



## Mechanical Properties of Carboxymethyl Cellulose Edible Films

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**Abstract:** This study was conducted to extract cellulose and synthesis carboxymethyl cellulose from flour bran. Fourier Transform Infrared Spectrometer (FT-IR) was used to confirm the existent of the carboxymethyl group. The sample edible films were prepared using 1, 2, 3, and 4 % CMC, and two types of plasticizers glycerol and sorbitol, (20, 40 and 60) %. Their qualitative, mechanical, reservation and thermal characteristics were studied. Tensile strength ranged 28-51.3 MPa and elongation percentage ranged between 65.5-91.0 %. The thickness of simple cellulose films were 0.018-0.078 mm. The values of solubility (19.05-36.31%) and the permeability values of simple cellulose film increased with the increasing of the plasticized ratio. The highest permeability was 11.99 g.mm/m<sup>2</sup>.h.kp at 60% glycerol and thermogravimetric analysis for some simple cellulose film plasticized by glycerol were 135, 146.29, 125 and 123.23° C.

**Keywords:** Synthesis, Carboxylmethyl cellulose, Edible films, Extraction.

### Introduction

Edible films can be defined as a thin layer of edible material encapsulated by food products acting as a barrier between product and external components (Bhattacharya, 2013). The recent use of bio-polymer as edible films has led to increased interest in food packaging when normal plastics have been replaced with biomaterials (Falguera *et al.*, 2011). Approximately 70% of all packaging materials made from plastic materials and many reports confirmed that these materials had a negative impact on human health due to interact with packaged foods, resulting in many health problems (Comstock *et al.*, 2004).

Using of polymers with high molecular weight such as protein and polysaccharides

showed a cohesive and flexible form for food packaging (Bergo & Sobral, 2007). Edible films are more suitable for food packaging because there were non-toxic effects, non-polluting and safe for human consumption (Rhim *et al.*, 2013). Wheat bran has a high percentage of cellulosic materials, and wheat bran is often disposed of by mills, causing environmental pollution (Xie *et al.*, 2008). The most abundant organic polymer in the world was Beta-glucose (Richardson & Gorton, 2003). Four forms of cellulose are used to form edible films: Hydroxypropyl cellulose (HPC), hydroxypropyl methylcellulose (HPMC), carboxymethyl cellulose (CMC) and methylcellulose (MC) (Murray, 2000). For water-related or hydrophobic and thermal-gel formation, leading to the formation of excellent edible

films used in the food industry (Turhan & Sahbaz, 2004). The aim of the present study is to investigate the mechanical and reservation properties for the edible film prepared from carboxymethyl cellulose synthesized from bran cellulose which is by product of wheat flour processing.

## Materials and Methods

### Materials

#### Flour Bran

The wheat flour bran was obtained from the General Grain Processing Company was obtained from local markets. All samples were cleaned and stored in polyethylene bags at a temperature of 4° C .

#### Extracts of cellulose from wheat flour bran

The method described by Liu *et al.* (2005) was followed to extracted cellulose from wheat bran.

#### Synthesis of carboxymethyl cellulose

For the synthesis of Carboxymethyl cellulose, the procedure of Rachtanapun *et al.* (2007) was followed.

#### Fourier Transform Infrared Spectrometer (FT-IR)

The FT-IR device was used at the Polymer Research Centre, Basrah University. The dried and powdered sample was mixed with potassium bromide and pressed as a tablet between the two disks of the device. The wavelength range was 400-4000 cm<sup>-1</sup> (Melo *et al.*, 2011).

#### Film formation

The cellulose films were used according to the method described by Asl *et al.* (2017), using carboxymethyl methyl cellulose (CMC), towns (glycerol, sorbitol) and distilled water in the preparation of films. The CMC was used in percentages (1, 2, 3, 4) w/w for water and plastic (20, 40 & 60) based on dry weight, prepare 24 different diameters were observed, and the components and CMC dissolved in distilled water at 80° C. The mix was mixed by a magnetic stirrer until a homogeneous mixture was added. After that, the plasticizer was added to the mixture and after obtaining a homogeneous solution Pour

the film to such a degree in plastic dishes with a diameter of 9 cm (size 10 ml) the films were left to dry at a temperature of 55° C (dried oven) for 24 hours. After dehydration, the films were removed from the dishes and placed in bags of polyethylene and kept in the refrigerator.

#### Films Condition

The simple and edible cellulose films were modified before they were tested (Shojaee-Aliabadi *et al.*, 2013) by placing the films in a desiccator. The chamber contained a saturated magnesium nitrate solution Mg<sub>2</sub> (NO<sub>3</sub>) for 48 hours and a temperature 25 ± 1° C to provide a relative humidity of 53 ± 1% before starting subsequent tests, and the thermo-graph was used to measure humidity and temperature within the glass dryer. Mechanical, thermal and mechanical tests for simple cellulose films Measure tensile strength and elongation until the cutting.

#### Mechanical & permeability tests of simple cellulose films

#### Tensile strength & Percentage elongation at break

The tensile strength and percentage of elongation were measured up to the cut using the German Texture analyzer supplied by 2.5 Zwick Roell which is located in the Polymer Research Centre, University of Basrah according to ASTM 91-D-882 (Ferreira *et al.*, 2009). After adjusting the films to the humidity of 53 ± 1% for 48 hours, cut in length 60 mm ribbons and 20 mm width and draw several layers of adhesive tape on both ends of the film which was installed well between the terminals of the machine. The sampling speed was 50 mm /min, indicating the results in the form of graphic markers drawn on the computer connected to the machine.

#### Films Thickness determination

The method which used by Zhong & Xia (2008) was used to measure film thickness using a digital micrometre at the Polymer Research Center / Basrah University.

#### Determination of film viability

The method proposed by Ojagh *et al.* (2010) was used to estimate the viability of films in water dissolving with some modifications:-

1. Cut the films into pieces of 2 x 2 cm.
2. Taking the primary weight to cut the film using a sensitive balance.
3. The films were submerged in 50 ml of distilled water in a glass bowl. Sodium azide was added at a concentration of 0.02% (weight/volume). The mugs were covered with aluminium foil and kept in a 25° C incubator for 24 hours.
4. Remove the film fragments from the mugs and wash with distilled water and blow in the oven at a temperature of 100 - 105° C and then rebalance to obtain the final weight of the sample (which represents the weight of the dry material not dissolved in water).

$$\frac{\text{Water solubility \%}}{\frac{\text{Primary weight of sample (gm)} - \text{Final sample weight (g)}}{\text{Primary weight of sample}}} = \text{ } \times 100$$

### Water Vapor Permeability

The method of Zhong & Xia (2008) was used to estimate the permeability of films for water vapor with some modifications. The film sample was well established using rubber on a cylindrical 30 mm diameter bowl with a height of 50 mm containing 10 g distilled water, taking the weight of the container with its contents. The vessel in Desiccator was set to 0% relative humidity using anhydrous copper sulphate at 25 ± 2° C. The vessel weight was taken every 6 hours for two days, the vacuum was measured again and the permeability was calculated by method (Dashipour *et al.*, 2014).

**First:** The relationship between weight loss (g) vs. hour was determined to estimate the slope (Slop) of the linear area and the correlation coefficient was 0.99. To estimate the rate of water vapor transfer (WVTR) equation (1)

$$WVTR = \frac{Slop}{Film} \frac{gm}{m^2.h} \dots\dots\dots(1)$$

Film area= Mug area of the inner diameter (m<sup>2</sup>)

**Second:** Application of equation (2) to estimate the partial pressure of water vapor below the film (P<sup>2</sup>)

$$\frac{WVTR}{MW} = \frac{P D L \ln (F-P_2)(F-P_1)}{R T \Delta Z} \dots\dots\dots(2)$$

Where:

WVTR= Water vapor transmission rate (gm/m<sup>2</sup>. h).

MW = Water molecular weight (18 gm/mol).

P = total pressure (1 atmosphere).

D = Diffusion of water vapor by air at absolute temperature (298K) = (1.02x10<sup>-1</sup>m/ s).

Ln = 1/ logarithm.

P<sub>1</sub> = pressure for saturated water vapor at absolute temperature (298K) = (3.13x10<sup>-2</sup> m. atmosphere).

P<sub>2</sub> = partial pressure of the water vapor below the film (atmosphere).

R = gas constant (88.1 x10<sup>-6</sup>) m<sup>3</sup> atmosphere / gm. mol is an absolute temperature.

T = absolute temperature (298K).

Δ Z = the height of the antenna vacuum which is calculated as follows:

$$\Delta Z = \frac{\text{Height at test start (m)} + \text{Height at end of examination (m)}}{2}$$

**Third:** To compensate the value (P<sub>2</sub>) calculated from equation (2) and the value of WVTR from equation (1) in equation (3), the water vapor permeability of the film (WVP) was estimated.

$$WVP = \frac{WVTR}{P_2 - P_3} \times L \dots\dots\dots(3)$$

P<sub>2</sub> = partial pressure of the water vapor below the film (atmosphere).

P<sub>3</sub>= Partial pressure of the water vapor above the film (atmosphere) in the dryer at 0% relative humidity and 25 ° C extracted from special tables.

L = film thickness rate (mm). (Yahya, 2000).

### Statistical analysis

Standard deviation (CRD) was estimated for the results of experiments in the study according to Al-Rawi and Khalafallah (2000).

## Results & Discussion

### Extraction & phenotypic diagnosis of cellulose & carboxymethyl cellulose

The cellulose was extracted from local wheat flour bran, which was described as a white powder, odourless, insoluble in water and non-viscous. Through phenotypic diagnosis, it was observed to be identical to that in Hu *et al.* (2011), Carboxymethyl cellulose was synthesized from bran cellulose, It is characterized as a white powder, colourless, odourless, odourless and easy to dissolve in water and is a basic transparent viscous substance or solution. Comparing with other studies, it is identical to results of Benchabane & Bekkour (2008).

### Fourier Transform Infrared Spectroscopy Test

FT-IR spectroscopy is widely used in cellulose research because it provides a simple way to obtain first-hand information about chemical changes occurring during different chemical processes (Din *et al.*, 2002). The infrared spectrum of the cellulose and CMC samples was taken to identify the completion of the CMC synthesis process as shown in Fig. (4). Hydroxyl free OH, as well as hydrogen bonds between and within the cellulose molecules. These results were consistent with the findings of (Pushpamalar *et al.*, 2006). The forms also showed a low density (2897.52 & 2917.77)  $\text{cm}^{-1}$  for both cellulose and carboxymethyl cellulose, respectively. This case is due to the vibration of the matrix C-H. These results are consistent with Biswal & Singh (2004). From the figure, the spectral analysis of the

carboxymethyl cellulose showed a relatively sharp and large package appearance at 1618.95  $\text{cm}^{-1}$  and the appearance of a weak packet at the range 1436.71  $\text{cm}^{-1}$ , indicating the carboxymethyl groups C = O. These results were consistent with Wang *et al.* (2007). Carboxymethyl groups and their salts showed two packets of frequency (1600 - 1640)  $\text{cm}^{-1}$  and (1400- 1450)  $\text{cm}^{-1}$  which indicate the appearance of aggregates of carboxymethyl substituted substitutions. These results are consistent with the results of Hong (2013). Figs. (1 & 2) showed that there were weak beams of 1360.53 and 1330.64  $\text{cm}^{-1}$  for both cellulose and carboxymethyl cellulose respectively, due to the O-H and the presence of strong bundles (1058.73 and 1053.91)  $\text{cm}^{-1}$ , which is due to vibration of the pyranose C-O-C rings. These results are consistent with Vieira *et al.* (2007). The shapes also showed the presence of large beams at the range (614.217 and 613.252)  $\text{cm}^{-1}$ , indicating the vibration of the C-H aggregates that matched what was found by Hong (2013).

### Tensile strength

The results showed the tensile strength of the simple cellulose films prepared using different concentrations of carboxymethyl cellulose (1, 2, 3 & 4) % and plasticizers (glycerol 1 and sorbitol) with concentrations (20, 40 & 60) % (table 1). The highest of tensile strength 51.3 MPa prepared from 1%CMC + 20% glycerol and a lowest strength 40.4 MPa for glycerol, while sorbitol had the highest strength 36.4 MPa prepared from 2% CMC + 40 % Sorbitol and a lower strength 28 MPa prepared from 3%CMC + 60% sorbitol. It is noted that the tensile strength of calculated polycrystalline films is higher than that of sorbitol-plasticized films. This is due to the interaction between the hydroxyl group of glycerol and the CMC-like

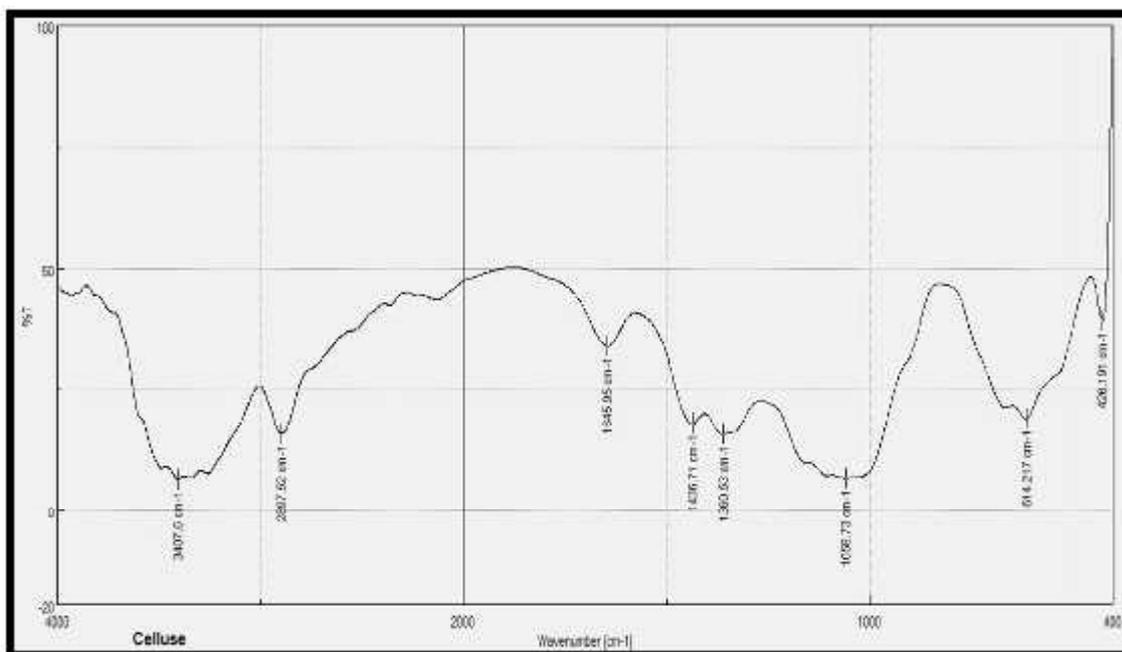


Fig. (1): Infra-red spectroscopy of cellulose.

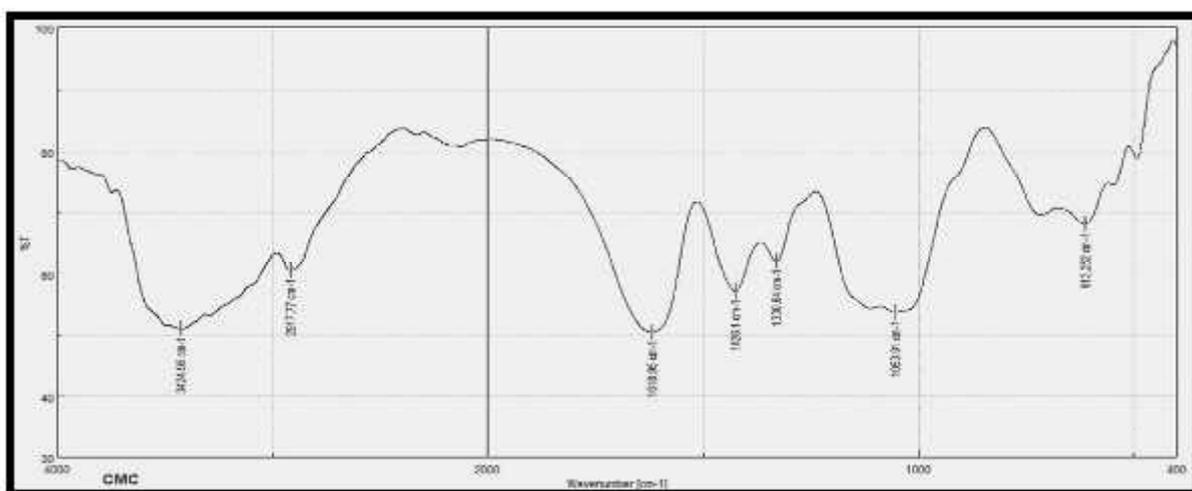


Fig. (2): Infra-red spectroscopy of the carboxymethyl cellulose created from cellulose

**Mechanical and reservation properties of simple cellulose films.**

carboxymethyl during the mixing and drying of films (Tongdeesontorn *et al.*, 2011). Bipolar reactions and the effect of existing charges can also increase the tensile strength (Li *et al.*, 2008). The higher concentration of the plasticizer is, the less the tensile strength; this is due to the ability of these materials to break down the hydrogen bonds between the polymer chains (Asl *et al.*, 2017). When comparing these results with other studies, they were consistent with the results of Nor *et*

*al.* (2017), noting that decreased tensile strength with increased concentration of the glycerol when preparing gelatinous films of chicken skin. This results in a reduction in tensile strength, according to Ahmadi *et al.* (2005), noting that the films made from carrageenan had the highest tensile strength of 1.25% by 102.50 MPa. When the concentration increased to 1.5%, the tensile strength decreased to 83.33 MPa; this is due to the increase in the strength of the

molecules between the polymer chain of the film, which made the film more rigid and fragile. The results showed no significant differences between the concentration of carboxymethyl acid and cellulose at a potential level ( $P < 0.05$ ).

#### **Elongation at Break (EAB)**

The results showed the change in the percentage of elongation of the simple cellulose films prepared using different concentrations of carboxymethyl cellulose (1, 2, 3 & 4) % and two types of plasticizers (glycerol and sorbitol) and different concentrations (20, 40 & 60) % (table 1). The highest percentage of elongation was 90.5% for the prepared film (4% CMC + 60%) and the lowest elongation rate (78.6%) for the prepared film (1% CMC + 20) %, As for the films prepared from sorbitol reached the highest elongation rates 81.5% for the prepared film (4% CMC + 60% sorbitol) and less elongation 65.5% for the prepared film (1% CMC + 20% sorbitol) between the polymer chains are prevents the formation of bonds of hydrogen and reduces the movement of molecules and thus the strings become capable of movement and increased flexibility, and also the CMC raises the viscosity of the film solution and cause greater flexibility in the polymer structure (Tongdeesontorn *et al.*, 2011). These results were consistent with Asl *et al.* (2017) and Rachtanapun (2009), who indicated that the elongation to the cut increases with the increased concentration of CMC. The results showed significant differences in the concentration of CMC on elongation at a potential level ( $P < 0.05$ ).

#### **Thickness**

The finding (Table 1) showed the change in the thickness of the simple cellulose films prepared using different concentrations of

carboxymethyl cellulose (1, 2, 3 & 4) % and different types of plasticizers (glycerol and sorbitol) with concentrations (20, 40 & 60) %. The thickness of the film ranged between (0.018-0.046) mm for the cities of the glycerol and (0.045-0.078) mm for the sorbitol cities. Therefore, the thickness of the Plasticization of sorbitol was higher than the thickness of the glycerol films. This may be due to the fact that sorbitol can penetrate the polymer network more quickly and more easily than the glycerol. Formation of stronger and thicker films that in turn can be hoped to the solid components of CMC as there is a direct relationship between the thickness and the concentration of the constituent material of the film, which applies to all polymers of water (polar) and the results were consistent with the findings of Tharanathan, (2003) and Tabari (2017). It should be noted that the thickness can increase with the increase of the proportion of plasticizer (glycerol) as a result of the overlap between the components of plasticized with the components of the film and increase the proportion of solids, making the resulting film thicker (Prodpran & Benjakul, 2005), and note the results of significant differences between the concentration the CMC of the film and the thickness of the resulting films are at a probabilistic level ( $P < 0.05$ ).

#### **Solubility in water**

Solubility is a major characteristic of edible films that are related to structural properties and film structure. The results of table (1) showed that the solubility of simple cellulose films prepared using different concentrations of carboxymethyl cellulose (1, 2, 3 & 4) % and two types of plasticizers (glycerol and sorbitol) at concentrations of (20, 40 & 60) %. The results showed that the highest solubility rate reached 36.1% for films prepared from

1% CMC + 20% glycerol while less than a reached 25.3% for films prepared from (4%CMC + 60% glycerol), and for plasticized with sorbitol, the highest solubility rate was 25.02% for films prepared from 1% CMC + 20% sorbitol the lowest percentage (19.05%) for films prepared from 4% CMC + 60% sorbitol, and therefore the value of solubility decreases as the percentage. This can be attributed to the interaction of the glycerol with the carboxymethyl cellulose by means of the astride bonds, which increases the size of the particles and surface area, leading to reduced solubility. Sorbitol increases the solubility due to its hydrophilic nature, which gives the polymer molecules a high ability to attract water. Reduces interactions between polymeric polymer molecules (Chick & Ustunol, 1998). It was found that there was an inverse relationship between the concentration of carboxymethyl cellulose in the preparation of film solution and the susceptibility of films to water solubility. The results showed that the increased concentration reduced the solubility, which reached 36.31% at the concentration of 1%, while 25.32% at the concentration of 4%. The decrease in solubility has led to increase concentrations of carboxymethyl cellulose due to increased estrogen bonds, which is consistent with the findings of Zillo *et al.* (2018) and Dashipour *et al.* (2015). Significant differences were found between the prepared films at a potential level ( $P < 0.05$ ).

### Water Vapor Permeability

The results (Table 1) showed the values of permeability of simple cellulose films for water vapor prepared using different concentrations of carboxymethyl cellulose (1, 2, 3 & 4) % and two types of plasticizers glycerol and sorbitol in concentrations (20,

40% & 60) %, as the table showed that the highest permeability of water vapor was 11.99 gm.mm /m<sup>2</sup>.h.kpa for films prepared from (4%CMC + 60% glycerol) and less permeability 5.2 gm.mm / m<sup>2</sup>.h.kpa for films prepared from (3% CMC + 40% glycerol), and for Plasticization with sorbitol reached highest permeability 7.75 gm.mm / m<sup>2</sup>.h.kpa for films prepared from (4% CMC + 60% sorbitol) and less permeability 5.43 gm.mm/m<sup>2</sup>.h. kpa for films prepared from 2%CMC + 20% sorbitol, where Ma *et al.* (2008) showed that there was a positive relationship between the concentration of CMC and the concentration of glycerol and permeability, which is increasing. This is due to the increase in carboxymethyl groups that were replaced with cellulose structure, as well as increased permeability due to increased cellulose polarity when converting it to carboxymethyl cellulose. The relative relationship between the concentration of the material involved in film preparation and permeability. As the concentration increased, the permeability values increased to a concentration of 4%. This was indicated by the results, which showed that increasing the concentration leads to increase of the pair while stirring the solution and thus the appearance of air bubbles, as a result, grow up air openings formed in the film, causing the increased permeability (Rachtanapun & Rattanapanone, 2011). The Plasticization with glycerol films was more permeable than plasticization than sorbitol. The reason is that sorbitol is more or less suitable for water molecules than the structure of the crystal, as well as differences in structure and hydrophilic properties. Thus, the periodic formation of sorbitol molecules is the main factor that reduced permeability values (Zhang & Han, 2006).

**Table (1): Mechanical and sequential properties of small cellulose films prepared from different concentrations of carboxymethyl cellulose with two types and concentrations of plasticizers (glycerol and sorbitol).**

Sample number	Ensil strength (MPa)	Elongat ion at break%	Thickness (mm)	Solubi lity%	Permeabil ity (gm.mm / m <sup>2</sup> .h. KPa)
1%+20%	51.3	78.6	0.018	36.31	6.47
1%+40%	46.8	81.2	0.021	35.11	7.26
1%+60%	40.8	83.9	0.023	34.55	11.33
2%+20%	48.6	84.5	0.026	33.91	5.8
2%+40%	47.2	84.7	0.028	32.57	6.15
2%+60%	45.9	85.8	0.031	31.61	7.77
3%+20%	48.1	87.8	0.034	30.73	5.2
3%+40%	46.5	88.4	0.037	29.26	6.26
3%+60%	44.2	91.0	0.039	28.53	10.36
4%+20%	42.9	89.6	0.040	27.85	10.21
4%+40%	41.5	89.8	0.043	26.41	11.02
4%+60%	40.4	90.5	0.046	25.32	11.99
1%+20%	35.8	65.5	0.045	25.02	5.98
1%+40%	34.6	65.9	0.048	23.31	6.05
1%+60%	31.0	66.3	0.050	23.49	7.71
2%+20%	36.2	70.2	0.052	23.44	5.43
2%+40%	36.4	71.3	0.055	23.28	6.34
2%+60%	33.8	71.8	0.057	23.01	7.12
3%+20%	29.3	75.6	0.061	21.67	5.86
3%+40%	29.1	76.4	0.067	20.39	6.17
3%+60%	28.0	77.1	0.069	20.04	6.69
4%+20%	30.7	80.5	0.072	19.85	5.48
4%+40%	30.4	81.2	0.074	19.23	6.13
4%+60%	30.0	81.5	0.078	19.05	7.75
R.L.S.D Concentration of CMC	N.S	7.456	0.019	5.012	1.985
R.L.S.D Plastic Type	N.S	N.S	N.S	N.S	1.922

The increased concentration of plasticizers reduces the molecular forces between polymer chains, increases free volume, segmental motions, allowing water molecules to diffuse easily and give higher permeability (Sothornvit & Krochta, 2001). The results were consistent with (Ghanbarzadeh *et al.*, 2007; Ghanbarzadeh and Almasi, 2011) who

observed that increasing the concentration of the glycerol increased the permeability of the zein films. Significant differences were also observed in the CMC concentration and the plasticity on the prepared films at a potential level ( $P < 0.05$ ).

## Conclusions

The results found that the possibility of synthesis of a polymer carboxymethyl cellulose extracted from flour bran. The possibility of preparing cellulose films from flour bran derivatives with good reserve and mechanical properties. Properties of films have been changed by type and proportion of plasticized and the concentration of carboxymethyl cellulose.

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