

## OKRA FRUIT GUM-CHITOSAN IMPREGNATED POLYMER NETWORK FILMS: FORMULATION AND SUBSTANTIAL DEPICTION

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### ABSTRACT

**Objective:** The present research is aimed at formulation and evaluation of okra fruit gum (OFG)-chitosan (CH) impregnated polymer network films.

**Methods:** The film forming property of the gum attained from fruits of *Abelmoschus esculentus* was enhanced by co-processing it with CH. Estimation of properties including swelling index (SI), film volume, volume index, film surface contact angle with buffer solutions (pH 1.2, 7.4 or 6.8), work of adhesion (Wa), and spreading coefficient was done.

**Results:** The contact angle and SI of OFG-CH film in both acidic and alkaline buffers were witnessed to be lowest when equated with the films prepared with difference in ratios of both gum and CH. Moreover, the Wa and spreading coefficient were less for this film. These results could be attributed to the optimum interaction between  $-COO^-$  groups of gum and  $-NH_3^+$  groups of CH.

**Conclusion:** The actual nature of the film found to be tough, flexible, and water resistant. Hence, the results have shown that the films produced have a high potential for use in modified drug release and in food and pharmaceuticals.

**Keywords:** Chitosan, Contact angle, Okra, Film, Spreading coefficient, Work of adhesion.

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### INTRODUCTION

The researchers are now focusing on the exploration of the components from green materials for becoming progressively endorsed alternative for cuisine industry and medicine. Using renewable resources condensed the obligation for nonrenewable resources as implication in packaging and other single use solicitations. Natural materials possess a merit of being profusely available, biodegradable and biocompatible and have shown good forbearance due to their non-toxic nature as compared to synthetic material. Furthermore, the chemical structure of these natural products proposed that the varied functional groups employed could be reacted/cross-linked with other ions/polymers for layout of new materials for unambiguous applications.

The fruit of Bhindi (in Hindi language) plant (*Abelmoschus esculentus*; Malvaceae family) is found copiously in many areas around the world. The mucilage of Bhindi has been used for treating various diseases such as diarrhea, dysentery, and gastric ulcer as calming softener. Alqasoumi [1] and Gürbüz *et al.* [2] testified that extract of okra in methanol possessed a gastroprotective effect in rats suffering from gastric ulcer induced with ethanol. Okra mucilage, when mixed with ripe banana, possess its application in the treatment of various GIT diseases including colitis, cystitis, hepatitis, and jaundice [1]. The gum has adequate film forming property which will further anguish its utilization in constitution of foodstuff and application in delivery of various drugs. The okra fruit gum (OFG) is acidic in nature, and the structure constitutes of galactose, rhamnose, and galacturonic/glucuronic acid [3]. This announces the pertinence of free  $-COO^-$  moieties. Thus, co-processing of okra with a polymer possessing excellent film forming property could lead to augmentation of the employment of okra in food and medicine. Chitosan (CH) is reported to possess films of valuable strength owing to its properties such as being safe, polycationic, bioadaptable, and ecological in nature [4,5]. CH has been considered as useful for regulating the release of drugs from their dosage forms. Contrary to this, the property of CH being soluble

in stomach acidic pH, efforts are put on derivatization or cross-linking it for amending drug release peculiarity [6,7]. The researchers have worked to enhance the film forming property of various polysaccharides such as chondroitin [8], sodium citrate [9], sodium alginate [10], and co-processing with CH. The drug delivery in colorectal drug release dosage forms [11], sustained release drug delivery systems [12] and transdermal film formulations [7], has been improved through these films.

The interactions of  $-COO^-$  functional groups of polysaccharides with the  $-NH_3^+$  groups of ionized CH molecule could result in the alteration of film forming capability and drug release regulation property [11]. Based on the phenomena,  $-COO^-$  groups present in galacturonic acid of OFG could be interacted with  $-NH_3^+$  groups present in CH molecules. This interaction can be conjectured to improvise the utilitarian properties of putative OFG and CH.

This article emphasize on the preparation of films using a combination of OFG and CH. In addition, the evaluation of films was done for reconnoitering their potential in food and pharmaceutical industry.

### MATERIALS AND METHODS

#### Materials

The purchase of Okra fruit was done from the local market of Chandigarh. CH was purchased from Merck Pharmaceuticals and Pvt. Ltd. (Ahmedabad, India). The supplier of ammonium acetate and acetic acid was Loba Chemie, India. All chemicals used in the research were of high analytical grade and were used without modification.

#### Extraction of OFG

The method used for extraction of the gum was amended as given by Nagpal *et al.* [13]. In brief, a slurry of cleaned, sliced fruits were prepared by mixing in 3% v/v glacial acetic acid solution, and gum was extracted in 1000 ml beaker by addition of appropriate amount of distilled water

for 2 hrs at 65°C. Afterward, the filtration of slurry was performed using a muslin cloth to remove debris. The precipitation of gum was made by the inclusion of abundant acetone. The master of precipitates was removed in a vacuum oven at 60°C, and OFG sample was purified by dialysis. Further, lyophilization was performed to obtain a greenish powder. OFG powder obtained was put in the balance for weighing and yield was calculated.

#### Preparation of CH-OFG conjoin

A CH-OFG (1:1) conjoin was prepared by the method mentioned below. In brief, CH solution (prepared by adding 3% v/v acetic acid) was mixed with the solution of OFG in water dropwise, using homogenizer, to obtain gelatin like precipitates. The gelatinous precipitates obtained were dried in oven at 60°C for 48 hrs and then microwaved (80 W, 10 seconds, 15 cycles) to remove any bound water.

#### Preparation of solution of CH-OFG for production of films

In this, an attempt was made to prepare films of CH-OFG in varied ratios, i.e., 10:90, 20:80, 30:70, 40:60, 50:50, 60:40, 70:30, 80:20, and 90:10. The total weight of the polymer (ammonium acetate) was kept constant at 2% w/v. 3% v/v acetic acid was used for solubilizing CH. OFG is soluble in water. Thus, in another beaker, OFG was dissolved discretely in lukewarm water. 4M ammonium acetate was solubilized in water. The solution of ammonium acetate was then divided into two parts. One part was added to the CH solution (A) while another part was added to the OFG solution (B). This was followed by dropwise addition of solution A to solution B accompanied by uninterrupted stirring leading to the formation of a transparent solution of CH-OFG.

#### Preparation of film containing CH-OFG

The films of CH-OFG were prepared using spray gun (Electrolab, PP201V, Mumbai, India) of 1 mm nozzle. The transparent solution prepared above was sprinkled on a smooth emollient surface of rotating drum. Warm air at 55°C is puffed to remove the solvent present in the film. The restrained water present was dislodged using the microwave at 80 W, 20 seconds, 4 cycles. The prepared films were stocked for future usage.

#### Substantial depiction of CH-OFG films

##### Depiction of contact angle of varied buffer solutions (pH 1.2, 7.4 or 6.8) with film surface

The procedure reported by Jindal *et al.* [4] was altered for attaining the angle of contact of different buffers with the surface of films engaging different ratios of CH:OFG. Picture was taken using a digital camera (CANON, 20 Megapixel, 20× zoom, in Mega mode) was used for this purpose. A micropipette (200 ml) was used to put a bead of each buffer solution softly over the surface of the film. The photographs grabbed were exaggerated and were produced on A4 paper. Manual calculation was done for the calculating the contact angle of the drop. The mean of 12 readings is reported in results.

##### Volume of films

For the computation of the volume of films ( $V_f$ ), films were allowed to swell. The weight of the swollen film in air ( $W_{air}$ ) and in n-heptane ( $W_{heptane}$ ) was determined, heptane being a nonsolvent for the CH-OFG films.  $V_f$  was figured with the help of buoyancy principle:

$$V_f = \frac{W_{air} - W_{heptane}}{\rho_{heptane}}$$

Where,  $\rho_{heptane}$  is the density (0.684 g/cm<sup>3</sup>) of n-heptane.

##### Volume index ( $V_i$ )

For attaining the volume index of the films ( $V_i$ ), film volume was assured in air ( $V_{air}$ ) and in n-heptane ( $V_{heptane}$ ) and calculated using the buoyancy principle:

$$V_i = \frac{V_{air} - V_{heptane}}{V_{heptane}}$$

##### Work of adhesion ( $W_a$ )

The contact angle was employed to calculate the  $W_a$  using the equation:

$$W_a = \gamma L (\cos\theta + 1)$$

Where,  $\gamma L$  is the surface tension of the respective buffer calculated using stalagmometer method.

##### Spreading coefficient

Spreading coefficient at the buffer film surface was calculated using the equation:

$$S = \gamma L (\cos\theta - 1)$$

Where,  $\gamma L$  is the surface tension of the respective buffer calculated using stalagmometer method.

## RESULTS

One of the major excuses for a restricted application of presumed forms of OFG and CH in the industry is due to their moderate film forming ability. CH is a natural polymer but has low or no solubility in aqueous solution but is conveniently soluble in dilute acids and possess favorable film forming properties [8].  $-NH_3^+$  groups are existent in CH molecule in an ionized form while,  $-COO^-$  groups are present in OFG. Thus, it is envisioned that the interaction of these groups can succumb a material with enhanced physicochemical, performance.

#### Preparation of CH-OFG films

The preparation of CH-OFG conjugate by addition of CH dissolved in a solution of acetic acid solution to solution of OFG BFG solution resulted in the formation of white gelatinous precipitates which do not have capability of forming films. This might be on account of instinctive reaction of CH with OFG. Acetic acid dissolved CH abide  $-NH_3^+$  group. OFG accommodate galacturonic acid that contains  $-COO^-$  group. Thus, it can be justified that the formation of insoluble white gelatinous precipitates is due to the reaction between  $-NH_3^+$  group of CH and  $-COO^-$  group of OFG. Thus, there was need of addition of ammonium acetate to control this ionic interaction between CH acetate and solution of OFG. The mixture of CH acetate solution and OFG solution, resulting in the formation of the clear solution, was sprayed over surface of glass and dried at 50°C. During the process of drying, there was slow evaporation of ammonium acetate and excess acetic acid resulting in the formation of unwrinkled, ductile and brown colored film. Films could not be formed using OFG alone. Furthermore, the films formed using CH alone were very tough and breakable. That is why batches containing 10:90, 20:80, 80:20, and 90:10 were rejected. Supplementary, satisfactory films were procured using CH: OFG in the ratio of 30:70, 40:60, 50:50, 60:40, and 70:30, which were further evaluated for physico-chemical peculiarity.

#### Substantial performance of CH-OFG films

##### Swelling index (SI)

SI announces the magnitude of water subdued innards the film [14,15]. Table 1 and Fig. 1 indicates SI of different batches of films in varied buffer pH1.2, 7.4, and 6.8. It was found that OC3 film has the lowest SI, which further differs ( $p < 0.05$ ) for films in all buffers except for F4 film in buffer of pH 1.2. This indicates the water resistant nature of the film.

##### Film volume ( $V_f$ )

A variation in diametric and/or thickness of films on coming in contact with buffer pH 1.2, 7.4 or 6.8 is indicated by  $V_f$ . As shown in Table 1 and Fig. 1, OC3 film has lowest  $V_f$  value as compared to other films. This pinpoint the minimum changes in the measurement of the film, which could be result of formation of high crosslinking between OFG: CH.

##### $V_i$

$V_i$  signifies corresponding transformation in specification of the volume of film with reference to the dried film. It was observed that value of

Table 1: Physical properties of different films containing CH-OFG

S. No.	Film code	Composition		Buffer	SI	V <sub>f</sub>	V <sub>i</sub>	θ	cosθ	Wa	S.C
		CH	OFG								
1	OC1	30	70	pH 1.2	5.6±0.12	66.60±1.23	16.41±1.12	40.7±1.12	-0.99	0.69±0.04	-139.54
				pH 7.4	4.9±0.15	75.82±2.12	20.32±0.09	59.8±1.31	-0.99	0.42±0.03	-141.97
				pH 6.8	4.4±0.16	25.10±1.45	8.25±1.14	63.3±2.15	0.89	136.63±0.03	-7.77
2	OC2	40	60	pH 1.2	4.5±0.05	31.24±2.12	9.19±0.97	44.3±1.25	0.95	136.73±0.04	-3.51
				pH 7.4	3.9±0.07	88.92±1.45	23.51±2.12	67.1±2.12	-0.43	40.60±0.05	-101.80
				pH 6.8	4.2±0.06	16.72±2.41	6.12±0.74	74.3±2.31	0.46	105.06±0.07	-39.34
3	OC3	50	50	pH 1.2	4.2±0.06	16.71±2.45	5.12±0.76	79.3±2.45	-0.72	19.31±0.17	-120.94
				pH 7.4	2.5±0.08	8.97±1.74	3.57±0.59	87.6±1.92	0.93	137.71±1.14	-4.6B
				pH 6.8	3.2±0.14	11.25±1.12	4.09±0.71	65.7±1.11	-0.64	26.03±0.98	-118.37
4	OC4	60	40	pH 1.2	4.8±0.08	40.99±1.23	11.26±0.74	49.2±2.23	0.48	105.66±1.25	-36.18
				pH 7.4	3.7±0.09	8.97±1.47	6.12±0.84	82.4±2.25	0.75	124.79±1.92	-17.61
				pH 6.8	4.7±0.13	11.25±1.71	11.12±0.71	82.9±2.12	0.34	97.11±1.17	-47.29
5	OC5	70	30	pH 1.2	5.4±0.07	16.29±1.41	16.31±1.12	44.2±2.15	0.98	138.74±1.58	-1.66
				pH 7.4	4.7±0.14	12.20±1.71	14.18±1.09	63.5±2.91	0.78	127.08±1.47	-15.31
				pH 6.8	4.9±0.11	13.31±0.98	13.28±0.91	79.8±1.85	-0.31	50.13±1.74	-94.27

OFG: Okra fruit gum, CH: Chitosan, SI: Swelling index, V<sub>f</sub>: Film volume, V<sub>i</sub>: Volume index, Wa: Work of adhesion

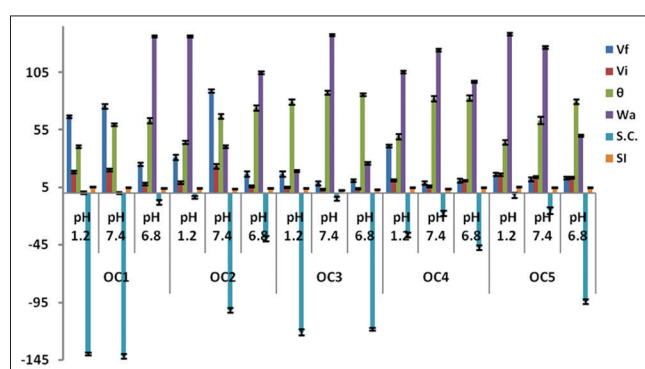


Fig. 1: Physical properties of different films containing chitosan-okra fruit gum

V<sub>i</sub> of OC3 film was found to be lowest and significantly different from volume index of other films in all other buffers. The lowest values of SI, V<sub>f</sub>, and V<sub>i</sub> of OC3 formulation proved maximum interaction between 50:50 proportion of CH and OFG that might have been a result of the appropriate interpenetrating network with a good amount of cross linking density (Table 1 and Fig. 1).

#### Film surface contact angle

The contact angle with film surface at different buffers (pH 1.2, 7.4 or 6.8) is envisioned to indicate the relative hydrophilic/lipophilic nature of the film. Moreover, an increase in contact angle increases the lipophilicity of the film surface. The contact angle of the OC3 formulation was found to be maximum in all the buffers as compared to all other formulation indicating lipophilic character of the film (Table 1 and Fig. 1).

#### Wa

Wa signifies bonding between the buffer and film surface. The results of Wa are shown in Table 1 and Fig. 1.

#### Spreading coefficient

All the film surfaces have negative spreading coefficient (Table 1 and Fig. 1).

#### DISCUSSION

From the results mentioned above, OC3 film formulation was found to be better than other films. It could be due to the most favorable interaction between -COO<sup>-</sup> groups of OFG and -NH<sub>3</sub><sup>+</sup> groups of CH during film formation resulting in slightest possibility of functional

groups of CH and OFG for hydrogen bonding with water. The contact angle of OC3 film was found to be maximum in buffer pH 7.4, followed by pH 6.8 and minimum in pH 1.2. It might be explained as from formulation OC1 to OC5, the percentage of CH increased where as that of OFG decreased. It can be considered in OC1 and OC2 films, the percentage of OFG would have been in excess of CH whereas, in OC4 and in OC5 formulations, the content of CH would have been in excess of OFG. OFG would be in unionized form in acidic pH, while CH would be unionized in alkaline pH. The data presented in Table 1 indicate that the contact angle of all other buffers increased for OC1 to OC3 films and then decreased for OC4 and OC5 films. The pattern recognized for contact angle for all the buffer solutions is defensible on the footing of the content of CH and BFG present in various films in ionized form. Wa signifies the effort needed to isolate the buffer from the exterior of film. The surface of OC3 film was more troublesome to wet when correlated to the surface of other films illustrated lowest work adhesion irrespective of the pH of the buffer. In addition, the Wa values of OC3 film was found to be significantly ( $p < 0.05$ ) different with respect to those of other films in all the buffers (Table 1). Negative value of spreading coefficient results in the formation of globule rather than film of liquid over a film surface [16]. Thus, the spreading coefficient signified comparatively less water-loving character of all the film surfaces. However, the spreading coefficient for OC3 film was found to be minimum for all the buffer. Hence, it could be proposed that the OC3 film formed by the maximal interaction of CH and OFG was least wetttable as compared to other film surfaces that probably contained few unreacted functional groups of CH or OFG. Those unreacted functional groups of CH and OFG could have been available for interaction with water molecules, thus yielding relatively hydrophilic surface as compared to the OC3 film.

#### CONCLUSION

It is, thus, concluded that -COO<sup>-</sup> moieties present in OFG and -NH<sub>3</sub><sup>+</sup> moieties present in CH interacted to form CH-OFG film. The values of SI and contact angle measurements suggested that OC5 film containing an equal proportion of CH and OFG in the ratio of 50:50 showed more lipophilic character in comparison to other films. This might due to non-availability of free functional groups in OC3 formulation to interact with water. The same justification manifested in lowest work of adhesion and spreading coefficient of OC3 formulation.

Thoroughly, in the above research, it was indicated that OC3 films containing 50:50 CH and OFG proved to play an auspicious role for modification of drug release, water permeability, protection of moisture sensitive edible items, etc. Furthermore, both CH and OFG are admitted to be safe for oral consumption. Hence, these films could be oppressed for use in food items.

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