



Scientific Research

## Biodegradable film of Sodium alginate film/flax seed mucilage/norbixin/tungsten oxide: investigation of color, crystalline, thermal, mechanical and antibacterial properties

Dadkhah, H. <sup>1\*</sup>, Pirsas, S. <sup>2</sup>, Javadi, A. <sup>3</sup>, Mohtarami, F. <sup>2</sup>

1. Department of Food Science and Technology, Mamaghan Branch, Islamic Azad University, Mamaghan, Iran
2. Department of Food Science and Technology, Faculty of Agriculture, Urmia University, Urmia, Iran
3. Department of Food Hygiene, Faculty of Veterinary, Tabriz Medical Sciences, Islamic Azad University, Tabriz, Iran.

### ARTICLE INFO

#### Article History:

Received 2023/ 01/ 11  
Accepted 2023/ 03/ 11

#### Keywords:

Biodegradable film,  
Thermal resistance,  
Mechanical resistance,  
Antibacterial polymers and Smart packaging.

DOI: 10.22034/FSCT.19.135.31  
DOR: 20.1001.1.20088787.1402.20.135.4.0

\*Corresponding Author E-Mail:  
H.d2960050241@gmail.com

### ABSTRACT

In this research, flax seed mucilage was extracted. Composite film of sodium alginate and flax seed mucilage was prepared. Norbixin pigment and tungsten oxide (WO<sub>3</sub>) nanoparticles were used to modify the film structure. The color, crystallite, thermal and mechanical properties of the films were investigated. Also, the antibacterial properties of the prepared films against *Escherichia coli* and *Staphylococcus aureus* bacteria were investigated. The obtained results showed that the pure alginate/mucilage film does not have very high transparency, which is reduced by adding tungsten oxide nanoparticles and norbixin pigment. The effect of tungsten oxide nanoparticles in reducing film transparency has been greater than that of norbixin. Examining the factor a shows that this factor has increased with the increase of norbixin and tungsten oxide nanoparticles. Examining factor b (blue-yellow) shows that with the increase of norbixin and tungsten oxide nanoparticles, this factor has increased. By examining the XRD spectrum of the pure alginate/mucilage film, it was found that this film showed two broad peaks at 2θ of 10° and 20°, which indicates the relatively amorphous structure of this film. In the alginate/mucilage film modified with tungsten oxide nanoparticles, the peaks related to the crystalline nanoparticles in 2θ of approximately 25, 30, 35, 40, 50, 55 and 65 degrees are quite clear, which shows that these nanoparticles improve the crystalline structure of the film. By examining the TGA curves of the films, it was found that the addition of tungsten oxide nanoparticles and norbixin pigment increased the thermal stability of the film. Examining the antibacterial property of the films showed that the addition of tungsten oxide nanoparticles and norbixin pigment increased the antibacterial property of the film significantly (p<0.05).

## 1. Introduction

In recent years, environmental problems and biological pollution caused by synthetic and petroleum polymers have made researchers think to look for suitable alternatives for these polymers in order to reduce the pollution caused by synthetic polymers and provide new polymers with new uses. Based on this, in recent years, the use of natural polymers as environmentally friendly and biodegradable materials has attracted the attention of researchers all over the world.]3-1[. Biodegradable polymers are prepared from natural sources such as mucilage and plant gums or animal sources such as proteins. Biodegradable and biobased polymers have a wide range and have many applications in pharmaceuticals, medicine, horticulture, agriculture, consumer electronics, automobiles, textiles, and especially packaging.]9-4[.

Flax seeds are rich in healthy omega-3 and omega-6 fats Dietary fibers. Among other properties of flax seed is its high antioxidant property. Lignan (antioxidant found in flax seed mucilage) is a type of polyphenolic fiber that has antioxidant properties. Regulation of hormones, cellular health and anti-aging properties are among the features of lignan. Polyphenols help the growth of probiotics (live microorganisms that are beneficial to health) and are effective in removing yeast and candida from the body. Lignans have antimicrobial and antiviral properties. Therefore, their regular consumption can reduce the frequency of colds and flu. One of the most important properties of flax seeds is having a high amount of mucilage. Mucilage is a type of fiber with a jelly state and soluble in water. Mucilage prevents the rapid emptying of food from the stomach to the small intestine. As a result, eating flax seeds causes a longer feeling of satiety and causes us to consume fewer calories and leads to weight loss.]13-10[.

Sodium alginate is a linear polysaccharide derived from alginic acid, which is present in the cell wall of brown algae. In recent decades, alginates have attracted the attention of researchers due to their unique physical and chemical properties and their wide applications as a natural polymer. This salt compound is derived from alginic acid. It is a polysaccharide acid that is widely present in the cell walls of brown algae

and forms a sticky gum when hydrated and has the ability to form polymer films. This material creates elastic gels in acidic and calcium-containing environments and is also known as a cold gelling agent; Because it does not need any kind of heat to gel. Sodium alginate is also used in the pharmaceutical industry due to its gelling and thickening properties. Of course, apart from the role it plays as an additive, this substance also has other uses in the world of medicine and pharmaceuticals.]16-14[.

Annatto color (Annatto) is an orange-colored food seasoning that is extracted from the seeds of the achiote tree that grows in the tropical regions of South and Central America. Annatto color is a natural food color that gives food a color similar to saffron or turmeric. Annatto contains many plant compounds with antioxidant properties such as carotenoids, terpenoids, flavonoids and tocotrienols. Antioxidants are compounds that can neutralize potentially harmful molecules called free radicals, which can damage your cells if they build up too much. Research has shown that damage caused by high levels of free radicals will lead to chronic diseases such as cancers, brain disorders, heart disease and diabetes. Also, research shows that this food coloring may have antimicrobial properties. Laboratory studies have shown that annatto extract inhibits the growth of various bacteria, including *Staphylococcus aureus* And *Escherichia coli* prevents. In addition, adding this color to bread will inhibit the growth of fungi and increase the shelf life of bread[17 and 18].

Tungsten oxide chemical compound with the formula  $WO_3$  is. It is a yellow solid crystal. In recent years, the structures of this oxide have attracted much attention due to their unique properties. Tungsten oxide is a versatile material with many advantages including availability and low manufacturing cost. The favorable semiconducting properties of this chemical compound allow it to be used as a photocatalyst. This metal oxide has a very low cost to produce, has very little toxicity to living organisms, and is environmentally friendly. These characteristics together with a high chemical stability in the pH range related to many applications and its semiconductor properties have led to its wide use in various industries. Tungsten oxide powder ( $WO_3$ ) Or Nano atoms In the form of nanofluids or

oxide particles with a high surface area, they have magnetism. Tungsten oxide photocatalyst is a type of light radiation, this chemical mixture cannot change itself during a chemical reaction, but it can increase the speed of chemical reactions. A photocatalyst naturally works by becoming a catalyst. Recent research of the specialty antibacterial Tungsten oxide has also been proven [20 and 19].

Considering the problems of environmental pollution caused by synthetic polymers and considering the useful features mentioned about natural polymers, flax seed mucilage, sodium alginate, norbixin pigment and tungsten oxide, the production of a film composed of these natural and beneficial materials will be very useful. Was. Therefore, in this research, an environmentally friendly film was prepared from natural materials with antimicrobial capabilities, which can be used in active food packaging and increase the shelf life of food sensitive to microbial spoilage.

## 2- Materials and methods

### 2-1- Chemicals

Flax seeds were obtained from medicinal plant store (Urmia, Iran). Tungsten oxide nanoparticles powder was purchased from Merck (Germany). Sodium alginate was purchased from Tinacom Dartabriz (Tabriz, Iran). 5% Norbixin solution was prepared and used from Araz Kimiai Jahan Company (Tabriz, Iran). Other chemical compounds such as silica gel, calcium sulfate, potassium sulfate, 2,2-diphenyl-1-picrylhydrazyl (DPPH), etc. were purchased and used from Merck (Germany) and Aldrich (USA).

### 2-2- Flax seed mucilage extraction

To extract the mucilage, flax seeds were mixed with distilled water at a ratio of 1 to 40 and heated with stirring (mechanical stirrer) at 50 degrees Celsius for 2 hours. In order to separate the insoluble components of the seed from the mucilage part, it was passed through a mesh cloth

in two steps and the mucilages were separated and kept in the refrigerator at 0°C until the tests.

### 2-3- Preparation of sodium alginate film/linseed

#### mucilage/norbixin/tungsten oxide

To prepare the films, 3 grams of flax seed mucilage was mixed with 100 ml of distilled water, then it was placed on a magnetic heater with a magnet for 15 minutes and the solution was passed through a filter. Next, 2 grams of sodium alginate was added to the solution and it was placed on a magnetic heater with a magnet for another 15 minutes to prepare a uniform jelly solution. Then, as a softener, 2 ml of glycerol was added to the above solution and stirred again for 5 minutes. Then the solution of Norbixin and tungsten oxide was added to the solution according to the statistical plan (Table 1-A) and then the mixed materials were placed on a heater at a temperature of 60 degrees Celsius for 15 minutes. Finally, mix for 15 minutes in an ultrasonic device (Hielscher UP400ST, Germany) It was ultrasounded and then heated for 5 minutes. 30 ml of the prepared solution was poured on polyethylene molds with a diameter of 10 cm and dried in ambient conditions for 48 hours. During this time, films of a certain thickness were prepared, and finally the prepared films were separated from the mold and stored in special zippered bags (Figure 1-a).

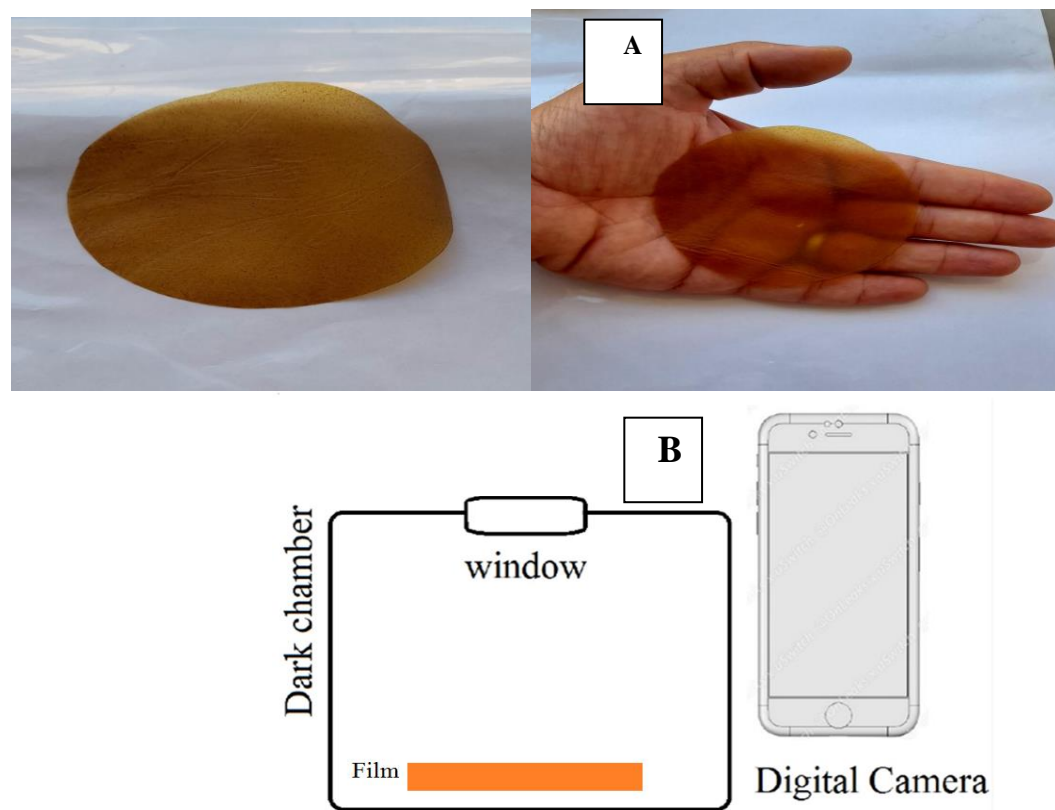
### 2-4- Measurement of color characteristics

To measure the color factors, a special system was designed as follows: a light-insulating plastic chamber was designed with dimensions of 10 x 10 x 20 cm, and a valve of 1 x 1 cm was installed in its upper part. This hole is built to place the camera lens. Color factors in terms of L (lightness-darkness), a (red-green) and b (yellow-blue) were measured by Samsung digital camera and using Color-Grab software according to Figure 1-b. Color-Grab software was placed in Lab mode and color factors were recorded.

[ DOI: 10.22034/FSCT.20.136.38 ]

[ DOR: 20.1001.1.20088787.1402.20.136.4.2 ]

[ Downloaded from fsct.modares.ac.ir on 2024-05-13 ]



**Fig 1** Examples of produced films (A) and film color factor measurement system (B)

## 2-5- X-ray diffractometer (XRD)

by XRD technique and with the help of X-ray diffractometer (Kristalloflex D500, Siemens, Germany) To investigate the crystalline or amorphous structure used. In this technique, film samples were first placed in the special position of the device (sample cell). The primary rays were irradiated to the sample at ambient temperature and the reflected rays were collected in the range of angle  $2\theta = 0-80$ . The XRD spectra of the samples were drawn automatically by the device. Cu  $K\alpha$  radiation source was used at a wavelength of 0.154 nm. The specifications of the device include X-ray generator at 40 kV and 40 mA.

## 6-2- Thermal gravimetric analysis (TGA)

To investigate the thermal stability of films from TGA test will be used by thermal analyzer (Linseis – L81A1750, Germany). For this purpose, films will be prepared in the form of 10 mg samples. Film samples under nitrogen atmosphere  $50 \text{ cm}^3/\text{min}$  in aluminum cups in the temperature range of 30-600 °C. The heating rate

will be 10 °C/min. The empty aluminum cup will be considered as a reference and the TGA curve will be drawn and recorded by the device.

## 2-7- Mechanical characteristics

The mechanical properties of the films include stretchability up to breaking point (EB) and tensile strength (TS) which are important factors for food packaging. To check the mechanical properties, the samples were conditioned for 24 hours in special conditions. Special conditions include relative humidity of 55%. The film was cut with a special cutter in the dimensions of 1 x 8 cm in a dumbbell shape. The films were placed between the two jaws of the device, and the initial distance between the two jaws was 50 mm. The upper jaw was moved relative to the lower jaw at a speed of 5 mm/min. The mechanical properties were recorded by a computer. From tissue measuring device with brand (Zwick/Roell model FR010, Germany) was used to perform this test.

## 8-2- Antimicrobial property

With a slight change, the agar diffusion method was used to determine the antibacterial properties

of the films. For this purpose, the films in the form

Discs (with a diameter of 15 mm) are cut and then on

on Mueller Hinton agar plates containing *Staphylococcus aureus* ATCC6538 and *Escherichia Coli* ATCC13706, with a concentration of ( $10^7$ CFU/mL) were placed. The plates were incubated for 24 hours at 37 degrees Celsius. After 24 hours, the radius of the halos *No bacterial growth* It was measured around the films (in millimeters) with a precise caliper.

## 9-2- Statistical analysis

The statistical analysis and analysis of the data obtained in this research was done in two parts. In the first part, the central composite statistical design (CCD) was used to investigate the effect of two factors, the amount of norbixin (ml) and the amount of tungsten oxide (mg) on the color properties (Table 1-A). Design Expert-10 software was used to design experiments and analyze data at the 95% probability level and draw 3D curves. In the second part, a factorial design was used at the 95% probability level to investigate the effect of two factors, the amount of norbixin (ml) and the amount of tungsten oxide (mg) on the crystalline, thermal and mechanical properties (Table 1-b).

**Table 1 A** List of films prepared based on the central composite design

Film	A: WO <sub>3</sub> (mg)	B: Norbixin (ml)
1	0.00	2.50
2	50.00	2.50
3	100.00	5.00
4	50.00	0.00
5	0.00	5.00
6	50.00	2.50
7	100.00	2.50
8	0.00	0.00
9	50.00	2.50
10	50.00	2.50
11	100.00	0.00
12	50.00	2.50
13	50.00	5.00

**Table 1 B** List of tests performed for crystalline, thermal and antimicrobial properties

Film	A: WO <sub>3</sub> (mg)	B: Norbixin (ml)
*Control (Pure Muc/Al)	0	0
Muc/Al/WO	100	0
Muc/Al/NB	0	5
Muc/Al/WO/NB	100	5

\* **Muc**: Mucilage; **Al**: Alginate; **WHERE**: WHERE<sub>3</sub>; **NB**:Norbixin

## 3. Results and Discussion

### 3-1- Color review of movies

The color of food packaging films is very important and significant. Because the color of food packaging, in addition to preventing light oxidation of food sensitive to light oxidation, can also be effective in attracting customers. Also, in some materials, the presence of photocatalytic

nanoparticles in the packaging film can have antibacterial properties by absorbing light while changing the color of the film surface with photocatalytic activity.

Figure 2 shows the color factors of mucilage/alginate film and its composites. The analysis of the L factor shows that the pure mucilage/alginate film is not very transparent and shows a transparency of about 55, which is



reduced by the addition of nanoparticles and Norbixin pigment, so that the mucilage/alginate film that contains both tungsten oxide nanoparticles and Norbixin It has the least transparency. The effect of tungsten oxide nanoparticles in reducing film transparency has been greater than the effect of Norbixin. Due to the fact that nanoparticles are physically distributed among the polymer chains, they fill the empty spaces of the polymer and prevent the passage of light and lead to a decrease in transparency.

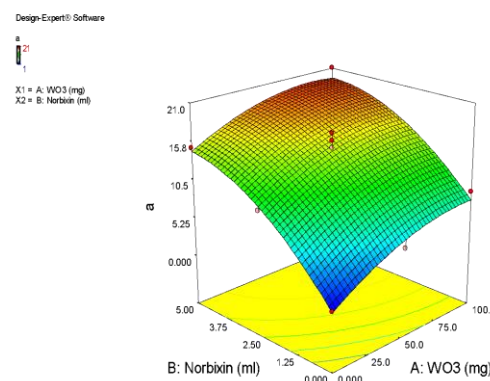
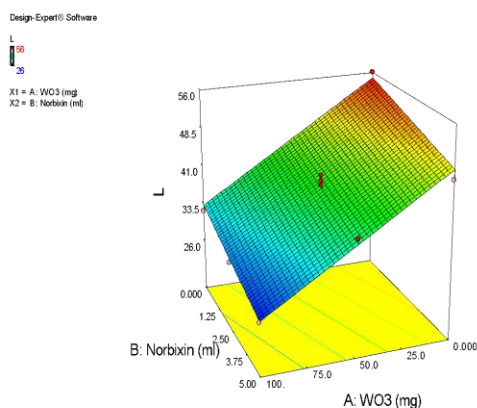
Examining factor a (green-red) shows that with the increase

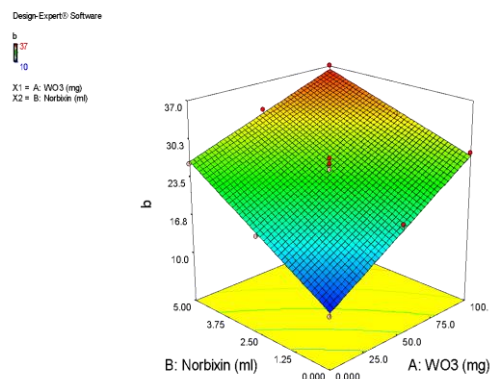
Norbixin and tungsten oxide nanoparticles have increased this factor, that is, it has moved towards more positive numbers that indicate redness. The effect of Norbixin in increasing factor a was more than that of tungsten oxide nanoparticles. According to the nature of the norbixin pigment, which has a yellow-orange color, an increase in factor a is expected due to the presence of norbixin. Tungsten oxide nanoparticles also have

a somewhat yellow color, and these particles also increase the a factor.

Also, the examination of factor b (blue-yellow) shows that with the increase of Norbixin and tungsten oxide nanoparticles, this factor has increased, that is, it has moved towards more positive numbers that indicate yellowness. The effect of Norbixin in increasing factor b is almost equal to that of tungsten oxide nanoparticles. Considering the nature of the pigment norbixin, which has a yellow-orange color, and the tungsten oxide nanoparticles also have a somewhat yellow color, an increase in factor b is expected due to the presence of norbixin and tungsten oxide nanoparticles.

Priyadarshi et al. (2021) They have investigated the effect of additives (nanoparticles) on the color characteristics of alginate film. They reported that nanoparticles can affect the color factors, transparency and light transmission of films. The results of this research are in good agreement with the results Priyadarshi et al]21[.





**Fig 2** Three-dimensional curves of the effect of WO<sub>3</sub> and norbixin on color factors of mucilage/alginate film and its composites

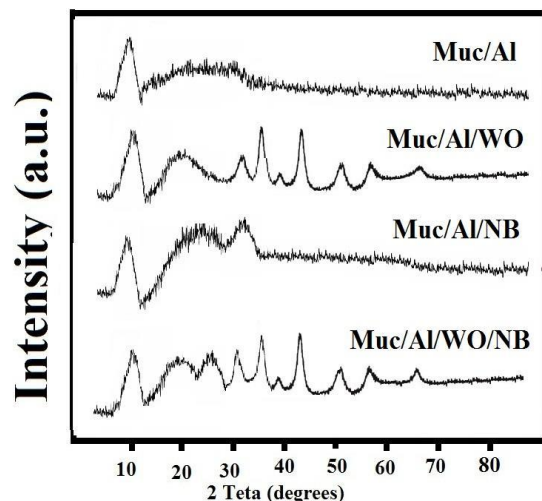
### 3-2- Investigation of crystalline properties

Figure 3 shows the XRD spectra of flax seed mucilage film/sodium alzinate and its various composites. By examining the spectrum of the pure mucilage/alginate film, it was found that this film shows two broad peaks at 2 theta of 10 and 20 degrees, which indicates the relatively amorphous structure of this film. The research conducted on the structure of pure sodium alginate film has introduced this film as a semi-crystalline film due to the presence of a peak in the region of 10 degrees, which is also seen in this study at 2 theta of 10 degrees, but since Previous studies have confirmed the amorphous structure of flaxseed mucilage, and considering that alginate and mucilage were used to prepare the film, the prepared film has a semi-crystalline structure with a tendency to form an amorphous structure. In the mucilage/alginate film modified with tungsten oxide nanoparticles, the peaks related to the crystalline nanoparticles in 2 theta approximately 25, 30, 35, 40, 50, 55 and 65 degrees are quite clear, which shows that these nanoparticles have improved the crystalline structure of the film. . The modified mucilage/alginate film also shows the semi-crystalline structure of the film. According to what has been reported in the references, norbixin is a semi-crystalline pigment with peaks in 2 theta between 15 and 30 degrees, and these peaks are also observed in the present study, and the presence of norbixin in the film structure is also confirmed. In the XRD spectrum of mucilage/alginate composite film modified with tungsten oxide nanoparticles and norbixin, all the

peaks related to the components of the film are observed, and the semi-crystalline to crystalline structure of the composite film is clearly evident. It is worth mentioning that the peak of the 20° 2-theta region is present in both mucilage, alginate and pure Norbixin, which overlaps this peak in the composite film and is observed as a single peak.

Prado et al. in 2018 have investigated the crystalline/amorphous structure of flax seed mucilage film. The results of their research confirm the results of the present research [22]. Bhagyaraj and Krupa (2020) studied the structure of sodium alginate composite using XRD technique. They reported that the film has a semi-crystalline structure, which is in good agreement with the results of the present study]23[. Perotti et al. (2020) have confirmed the semi-crystalline structure of annatto using XRD technique, and the results of this research showed good agreement with their results.]24[. Pang et al. (2012) have investigated different crystal structures of tungsten oxide. The results of their research in terms of peaks related to tungsten oxide in the XRD spectrum confirm the results of the current research]25[.





**Fig 3** XRD spectra of flax seed mucilage film/sodium alginate and its various composites

### 3-3- Examination of thermal properties

Figure 4 shows the TGA curve of mucilage/alginate film and its composites. As it is clear from the curves, in all 4 examined films, weight loss due to temperature increase has occurred in two stages. The weight loss of the first stage has occurred in the temperature range of 70 to 130 degrees Celsius, which is related to the evaporation of water molecules and other possible volatile compounds such as glycerol in the structure of the films. The weight loss of the second stage occurred in the temperature range of 200 to 280 degrees Celsius, and this weight loss is related to the destruction of the polymer

structure of the composite film. By more detailed examination and comparison of the TGA curves of the films, it is clear that the weight loss of the second stage in the modified films with tungsten oxide nanoparticles and Norbixin occurred at a higher temperature than the pure mucilage/alginate film, and also in the modified films. The weight loss in the second stage was less compared to pure film. These results show that tungsten oxide nanoparticles and norbixin have increased the thermal stability of the film. Considering that the temperature of tungsten oxide nanoparticles degradation is very high, therefore, reducing the degradation rate of the second stage in the presence of tungsten oxide seems natural. Also, it is possible that Norbixin and tungsten oxide nanoparticles improve the chemical connections between polymers by creating electrostatic interactions (the result confirmed by FTIR spectra) with polymer chains of alginate and flax seed mucilage, which leads to the thermal stability of the composite film. has been

Salis et al. (2016) and Prado et al. (2018) have investigated the thermal behavior of alginate films and flax seed mucilage film. The results of this research confirm the results of the present research [22 and 26]. Tehet et al. (2004) have investigated the thermal behavior of tungsten oxide, and their research results confirm the effect of tungsten oxide on the thermal behavior of biodegradable film [27].

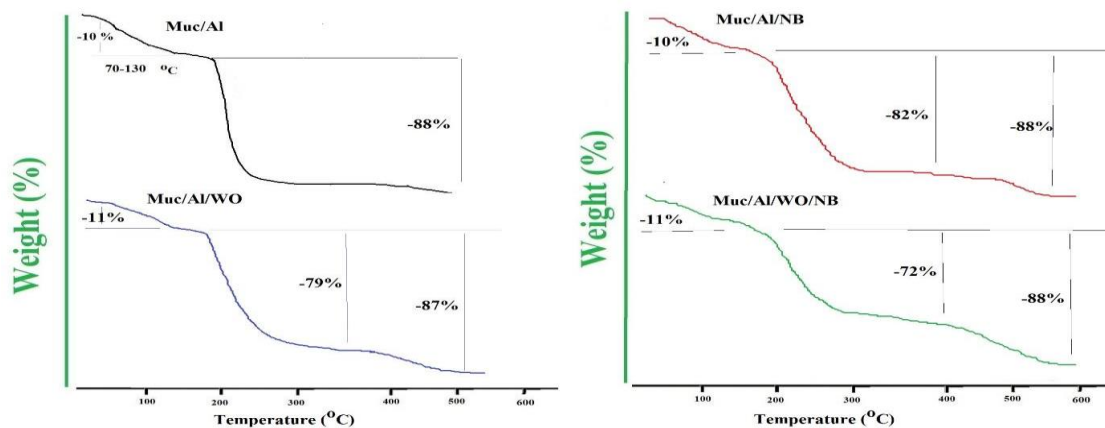


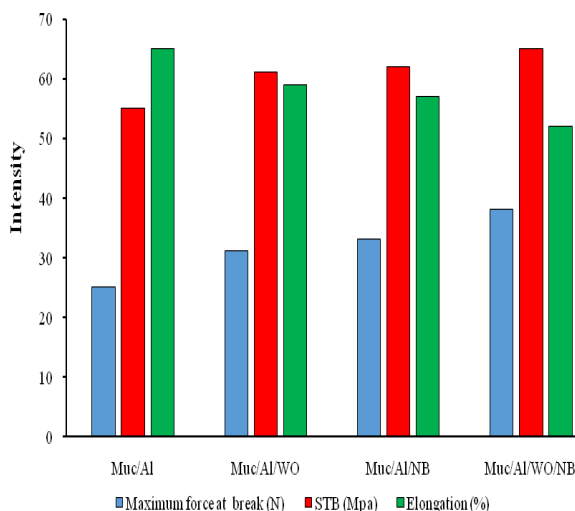
Fig 4 TGA curve of mucilage/alginate film and its composites

### 3-4- Examination of mechanical properties

Figure 5 shows the column graphs of mechanical properties of mucilage/alginate film and its various composites. The obtained data was the average of three repetitions. Examining the mechanical results of the films shows that the pure mucilage/alginate film has good mechanical resistance. This film has a tensile strength of up to 65%, and its tensile strength is 55 MPa, which is suitable for food packaging. By comparing the column graphs, it can be concluded that the addition of tungsten oxide nanoparticles and norbixin pigment has increased the tensile strength and the maximum force required to break the film, so that the film containing both materials (tungsten oxide nanoparticles and norbixin pigment) has the highest tensile strength and maximum It is the necessary force to break the film. The reason for this behavior can be explained by adding tungsten oxide nanoparticles and norbixin pigment to the film, creating electrostatic forces between mucilage/alginate polymer chains and tungsten oxide nanoparticles and norbixin pigment, and increasing the structural strength of polymer chains. This phenomenon is also observed in the FTIR spectra, which confirms the creation of electrostatic interactions between mucilage/alginate polymer

chains and tungsten oxide nanoparticles and norbixin pigment. Although the addition of tungsten oxide nanoparticles and norbixin pigment to the film has increased the tensile strength and the maximum force required to break the film, on the other hand, it has decreased the elasticity of the film. The decrease in the elasticity of the films is perhaps due to the greater structural strength of the films and the strong transverse connections of the film in the presence of tungsten oxide nanoparticles and norbixin pigment, which causes the film to lose its flexibility to some extent.

Pereira et al. (2011) have investigated the mechanical properties of alginate film and its various composites. The results of their research confirm the results of the present research to a large extent [28]. Prado et al. (2018) have investigated and confirmed the effect of nanostructured additives on improving the mechanical properties of the film based on flaxseed mucilage. The results of this research are in good agreement with the results of Prado et al]22[.



**Fig 5** Bar graphs of mechanical properties of mucilage/alginate film and its different composites

### 3-5- Property antibacterial

Movies and Polymers Antibacterials called pesticide polymers are also introduced. These films are a group of polymer materials that have antimicrobial properties and can prevent the growth of very small organisms such as fungi and bacteria. Antibacterial polymers are engineered in such a way that they can behave similarly to antibacterial peptides and can destroy spoilage agents in food and other packaging. Usually, antibacterial films and polymers are produced by connecting or placing an antibacterial agent (such as nanoparticles, extracts or pigments) inside the main chain of the polymer material with the help of alkyl or acetyl linkers. These films in the packaging of food products by inhibiting the growth of spoilage bacteria, delay the degradation of food and increase the shelf life of food.

Table 2 shows the antibacterial properties of two types of Gram-negative (*Escherichia coli*) and Gram-positive (*Staphylococcus aureus*) bacteria in the presence of mucilage/alginate film and its composites. Examining the antibacterial properties of 4 types of films shows that all films

(even the pure mucilage/alginate film) have antimicrobial activity against Gram-positive and Gram-negative bacteria. The addition of tungsten oxide nanoparticles and norbixin pigment has increased the antibacterial property of the film significantly ( $p < 0.05$ ). The most antibacterial property is related to mucilage/alginate film modified with tungsten oxide nanoparticles and norbixin.

The fact that the pure mucilage/alginate film also had antibacterial properties can be due to two reasons, firstly, this film can physically inactivate bacteria and show antibacterial properties, and secondly, the mucilage of plant seeds contains polyphenols. The rest are in their structure, which can induce antibacterial properties to the film. The antimicrobial behavior of nanoparticles has been investigated and confirmed many times by researchers, the research conducted on tungsten oxide confirms its antimicrobial properties (Duan et al., 2019). Penetration of tungsten oxide nanoparticles into the cell structure of bacteria and disrupting the life cycle of bacteria causes the destruction of bacteria and destroys them. Also, previous researches have confirmed the antibacterial property of Norbixin pigment [29]. By comparing the antibacterial properties of 4 films against Gram-positive and Gram-negative bacteria, it was found that all 4 films had more antimicrobial activity against Gram-positive bacteria (*Staphylococcus aureus*) than Gram-negative bacteria (*Escherichia coli*). Since the structure of the cell wall in Gram positive bacteria is simpler than in Gram negative bacteria and the penetration of antimicrobial agents (nanoparticles and pigments) into the bacterial cell and their destruction is easier in Gram positive bacteria, so the result is observed. It was to be expected. Viuda-Martos et al. (2012) extracted annatto extract and investigated its antibacterial properties. The results of their research confirm the results of the present research [30].

**Table 2** Antibacterial properties of mucilage/alginate film and its composites

Diameter of non-growth area (mm)		B: Norbixin (ml)	A: WO <sub>3</sub> (mg)	Film
<i>E. coli</i>	<i>S. aureus</i>			

12±0.3 <sup>a</sup>	18±0.65 <sup>a</sup>	0	0	Control (Pure Muc/Al)
23±0.4 <sup>c</sup>	28±0.8 <sup>c</sup>	0	100	Muc/Al/WO
17±0.4 <sup>b</sup>	22±0.3 <sup>b</sup>	5	0	Muc/Al/NB
25±0.7 <sup>d</sup>	32±0.7 <sup>d</sup>	5	100	Muc/Al/WO/NB

\* In each column, the non-identical letter indicates the significance of the differences at the 95% probability level.

## 4 - Conclusion

Considering the problems of environmental pollution caused by synthetic polymers and considering the useful features mentioned about natural polymers, flax seed mucilage, sodium alginate, norbixin pigment and tungsten oxide, the production of a film composed of these natural and beneficial materials will be very useful. Was. Therefore, in this research, an environmentally friendly film was prepared from natural materials with antimicrobial capabilities, and its color, mechanical and thermal properties were investigated. The analysis of the L factor showed that the pure mucilage/alginate film does not have a very high transparency and shows a transparency of about 55, which decreased with the addition of tungsten oxide nanoparticles and norbixin pigment, so that the mucilage/alginate film containing both tungsten oxide nanoparticles and norbixin Haste had the least transparency. Examining the factor a (green-red) showed that this factor increased with the increase of Norbixin and tungsten oxide nanoparticles, that is, it moved towards more positive numbers that indicate redness. The effect of Norbixin in increasing factor a was more than that of tungsten oxide nanoparticles. According to the nature of the norbixin pigment, which has a yellow-orange color, an increase in factor a is expected due to the presence of norbixin. Tungsten oxide nanoparticles also have a somewhat yellow color, and these particles also increase the a factor. Examining factor b (blue-yellow) showed that with the increase of Norbixin and tungsten oxide nanoparticles, this factor increased, that is, it moved towards more positive numbers that indicate yellowness. By examining the spectrum of the pure mucilage/alginate film, it was found that this film shows two broad peaks at 2 theta of 10 and 20 degrees, which indicates the relatively amorphous structure of this film. The research conducted on the structure of pure sodium alginate film has introduced this film as a semi-crystalline film due to the presence of a peak in

the region of 10 degrees, which is also seen in this study at 2 theta of 10 degrees, but since Previous studies have confirmed the amorphous structure of flax seed mucilage, and considering that alginate and mucilage were used to prepare the film, the prepared film had a semi-crystalline structure with a tendency to form an amorphous structure. By examining the TGA curves of the films, it is clear that the weight loss of the second stage in the modified films with tungsten oxide nanoparticles and norbixin occurred at a higher temperature than the pure mucilage/alginate film, and also in the modified films, the weight loss in the second stage The second has been less compared to pure film. These results show that tungsten oxide nanoparticles and norbixin have increased the thermal stability of the film. The mechanical results of the films showed that the pure mucilage/alginate film has good mechanical resistance. This film has up to 65% tensile strength and its tensile strength is 55 MPa, which is suitable for food packaging. Examining the antibacterial properties of 4 types of films shows that all films (even the pure mucilage/alginate film) have antimicrobial activity against Gram-positive and Gram-negative bacteria. The addition of tungsten oxide nanoparticles and norbixin pigment increased the antibacterial property of the film significantly ( $p < 0.05$ ). The highest antibacterial property was related to mucilage/alginate film modified with tungsten oxide nanoparticles and norbixin.

## 5- Resources

- [1] Jabraili, A., Pirs, S., Pirouzifard, M.K. and Amiri, S., 2021. Biodegradable nanocomposite film based on gluten/silica/calcium chloride: physicochemical properties and bioactive compounds extraction capacity. *Journal of Polymers and the Environment*, 29(8), pp.2557-2571.
- [2] Hosseini, S.N., Pirs, S. and Farzi, J., 2021. Biodegradable nano composite film based on modified starch-albumin/MgO; antibacterial,

- antioxidant and structural properties. *Polymer Testing*, 97, p.107182.
- [3] Pirsá, S. and Asadi, S., 2021. Innovative smart and biodegradable packaging for margarine based on a nano composite polylactic acid/lycopene film. *Food Additives & Contaminants: Part A*, 38(5), pp.856-869.
- [4] Acik, G., 2020. Preparation of antimicrobial and biodegradable hybrid soybean oil and poly (L-lactide) based polymer with quaternized ammonium salt. *Polymer Degradation and Stability*, 181, p.109317.
- [5] Jiménez-Rosado, M., Perez-Puyana, V., Rubio-Valle, J.F., Guerrero, A. and Romero, A., 2020. Processing of biodegradable and multifunctional protein-based polymer materials for the potential controlled release of zinc and water in horticulture. *Journal of Applied Polymer Science*, 137(46), p.49419.
- [6] Pirsá, S. and AghbolaghSharifi, K., 2020. A review of the applications of bioproteins in the preparation of biodegradable films and polymers. *Journal of Chemistry Letters*, 1(2), pp.47-58.
- [7] Sani, I.K., Geshlaghi, S.P., Pirsá, S. and Asdagh, A., 2021. Composite film based on potato starch/apple peel pectin/ZrO<sub>2</sub> nanoparticles/microencapsulated *Zataria multiflora* essential oil; investigation of physicochemical properties and use in quail meat packaging. *Food Hydrocolloids*, 117, p.106719.
- [8] Asdagh, A., Karimi Sani, I., Pirsá, S., Amiri, S., Shariatifar, N., Eghbaljoo-Gharehgheshlaghi, H., Shabahang, Z. and Taniyan, A., 2021. Production and characterization of nanocomposite film based on whey protein isolated/copper oxide nanoparticles containing coconut essential oil and paprika extract. *Journal of Polymers and the Environment*, 29(1), pp.335-349.
- [9] Sharifi, K.A. and Pirsá, S., 2021. Biodegradable film of black mulberry pulp pectin/chlorophyll of black mulberry leaf encapsulated with carboxymethylcellulose/silica nanoparticles: Investigation of physicochemical and antimicrobial properties. *Materials Chemistry and Physics*, 267, p.124580.
- [10] Fabre, J.F., Lacroux, E., Valentin, R. and Mouloungui, Z., 2015. Ultrasonication as a highly efficient method of flaxseed mucilage extraction. *Industrial Crops and Products*, 65, pp.354-360.
- [11] Roulard, R., Petit, E., Mesnard, F. and Rhazi, L., 2016. Molecular investigations of flaxseed mucilage polysaccharides. *International journal of biological macromolecules*, 86, pp.840-847.
- [12] Pirsá, S., Dalili, R. and Yazdani, I., 2018. Effects of Quince Seed Mucilage and Guar Gum on the Physicochemical and Sensory Properties of Doogh. *Journal of Agricultural Science and Technology*, 20(3), pp.485-494.
- [13] Daei, S., Mohtarami, F. and Pirsá, S., 2022. A biodegradable film based on carrageenan gum/*Plantago psyllium* mucilage/red beet extract: physicochemical properties, biodegradability and water absorption kinetic. *Polymer Bulletin*, pp.1-22.
- [14] Ye, J., Ma, D., Qin, W. and Liu, Y., 2018. Physical and antibacterial properties of sodium alginate—sodium carboxymethylcellulose films containing *Lactococcus lactis*. *Molecules*, 23(10), p.2645.
- [15] Hecht, H. and Srebnik, S., 2016. Structural characterization of sodium alginate and calcium alginate. *Biomacromolecules*, 17(6), pp.2160-2167.
- [16] Shabkhiz, M.A., Pirouzifard, M.K., Pirsá, S. and Mahdavinia, G.R., 2021. Alginate hydrogel beads containing *Thymus daenensis* essential oils/Glycyrrhizic acid loaded in  $\beta$ -cyclodextrin. Investigation of structural, antioxidant/antimicrobial properties and release assessment. *Journal of Molecular Liquids*, 344, p.117738.
- [17] Scotter, M., 2009. The chemistry and analysis of annatto food colouring: a review. *Food Additives and Contaminants*, 26(8), pp.1123-1145.
- [18] Mahendranath, G., Venugopalan, A., Parimalan, R., Giridhar, P. and Ravishankar, G.A. *Plant Cell, Tissue and Organ Culture (PCTOC)*, 106(3), pp.517-522.
- [19] Duan, G., Chen, L., Jing, Z., De Luna, P., Wen, L., Zhang, L., Zhao, L., Xu, J., Li, Z., Yang, Z. and Zhou, R., 2019. Robust antibacterial activity of tungsten oxide (WO<sub>3</sub>-X) nanodots. *Chemical research in toxicology*, 32(7), pp.1357-1366.
- [20] Wang, Q., Wang, H., Zhang, T., Hu, Z., Xia, L., Li, L., Chen, J. and Jiang, S., 2021.



- Antibacterial Activity of Polyvinyl Alcohol/WO<sub>3</sub> Films Assisted by Near-Infrared Light and Its Application in Freshness Monitoring. *Journal of Agricultural and Food Chemistry*, 69(3), pp.1068-1078.
- [21] Priyadarshi, R., Kim, H.J. and Rhim, J.W., 2021. Effect of sulfur nanoparticles on properties of alginate-based films for active food packaging applications. *Food Hydrocolloids*, 110, p.106155.
- [22] Prado, N.S., Silva, I.S.V.D., Silva, T.A.L., Oliveira, W.J.D., Motta, L.A.D.C., Pasquini, D. and Otaguro, H., 2018. Nanocomposite films based on flaxseed gum and cellulose nanocrystals. *Materials Research*, 21.
- [23] Bhagyaraj, S. and Krupa, I., 2020. Alginate-mediated synthesis of hetero-shaped silver nanoparticles and their hydrogen peroxide sensing ability. *Molecules*, 25(3), p.435.
- [24] Perotti, G.F., Silva, F.F., de Couto, R.A., Lima, F.C., Petrilli, H.M., Leroux, F., Ferreira, A. and Constantino, V.R., 2020. Intercalation of Apocarotenoids from Annatto (*Bixaorellana L.*) into Layered Double Hydroxides. *Journal of the Brazilian Chemical Society*, 31, pp.2211-2223.
- [25] Pang, H.F., Xiang, X., Li, Z.J., Fu, Y.Q. and Zu, X.T., 2012. Hydrothermal synthesis and optical properties of hexagonal tungsten oxide nanocrystals assisted by ammonium tartrate. *physica status solidi (a)*, 209(3), pp.537-544.
- [26] Salisu, A., Sanagi, M.M., Abu Naim, A., Abd Karim, K.J., Wan Ibrahim, W.A. and Abdulganiyu, U., 2016. Alginate graft polyacrylonitrile beads for the removal of lead from aqueous solutions. *Polymer Bulletin*, 73(2), pp.519-537.
- [27] Teoh, L.G., Shieh, J., Lau, W.H. and Hon, M.H., 2004. Effects of mesoporous structure on grain growth of nanostructured tungsten oxide. *Journal of materials research*, 19(9), pp.2687-2693.
- [28] Pereira, R., Tojeira, A., Vaz, D.C., Mendes, A. and Bártolo, P., 2011. Preparation and characterization of films based on alginate and aloe vera. *International Journal of Polymer Analysis and Characterization*, 16(7), pp.449-464.
- [29] Handayani, I., Haryanti, P. and Sulisty, S.B., 2021. Color and antibacterial activity of annatto extracts at various pH of distilled water solvent and extraction temperature. *Food Research*, 5(6), pp.247-253.
- [30] Viuda-Martos, M., Ciro-Gómez, G.L., Ruiz-Navajas, Y., Zapata-Montoya, J. E., Sendra, E., Pérez-Álvarez, J. A. and Fernández-López, J., 2012. In vitro Antioxidant and Antibacterial Activities of Extracts from Annatto (*Bixaorellana L.*) Leaves and Seeds. *Journal of Food Safety*, 32(4), pp.399-406.



فیلم زیست تخریب پذیر آلزینات سدیم/موسیلایز دانه کتان/نوریکسین/نانوذرات اکسید تنگستن: بررسی

خواص رنگی، کریستالی، حرارتی، مکانیکی و ضدباکتریایی

حسین دادخواه<sup>۱\*</sup>، سجاد پیرسا<sup>۲</sup>، افشین جوادی<sup>۳</sup>، فروغ محترمی<sup>۲</sup>

۱- گروه علوم و صنایع غذایی، واحد ممقان، دانشگاه آزاد اسلامی، ممقان، ایران.

۲- گروه علوم و صنایع غذایی، دانشگاه ارومیه، ارومیه، ایران.

۳- گروه بهداشت مواد غذایی، دانشکده دامپزشکی، علوم پزشکی تبریز، دانشگاه آزاد اسلامی، تبریز، ایران.

## اطلاعات مقاله

## چکیده

### تاریخ های مقاله :

تاریخ دریافت: ۱۴۰۱/۱۰/۲۱

تاریخ پذیرش: ۱۴۰۱/۱۲/۲۰

### کلمات کلیدی:

فیلم زیست تخریب پذیر،

مقاومت حرارتی،

مقاومت مکانیکی،

پلیمرهای ضدباکتریایی و بسته بندی هوشمند.

DOI: 10.22034/FSCT.19.135.31

DOR: 20.1001.1.20088787.1402.20.135.4.0

\* مسئول مکاتبات:

H.d2960050241@gmail.com

در این تحقیق موسیلایز دانه کتان استخراج شده و فیلم مرکب آلزینات سدیم و موسیلایز دانه کتان تهیه شد. از رنگدانه نوریکسین و نانوذرات اکسید تنگستن ( $WO_3$ ) برای اصلاح ساختار فیلم استفاده شد. خواص رنگی، کریستالی، حرارتی و مکانیکی فیلم ها بررسی شد. همچنین خاصیت ضد باکتریایی فیلم های تهیه شده نسبت به باکتری اشریشیاکلی و استافیلوکوکوس اورئوس بررسی شد. نتایج بدست آمده نشان داد که با افزودن نانوذرات اکسید تنگستن و رنگدانه نوریکسین شفافیت کاهش یافته است. تاثیر نانوذرات تنگستن در کاهش شفافیت فیلم بیشتر از تاثیر نوریکسین بوده است. بررسی فاکتور a نشان داد که با افزایش نوریکسین و نانوذرات اکسید تنگستن این فاکتور افزایش یافت. بررسی فاکتور b نشان داد که با افزایش نوریکسین و نانوذرات اکسید تنگستن این فاکتور افزایش یافت. با بررسی طیف XRD فیلم خالص موسیلایز/آلزینات مشخص شد که این فیلم دو پیک پهنا ۲ تا ۱۰ و ۲۰ درجه نشان می دهد که نشان دهنده ساختار نسبتا آمورف این فیلم می باشد. در فیلم موسیلایز/آلزینات اصلاح شده با نانوذرات اکسید تنگستن پیک های مربوط به نانوذرات کریستالی در ۲ تا ۲۵ تقریبی، ۳۰، ۳۵، ۴۰، ۵۰، ۵۵ و ۶۵ درجه کاملا مشخص هست که نشان می دهد این نانوذرات ساختار کریستالی فیلم را بهبود بخشیده است. با بررسی منحنی های TGA فیلم ها مشخص شد که نانوذرات اکسید تنگستن و نوریکسین پایداری حرارتی فیلم را افزایش داده است. بررسی خاصیت ضدباکتریایی فیلم ها نشان داد که افزودن نانوذرات اکسید تنگستن و رنگدانه نوریکسین خاصیت ضدباکتریایی فیلم را به طور معنی داری ( $p < 0.05$ ) افزایش داده است.