm⁻¹ (O-H stretch), 1594.29 cm⁻¹ (C=C stretch), 1393.43 cm⁻¹ (S=O stretch), 1061.27 cm⁻¹ (C-O stretch), 872.27 cm⁻¹ (C=C stretch), and 710.81 cm⁻¹ (C-H stretch). This FTIR characterization was done to determine the component present in each hybrid alginate/ cellulose aerogel in order to find the best ingredient for drug delivery system.



Figure 5: FTIR spectrum of blank hybrid alginate/ cellulose aerogel using air drying method: a) 1wt % alginate+1wt % CMC, b) 1wt % alginate+2wt % CMC, c) 1wt % alginate+3wt % CMC, d) 2wt % alginate+1wt % CMC, e) 3wt % alginate+1wt % CMC, f) pure CMC

Figure 6 illustrate several characteristics absorption bands that can be observed at 3339 cm⁻¹ (N-H stretch), 1599.27 cm⁻¹ (C=C stretch), 1398.75 cm⁻¹ (O-H stretch), 1156.62 cm⁻¹ (S=O stretch), 1023.78 cm⁻¹ (C-O stretch), 870.96 cm⁻¹ (C=C stretch), and 711.27 cm⁻¹ (C-H stretch), for FTIR spectrum hybrid alginate/ cellulose aerogel loaded with paracetamol of 1wt % alginate+1wt % CMC+3% paracetamol. In FTIR spectrum of hybrid alginate/ cellulose aerogel loaded with paracetamol of 1wt % alginate+2wt % CMC+3% paracetamol the absorption bands present were 3742.76 cm⁻¹ (O-H stretch), 3380.77 cm⁻¹ (N-H stretch), 1551.81 cm⁻¹ (C=C stretch), 1411.89 cm⁻¹ (S=O stretch), 1033.51 cm⁻¹ (S=O stretch), 756.82 cm⁻¹ (C-H stretch), and 609.27 cm⁻¹ (C-Br stretch). In addition at FTIR spectrum of hybrid alginate/ cellulose aerogel loaded with paracetamol of 1wt % alginate+3wt % CMC+3% paracetamol, the absorption bands present were 3286.77 cm⁻¹ (O-H stretch), 1795.96 cm⁻¹ (C=O stretch), 1603.21 cm⁻¹ (C=C stretch), 1398.64 cm⁻¹ (S=O stretch), 1020.30 cm⁻¹ (C-O stretch), 871.38 cm⁻¹ ¹ (=C-H stretch), and 711.59 cm⁻¹ (C-H stretch). At FTIR spectrum of hybrid alginate/ cellulose aerogel of 2wt % alginate+1wt % CMC+3% paracetamol loaded with paracetamol the absorption bands present were 3288.54 cm⁻¹ (O-H stretch), 2159.43 cm⁻¹ (C=C stretch), 1603.01 cm⁻¹ (C=C stretch), 1411.10 cm⁻¹ (S=O stretch), 1025.17 cm⁻¹ (C-O stretch), 873.02 cm⁻¹ (C=C stretch), and 710.63 cm⁻¹ (C-H stretch). The FTIR spectrum of hybrid alginate/ cellulose aerogel of 3wt % alginate+1wt % CMC+3% paracetamol loaded with paracetamol the absorption bands present were 3276.98 cm⁻¹ (O-H stretch), 1599.20 cm⁻¹ (C=C stretch), 1409.65cm⁻¹ (S=O stretch), 1025.94cm⁻¹ (C-O stretch), 947.19 cm⁻¹ (C=C stretch), and 620.26 cm⁻¹ (C-Br stretch). Some of the absorption bands appear in FTIR spectrum of hybrid alginate/ cellulose aerogel loaded with paracetamol for pure CMC were 3256.82 cm⁻¹ (O-H stretch). 1594.29 cm⁻¹ (C=C stretch), 1393.43 cm⁻¹ (S=O stretch), 1061.27 cm⁻¹ (C-O stretch), 872.27 cm⁻¹ (C=C stretch), and 710.81 cm⁻¹ (C-H stretch). Lastly the absorption bands appeared at FTIR spectrum of hybrid alginate/ cellulose aerogel loaded with paracetamol for pure paracetamol were 3157.08 cm⁻¹ (O-H stretch), 1650.58 cm⁻¹ (C=O stretch), 1505.72 cm⁻¹ (C=C stretch), 1225.25 cm⁻¹ (C-O stretch), 1015.65 cm⁻¹ (C=O stretch), and 682.37 cm⁻¹ (C-H stretch). This FTIR characterization was done to determine the component present in each hybrid alginate/ cellulose aerogel in order to find the best ingredient for drug delivery system.



Figure 6: FTIR spectrum of hybrid alginate/ cellulose aerogel loaded with paracetamol using air drying method: a) 1wt % alginate+1wt % CMC+3% paracetamol, b) 1wt % alginate+2wt % CMC+3% paracetamol, c) 1wt % alginate+3wt % CMC+3% paracetamol, d) 2wt % alginate+1wt % CMC+3% paracetamol, e) 3wt % alginate+1wt % CMC+3% paracetamol, f) pure CMC, g) pure paracetamol.

FTIR analysis was used in this experiment to study the component present in the hybrid alginate/ CMC aerogel. As shown in figure 7, several characteristics absorption bands can be observed at 731 cm⁻¹ (C=Cl stretch), 881 cm⁻¹ (C-H stretch), 1051.75 cm⁻¹ (C-O stretch), 1435 cm⁻¹ (C=C stretch), 1663 cm⁻¹ (C=C stretch) and 3000 cm⁻¹ (C-O stretch) for FTIR spectrum of blank hybrid alginate/ cellulose aerogel of 1wt % alginate+1wt % CMC. In FTIR spectrum of blank hybrid alginate/ cellulose aerogel of 1wt % alginate+2wt % CMC the absorption bands present were 715 cm⁻¹ (C=Cl stretch), 881 cm⁻¹ (C-H stretch), 1054 cm⁻¹ (C-O stretch), 1876 cm⁻¹ (C=O stretch), 2598 cm⁻¹ (O-H stretch), and 2985 cm⁻¹ (C-H stretch). In addition at FTIR spectrum of blank hybrid alginate/ cellulose aerogel of 1wt % alginate+3wt % CMC the absorption bands present were 735 cm⁻¹ (C=Cl stretch), 1793 cm⁻¹ (C=O stretch), 880 cm⁻¹ (C-H stretch), 1434 cm⁻¹ (-C-H stretch), 1628 cm⁻¹ (C=C stretch), and 3000 cm⁻¹ (C-H stretch). At FTIR spectrum of blank hybrid alginate/ cellulose aerogel of 2wt % alginate+1wt % CMC the absorption bands present were 946 cm⁻¹ (O-H stretch), 1090 cm⁻¹ (C=C stretch), 1434 cm⁻¹ (C=C stretch), 1412.66 cm⁻¹ (S=O stretch), 1029.19 cm⁻¹ (C-O stretch), and 872.69 cm⁻¹ (C=C stretch). The FTIR spectrum of blank hybrid alginate/ cellulose aerogel of 3wt % alginate+1wt % CMC the absorption bands present were 946 cm⁻¹ (C-O stretch), 1039 cm⁻¹ (C-F stretch) and 2196 cm⁻¹ (-C \equiv C- stretch). Some of the absorption bands appear in FTIR spectrum of blank hybrid alginate/ cellulose aerogel for pure CMC were 714 cm⁻¹ (C-Cl stretch), 879 cm⁻¹ (C=C stretch), 1622 cm⁻¹ (C=C stretch), 2997 cm⁻¹ (C-H stretch) and 3823 cm⁻¹ (N-H stretch). This FTIR characterization was done to determine the component present in each hybrid alginate/ cellulose aerogel in order to find the best ingredient for drug delivery system.



Figure 7: FTIR spectrum of blank hybrid alginate/ cellulose aerogel using supercritical drying method: a) 1wt % alginate+1wt % CMC, b) 1wt % alginate+2wt % CMC, c) 1wt % alginate+3wt % CMC,

d) 2wt % alginate+1wt % CMC, e) 3wt % alginate+1wt % CMC, f) pure CMC

Figure 8 illustrate several characteristics absorption bands that can be observed at 715 cm⁻¹ (C=Cl stretch), 881 cm⁻¹ (C-H stretch), 1053 cm⁻¹ (C-O stretch), 1097 cm⁻¹ (C-O stretch), 1447 cm⁻¹ (-C-H stretch), 1638 cm⁻¹ (C=C stretch), and 2991 cm⁻¹ (C-H stretch), for FTIR spectrum hybrid alginate/ cellulose aerogel loaded with paracetamol of 1wt % alginate+1wt % CMC+3% paracetamol. In FTIR spectrum of hybrid alginate/ cellulose aerogel loaded with paracetamol of 1wt % alginate+2wt % CMC+3% paracetamol the absorption bands present were 716 cm⁻¹ (C=Cl stretch), 879 cm⁻¹ (C-H stretch), 1437 cm⁻¹ (C-H stretch), 1628 cm⁻¹ (C=C stretch) and 3000 cm⁻¹ (C-H stretch. In addition at FTIR spectrum of hybrid alginate/ cellulose aerogel loaded with paracetamol of 1wt % alginate+3wt % CMC+3% paracetamol, the absorption bands present were 742 cm⁻¹ (C=Cl stretch), 881 cm⁻¹ (C-H stretch) and 1441 cm⁻¹ (-C-H stretch). At FTIR spectrum of hybrid alginate/ cellulose aerogel of 2wt % alginate+1wt % CMC+3% paracetamol loaded with paracetamol the absorption bands present were 744 cm⁻¹ (C-Cl stretch), 881 cm⁻¹ (C-H stretch), 1052 cm⁻¹ (C-O stretch), 1435 cm⁻¹ (C=C stretch) and 1634 cm⁻¹ (C=C stretch). The FTIR spectrum of hybrid alginate/ cellulose aerogel of 3wt % alginate+1wt % CMC+3% paracetamol loaded with paracetamol the absorption bands present were 770 cm⁻¹ (C-Cl stretch), 881 cm⁻ ¹ (C-H stretch), 1047 cm⁻¹ (C-O stretch), 1437 cm⁻¹ (C=C stretch) and 1650 cm⁻¹ (C=C stretch). Some of the absorption bands appear in FTIR spectrum of hybrid alginate/ cellulose aerogel loaded with paracetamol for pure CMC 714 cm⁻¹ (C-Cl stretch), 879 cm⁻¹ (C=C stretch), 1622 cm⁻¹ (C=C stretch), 2997 cm⁻¹ (C-H stretch) and 3823 cm⁻¹ (N-H stretch). Lastly the absorption bands appeared at FTIR spectrum of hybrid alginate/ cellulose aerogel loaded with paracetamol for pure paracetamol were 717 cm⁻¹ (C-Cl stretch), 840 cm⁻¹ (C-H stretch), 1000 cm⁻¹ (C-F stretch), 1261 cm⁻¹ (C-N stretch), 1381 cm⁻¹ (-C-H stretch), 1417 cm⁻¹ (C=C stretch), 1560 cm⁻¹ (C-H stretch), 1615 cm⁻¹ (C=C stretch), 1661 cm⁻¹ (C=C stretch) and 3334 cm⁻¹ (N-H stretch). This FTIR characterization was done to determine the component present in each hybrid alginate/ cellulose aerogel in order to find the best ingredient for drug delivery system.



Figure 8: FTIR spectrum of hybrid alginate/ cellulose aerogel loaded with paracetamol using supercritical drying method: a) 1wt % alginate+1wt % CMC+3% paracetamol, b) 1wt % alginate+2wt % CMC+3% paracetamol, c) 1wt % alginate+3wt % CMC+3% paracetamol, d) 2wt % alginate+1wt % CMC+3% paracetamol, e) 3wt % alginate+1wt % CMC+3% paracetamol, f) pure CMC, g) pure paracetamol.

4 CONCLUSION

The conclusion here is that based on synthesis and characterization, hybrid alginate/ cellulose aerogel load with paracetamol showed better result for drug delivery system compared to hybrid alginate/ cellulose aerogel blank that is not loaded with paracetamol.

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References

- 1. Sachithanadam, M. and S.C. Joshi, *Silica Aerogel Composites*. 2016: Springer.
- Raman, S., P. Gurikov, and I. Smirnova, *Hybrid* alginate based aerogels by carbon dioxide induced gelation: Novel technique for multiple applications. The Journal of Supercritical Fluids, 2015. 106: p. 23-33.
- 3. Brunetti, M., *Alginate polymers for drug delivery*. 2006, Citeseer.
- 4. Mustapa, A., et al., *Impregnation of medicinal plant phytochemical compounds into silica and alginate aerogels.* The Journal of Supercritical Fluids, 2016. **116**: p. 251-263.
- Hosseini, S.M., et al., Preparation and characterization of alginate and alginate-resistant starch microparticles containing nisin. Carbohydrate polymers, 2014. 103: p. 573-580.
- Esa, F., S.M. Tasirin, and N.A. Rahman, *Overview* of bacterial cellulose production and application. Agriculture and Agricultural Science Procedia, 2014. 2: p. 113-119.
- 7. Yu, M., J. Li, and L. Wang, *KOH-activated carbon* aerogels derived from sodium carboxymethyl cellulose for high-performance supercapacitors and dye adsorption. Chemical Engineering Journal, 2017. **310**: p. 300-306.
- Devi, B.M., et al., Synthesis and characterisation of chitosan/sodium alginate/carboxymethyl cellulose beads. Scholar Research Library, 2014. 6: p. 389-395.