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Development of antibacterial fibers and study on effect of guar-gum addition on properties of carboxymethylcellulose (CMC)/alginate fibers

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Abstract. The effect of carboxymethylcellulose (CMC) addition on the tensile and absorption properties of alginate fibers had been reported in the previous research. Great increase in absorbency was observed due to addition of CMC to calcium alginate fibers but was not balanced with mechanical strength. By addition of CMC dry and wet strength of composite fibers was reduced, therefore, a balance was needed between the absorbency of the fibers and mechanical strength. The effect to introduce another component i.e. Guar-gum on overall mechanical and absorbency characteristics is reported here. In comparison to CMC/alginate co-spun fibers, it was shown that inclusion of guar-gum improves the tensile strength as well as absorbency of composite fibers. CMC/guar-gum/alginate fibers were extruded through two different coagulation baths. In first step the dope solution was extruded through $CaCl_2$ coagulation bath. The dope of optimized composite fiber formulation with adjusted mechanical property and absorption was extruded, then, through 50:50 $CaCl_2$ and $ZnCl_2$ coagulation bath. The formed CMC/guar-gum/Zn/alginate composite fibers were found excellent for medical applications with their super absorbency along with equivalent mechanical strength and high antibacterial efficacy.

1. Introduction

Seaweeds have been considered a rich source of polysaccharide because of their durability, reliability, availability, and novel applications with high efficiency. Various research studies have been conducted owing to their biocompatibility, biodegradability and eco-friendliness. Alginate from brown seaweed has been used in biomaterials for last many years due to its unique properties. It forms gel by exchanging its calcium ion with sodium ions of the blood. Gel forming property depends upon the quantity of β-D-mannuronic acid and α-L-guluronic acid [3]. Its swelling, hemocompatibility and hemostatic properties make it ideal for wound care materials [1- 4]. In wound management, alginate dressings are widely accepted and most prevalent [5], because, they are highly absorbent due of gel formation [6], hemostatic due to calcium ions exchange with fluid of wound and good in fluid handling permitting better moisture and oxygen transfer. These essential attributes make alginate best choice for highly exuding wounds [3], [4].

Wound occurs due to epithelial cell destruction of skin by some external agent. Skin has inherent ability to reconstruct this tissue discontinuity at wound site described as innate wound healing property that is a complex and vital natural response [7]. The prime objective is to heal the wound in minimum possible time without infecting it that occurs when the number of pathogens at wound site increases than immune system. Usually infected wound is responsible for delayed wound healing process [8]. Mostly, skin pathogens like *Staphylococcus aureus, Escherichia coli* and *Pseudomonas aeruginosa* are responsible for such infection [9]. Absorption, tensile strength, healing, antibacterial activity and protection of wound from toxicity are main functions of wound care material [10].

The properties of alginate dressings can be enhanced by the addition of non-alginate polymers. Quin and gilding [11] made fibers by using alginate and CMC. The concentration of CMC determines some of the properties such as solubility, swelling, chemical and bacterial resistance due to their entrapment in gel. The major problem in the alginate and CMC co-spun fibers was the high absorbency but not balanced with tensile strength. This issue was resolved by addition of guar gum. Guar gum also know by name of cluster bean, exhausted from refined endosperm of cluster bean is a natural nonionic polysaccharide which is soluble in water. It has the capability to change the physical and chemical properties and to form gels [12]. So, it can be used in wound care materials for the absorption of exudate [13] and to increase the strength of composite fibers. The aim of the study was to obtain optimum values of absorbency of composite fibers without compromising the mechanical properties.

2. Material and Method

2.1. Materials

Sodium alginate (protnal LF 60/10, guluronic acid content 65-75%) supplied by FMC biopolymer,Norway. Zinc chloride $ZnCl_2$ (UNI.Chem, lab grade extra pure), Guar gum and CMC of commercial grade were purchased from local market. Calcium chloride dihydrate ($CaCl_2.2H_2O$, RdH Laboratories GmbH & Co, analytical grade), Acetone (99%, Sigma Aldrich), and Sodium Chloride (NaCl, reagent grade UNI.Chem.).

2.2. Methods

2.2.1. Fiber Production. A multifunctional wet extruder ESM 4480 designed in house by NTRC and manufactured by PACE electronics Pvt. Ltd had been used for fibers production. Solution of CMC/guar-gum/alginate was prepared by 3-4 hour stirring of different concentrations of each biopolymer in distilled water to make it uniform at room temperature as shown in table 1.

Table 1. Design of experiment for fiber formation.

Sr #	Na Alginate (%)	CMC (%)	Guar gum (%)
S1	2.5	0.5	1
S2	2.5	0.75	1
S3	2.5	1	1
S4	3	0.5	0.5
S5	3	0.75	0.5
S6	3	1	0.5
S7	4	0.5	0.75
S8	4	0.75	0.75
S9	4	1	0.75

CMC was slowly fed in and stirred in which total solid contents was not more than 5% w/v as shown in Table 1. The solution pH was maintained at 7 ±1 to avoid degradation of polymeric solution at higher pH [1]. The prepared dope was filtered, shifted into the hose and left overnight to release gases at ambient conditions. The dope was pumped through extruder with 12 holes spinneret at the constant rate of 11 rpm, draw ratio of 38 rpm, into coagulation bath with concentration of 2%. The spun filaments were washed with distilled water and dried in aqueous acetone. The whole process of fiber formation is shown in Figure 1.

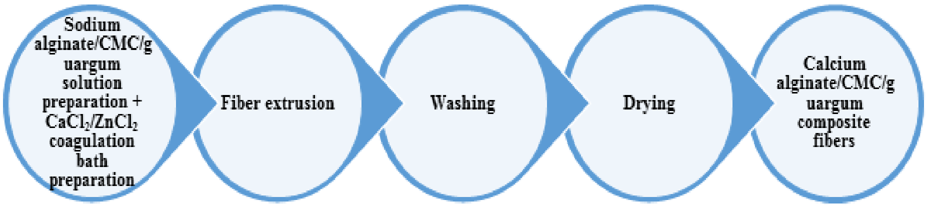

Figure 1. Flow chart for fiber development.

2.2.2. Liquid absorption testing. Liquid absorption of CMC/guar-gum/alginate composite fibers was calculated by placing fibers in three different solutions; Normal saline solution (0.9 % NaCl), distilled water and solution A (0.8298 % NaCl and 0.0368 % $CaCl_2$) by using the formula,

$$\text{Absorbance (gram/gram)} = W_w\text{-}W_d/W_d \quad (1)$$

Where, W_w is wet weight and W_d is oven dry weigh

2.2.3. Tensile strength measurement. Tensile strength, percentage elongation, and tenacity of composite fibers were measured by using single fiber strength tester according to standard EN ISO 5079/ ASTM D 5079 which was based on constant load.

2.2.4. Antibacterial testing. The fibers were investigated for their qualitative antibacterial activity by standard test procedure using AATCC 147-1998. Inhibition zone was observed for antibacterial fibers.

3. Result and discussion

3.1. SEM analysis

It can be seen from SEM micrographs (figure 2) that alginate fiber surface has more cracks without CMC/guar-gum (figure 2a). But, after their addition smoother surface was obtained (Figure 2b).

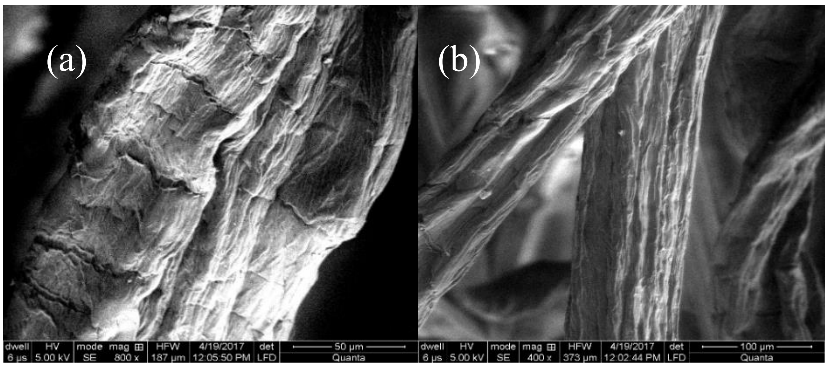

Figure 2. SEM micrograph of fibers; (a) alginate fibers; (b) CMC/guar-gum/alginate fibers

3.2. Liquid absorption testing

Basic chemical structure of all polysaccharide is nearly same and they are highly absorbent biodegradable biopolymers. Different polysaccharides are blended together to develop synergistic mixture with improved gelation characteristics. Absorption characteristics are highly dependent on Ion-exchange mechanism. Insoluble three-dimensional gel network is formed by inter-linkage of two or more molecular chains known as “Junction zones”. These “junction zones” are cross-linking of negatively charged polymeric chains with ions typically cations like (Ca^{++}, Zn^{++}). Ions (Ca^{++}, Zn^{++}) in composite fibers were changed by sodium ions in any fluid (wound exodus, Saline solution or Solution A) that come in contact with them and changing these fibers into soluble sodium alginate that absorbs high quantity of water. Thus, ion-exchange mechanism is responsible for swelling and gel formation of fibers [2]. Results are shown by graph (Figure 3). It is clearly visible that absorbency of saline solution by all samples is highest and that of water is least. While absorbency of samples for solution A exists in between these two limits. The main reason behind was the highest amount of sodium ions contained in saline solution that were exchanged by calcium ions in the fibers and made the fibers highly absorbent. Also, by the addition of CMC the performance of fibers can be improved in the same way as by the addition of sodium ions.

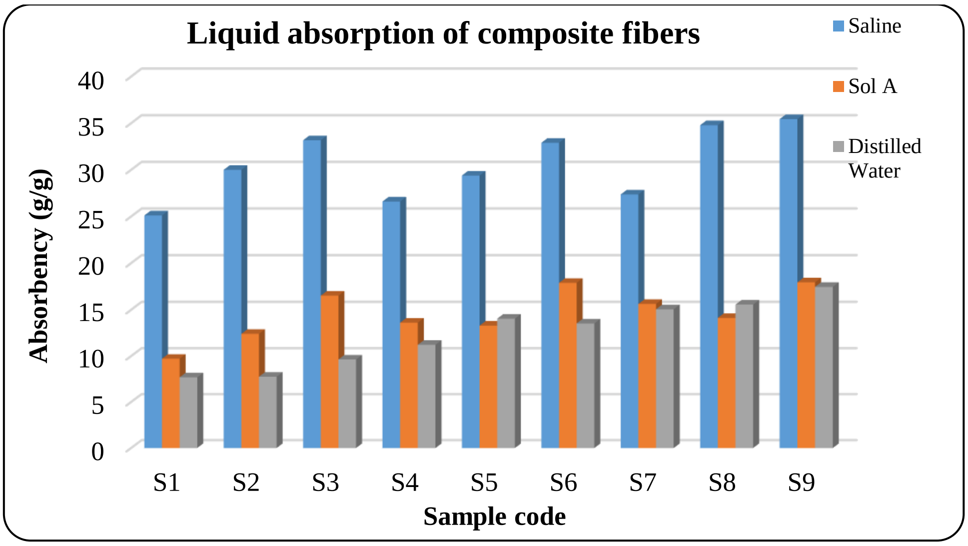

Figure 3. Liquid absorption of composite fibers for saline, solution A and distilled water.

Therefore, due to addition of this polysaccharides into composite fibers, the absorbency was increased too many folds than the simple alginate fibers. On other side, water didn't contain any ion therefore, no ion exchange occurred between fibers and water solution. But, due to addition of CMC absorbency was higher than simple alginate fibers but less than both saline and solution A. As, solution A contained both sodium and calcium ions, but less sodium ions than saline was having medium absorbency and swelling. Guar-gum showed little effect on absorbency of fibers compared to the CMC. By increasing concentration of alginate, CMC and guar-gum, the absorbency was increased. When the effect of each polysaccharide (CMC and guar-gum) for one concentration of alginate fibers was observed, increase in the concentration of CMC had more affect than guar-gum. It can be seen clearly by graph (figure 3). Highest absorbency was observed for sample 9 that contained highest amount of alginate and CMC. Due to addition of CMC regular structure of alginate is disrupted and due to more flexible structure the fibers could be easily swollen [14].

3.3. Mechanical strength

The fibers must have enough strength (6-8 cN/Tex) to be able to bear the stresses of non-woven process during formation of dressing and dressing application on wound. Therefore, balance was made between absorbency and mechanical strength of alginate fibers by addition of CMC and guar-gum.Absorbency and swelling properties can be enhanced by CMC but at the same time dry and wet strength of fibers can be negatively affected most likely due to interference of CMC within the regular polymeric chain structure of alginate fibers. Regular pattern of alginate had been broken and mechanical strength of formed fibers was reduced [14].

Average of ten testing was taken in which the effect of different concentrations of CMC, sodium alginate and guar gum on fiber strength and elongation at break was analyzed. It can be seen from (figure 4) that by increasing concentration of sodium alginate, decrease in mechanical properties was observed. As the concentration increased, the fibers became more harsh and brittle, thus, the fibers lose strength with small elongation at break. By increasing the concentration of CMC % decrease of mechanical properties was observed due to interference in regular structure of alginate. Increase in guar gum % showed increase in mechanical properties. Highest mechanical properties were found for S1, in which concentration of alginate and CMC was minimum but of guar gum was maximum. If we consider the effect of CMC due to its less concentration, alginate structure interference was less, chains were better oriented and can move smoothly [15], due to which the strength and tenacity were higher and fibres elongated more before their breakage.

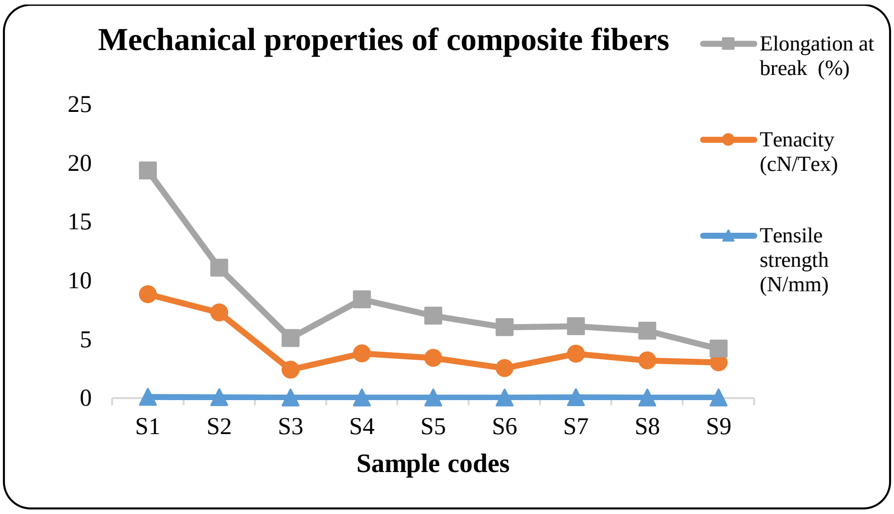

Figure 4: Graphical representation showing effect of sodium alginate, CMC and guar-gum on mechanical properties

Tenacity of CMC/alginate fibers without guar-gum was 2 cN/Tex (2.5% alginate, 0.75% CMC), but on addition of guar-gum (1%) it increased from 2cN/Tex to 7.2 cN/Tex, which is major consideration of wound dressing fibers. Absorbency of alginate fibers (2.5% alginate) was 15g/g for saline solution which on addition of CMC (0.75%) became 27.01 g/g, that further increased upto 30.007 g/g on addition of guar-gum (1%). Though guragum had minimum effect on absorbency but there was great increase in tenacity of fibers. On the basis of above mentioned results sample 2 (S2) was considered as best fibers with maximum strength and absorbency.

3.4. Antibacterial activity

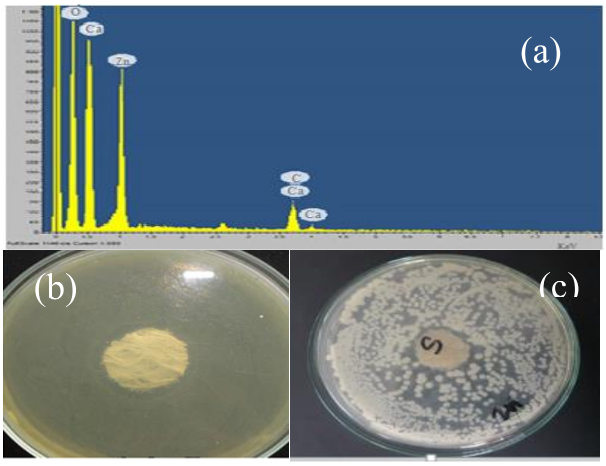

Figure 5. a) EDX analysis of fibers containing zinc ions; (b) Antibacterial property of fibers against *E.coli*; (c) Antibacterial property of fibers against *S.aureus.*

After optimization of mechanical and absorbency characteristics the same dope solution was extruded through coagulation bath containing $CaCl_2$ and $ZnCl_2$ in ratio 50:50. Energy dispersive x-ray (EDX) spectroscopy of the fibers was done for elemental analysis that confirmed the presence of zinc along with calcium ions (figure 5a). These fibers were then investigated for their antibacterial activity against two different bacterial strains. Clear inhibition zones were shown by Gram-negative bacteria (figure 4b) and Gram-positive bacteria (figure 5c). But, zone was bigger for *E.Coli* than zone made by *S.aureus* which has already established in literature [16]. These zones could be due to release of zinc ions from the fibers which were swollen by absorbing water from agar bed thus releasing antibacterial agent that possibly blocked the cell membrane permeability of bacteria causing its death [17].

4. Conclusions

Literature review has shown that increased concentration of sodium alginate increased the water uptake of fiber, but, it decreased tensile strength and elongation at break. This study showed that by increasing the CMC%, 48.2% increase in water take up of fiber occurred, but, at the same time it decreased the wet and dry tensile strength and elongation at break. Guar gum showed little increase in fiber absorbency (9.99%) but mechanical properties increased too many folds (72.2%) by its addition. These composite fibers showed excellent antibacterial efficacy against different bacterial strains and best choice for wounds.

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