Films from an Aqueous Suspension of Alkaline Pretreated and Fine Milled Chicken Feathers

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Abstract

The purpose was to test the feasibility of preparing cast films directly from an aqueous suspension of alkaline pretreated and fine milled chicken feathers, and to evaluate the impact of different additives on film formation and the tensile properties of the resulting films. The feather suspension consisted of stiff and sharp-pointed fibers together with more round-shaped fines. Films cast from this suspension were opaque and porous. While films without additives were fragile with drying-induced defects, film formation was improved with additives, especially with ethanolamine and maleic acid at 20% and 30% concentrations. A synergistic plasticizing effect was observed with ethanolamine and formamide, and strength of the films was improved with sodium alginate. However, the overall impact of additives on the tensile properties in general and strain at break in specific was limited. This was likely due to the dominating role of the porous film structure and the stiff fibers with a limited reactivity towards the additives.

Keywords

Feather, Films, Mechanical Properties

1. Introduction

The target of sustainable economy is to utilize biomass-derived materials for high-volume applications [1]. There is interest, for example, in packaging industry towards more sustainable materials, and biobased plastics and biopolymers have been widely studied. Interesting sources of biomass are by-products from animal sources (legs, heads, bones and feather) currently used for fertilizers, animal feed and pet food.

The global production of broiler meat was estimated to reach 98.4 million tons in 2019 [2]. Feathers constitute up to 10% out of chicken body weight [3].
This equals 23.4 million tons of feather waste annually. Although chicken feathers and keratin, the main component of feathers, have been tested for various applications, there is still a limited use of feathers as a raw material in industrial applications mainly due to the lack of technologies for processing keratin and established routes for utilizing keratin-based products. Most of feather waste is disposed of in landfills, incinerated, or converted into low-value products. Disposal by landfilling or incinerating is, however, becoming increasingly challenging [4].

Chicken feathers vary in form. In general, they are half fibrous material and half central core with a hollow tube structure [5]. Feather components are hydrophobic by nature resulting in poor wettability compared to cotton and wood pulp fibers [6]. Chemically feathers consist of approximately 90% protein, 8% water, and 1% fat [7]. The amino acids are tightly-packed and crosslinked making extraction of keratin challenging [5] [8]. However, keratin can be extracted by chemical and enzymatic means, superheated water, and with ionic liquids [9] [10]. This adds to the complexity of the feather processing.

Processing of keratin into plastic products has been studied extensively [9] [11] [12] [13] [14]. Feathers, on the other hand, have been tested for fabrics, construction materials, composites, and environmental applications [5] [15] [16] [17] [18]. Feathers are not thermoplastic, and chemically modified and hydrolyzed thermoplastic feathers have therefore been prepared and blended with glycerol for making films by compression molding [18] [19] [20] [21]. Chemical modification adds costs and potentially impairs the biodegradability of the feathers [18].

Feather, keratin, and other biopolymer films require additives, such as plasticizers and crosslinking agents, to adjust mechanical properties and moisture stability. Besides glycerol [18] [22] also other plasticizers have been tested for biopolymer films, such as 1,4-butanediol [23], ethanolamine [24] [25] [26], maleic and citric acids [24], sorbitol [27], and urea [26] [28]. Formamide has been used as a softener for paper, gums, and glues, and to prevent cracking in sol-gel coatings [29] [30]. It has also been used as a plasticizer together with urea for thermoplastic biopolymers [31] [32]. Typical crosslinking agents include citric acid [18] [33] and glyoxal [34] [35]. Sodium alginate has been blended with keratin and gelatin, and it crosslinks with different metal ions, especially with calcium [36] [37].

The purpose of this study was to test the feasibility of preparing cast films from an aqueous suspension of alkaline pretreated and milled chicken feathers, and to evaluate the impact of additives on the film formation and the tensile properties of the resulting films.

2. Materials and Methods

This study was carried out at VTT between January and August 2019.

2.1. Materials

Sanitized chicken feathers were supplied by Grupo Sada (Madrid, Spain).
Additives tested for the feather suspension included 1,4-butanediol (Sigma-Aldrich), calcium chloride (CaCl₂; Merck), citric acid (Sigma-Aldrich), D-sorbitol (Sigma-Aldrich), ethanolamine (Fluka), ferric perchlorate (Fe(ClO₄)₃∙9H₂O; Fluka), formamide (Sigma-Aldrich), glycerol (Sigma-Aldrich), glyoxal (BASF), magnesium chloride hexahydrate (MgCl₂·6H₂O; VWR Chemicals), maleic acid (Sigma-Aldrich), sodium alginate (Sigma-Aldrich), sodium hypophosphite monohydrate (NaPO₂H₂·H₂O; Alfa Aesar), and urea (Sigma-Aldrich). Additives in solid form were dissolved into water (4% consistency) except sorbitol that was used at 50% consistency.

2.2. Methods

2.2.1. Preparing Feather Suspension

Feathers were refined using a combination of mechanical and chemical treatments. Images of original and refined feathers are presented in Figure 1. First, a mild alkali pre-treatment was performed for the feather material. Alkaline treatment was applied with NaOH (2 mol/kg dry feather) and liquid/feather ratio of 5:1 (W/W) at 50°C for 40 min in order to avoid dissolving. The feather pulp was then washed with deionised water and dried in oven at 105°C. After the alkaline pre-treatment the feathers were processed using a compactor in which the feathers were pressed through a die using pan grinder rollers and crushed to an approximately 1 - 3 cm length to enable better feeding into a grinder.

Pre-treated feathers were soaked in deionised water at 10% consistency for approximately one hour and dispersed using a ladle. The suspension was then fed into a Masuko Sangyo’s Supermasscolloider (Masuko Sangyo Co.) type MKZA10-15J. Three passes were ground with a standard stone type MKE10-46 and three following passes with a modified stone type MKGA10-80. The feather suspension passed six times through the grinder and with increasing operating power from 2.0 to 2.4 kW. The gap width was gradually decreased from 0.3 mm to 0.2 mm. Operating speed was fixed at 1500 rpm. Finally, the ground feather suspension passed four times through the Microfluidizer type M110-EH. The suspension was diluted with deionised water to 6% consistency before the microfluidization. Two passes were processed through the chambers having diameters of 400 µm and 200 µm at 1100 bar. Three following passes were processed.
through the chambers having diameters of 400 µm and 100 µm at 1800 bar operating pressure.

The feather suspension was characterized for fiber type and length with an optical microscope (Nikon ECLIPSE Ci) and FiberLab™ analyzer (Metso Automation), and for consistency, pH and conductivity.

2.2.2. Formulations
The feather suspension was diluted with tap water to 4% consistency to lower viscosity.

The diluted suspension was mixed carefully for 20 min at room temperature with a mechanical overhead laboratory stirrer (IKA RW 20 digital). The suspension was placed in a hot water bath at 50°C and the plasticizer was added to the suspension. Mixing was continued for at least 10 min prior adding other possible additives, and the final formulation was mixed for a further 20 min at 50°C. Tested formulations are presented in Tables 1-3.

Table 1. Formulations (weight %) with a single plasticizer.

| Formulation | 1  | 2  | 3  | 4  | 5  | 6  | 7  | 8  | 9  |
|-------------|----|----|----|----|----|----|----|----|----|
| Feathers    | 80 | 80 | 80 | 80 | 80 | 80 | 70 | 70 | 70 |
| Plasticizers|    |    |    |    |    |    |    |    |    |
| 1,4-butanediol | 20 |    |    |    |    |    |    |    |    |
| Ethanolamine | 20 |    |    |    |    |    |    |    |    |
| Glycerol    |    |    |    |    |    |    | 20 |    |    |
| Maleic acid |    |    |    |    | 20 |    |    |    | 30 |
| Sorbitol    |    |    |    |    |    |    |    | 20 |    |
| Urea        |    |    |    |    |    |    |    |    | 20 |

Table 2. Formulations (weight %) with ethanolamine and other additives.

| Formulation | 10 | 11 | 12 | 13 | 14 | 15 | 16 | 17 | 18 |
|-------------|----|----|----|----|----|----|----|----|----|
| Feathers    | 75 | 70 | 65 | 60 | 62.5| 55 | 65 | 64.75| 65 |
| Plasticizers|    |    |    |    |    |    |    |    |    |
| Ethanolamine| 20 | 20 | 30 | 30 | 30 | 30 | 30 | 30 | 30 |
| Other additives | Citric acid | 5 | 10 |    |    |    |    |    |    |
|              | Formamide | 5 | 10 | 5 | 10 |    |    |    |    |
|              | Sodium alginate | 5 | 5 |    |    |    |    |    |    |
|              | Glyoxal |    |    |    |    |    |    | 5 |    |
|              | CaCl2 |    |    |    |    |    |    |    | 0.25|
|              | NaPO₄H₂H₂O | 2.5 | 5 |    |    |    |    |    |    |
Table 3. Formulations (weight %) with ethanolamine or maleic acid and other additives.

| Formulation | 19 | 20 | 21 | 22 | 23 | 24 | 25 | 26 | 27 | 28 |
|-------------|----|----|----|----|----|----|----|----|----|----|
| Feathers    | 65 | 60 | 62.5| 64.9| 64.75| 62.5| 62.5| 60 | 57.5| 62.5|
| Plasticizers| Ethanolamine | 30 | 30 | 30 | 30 | 30 | 30 |    |    |    |
|             | Maleic acid | 30 | 30 | 30 |    |    |    |    |    |
| Other additives | Citric acid | 5  |    |    |    |    |    |    |    |
|              | Formamide   | 5  | 10 |    |    |    |    |    |    |
|              | Sodium alginate | 5 | 5 | 5 | 5 | 10 | 10 | 10 |    |    |
|              | CaCl₂       | 0.1|    |    |    |    |    |    |    |
|              | Fe(ClO₄)₃·9H₂O | 2.5|    |    |    |    |    |    |    |
|              | MgCl₂·6H₂O  | 0.25| 2.5| 2.5 | 5  |    |    |    |    |
|              | NaPO₂H₂·H₂O | 2.5|    |    |    |    |    |    |    |

2.2.3. Film Casting

30 - 40 ml of each formulation was poured onto three plastic Petri dishes with a diameter of 140 mm (VWR International). To prevent films from adhering to the Petri dish during drying, a sheet of a one side siliconized 30 µm polyester film provided by UPM Raflatac (Tampere, Finland) was first placed at the bottom of the dish. Occasional air bubbles were removed with a single purpose pipette from the suspension. The films were dried in an air circulating oven at 50˚C at least for overnight. Films from formulations 14 and 15 were also heat treated with an L&W Rapid Dryer (ABB AB, Lorentzen & Wettre) at 150˚C for 5 min.

2.2.4. Film Characterization

The films were evaluated visually, and one film of each formulation was photographed.

Cross-sectional surface was prepared from selected films by fracturing a strip of material in liquid nitrogen. The strip was mounted on an aluminum stub with conductive carbon adhesive tape and sputter-coated with approx. 4 nm of gold-palladium. SEM images (Field Emission Scanning Electron Microscope, Zeiss Merlin) were taken from a tilted (50˚) stub to show simultaneously the material surface and the fractured cross-section. The acceleration voltage was 2 kV and probe current 60 pA.

Tensile properties of the films were measured using Lloyd LS5 materials testing machine. Load cell was 100 N, crosshead speed 2 mm/min, and initial gauge length 20 mm. In total 3 - 4 strips 15 mm wide of each sample were tested at 23˚C and 50% RH. Film thickness was measured twice from each strip tested. Properties of interest were tensile strength (MPa), strain at break (%), and
Young’s modulus (MPa). Before testing, all specimens were conditioned at 23°C and 50% RH for several days.

3. Results

3.1. Feather Suspension

The feather suspension was a viscous fluid with a brownish color. Consistency of the suspension was 5.68%, pH 8.9, and conductivity 453 µS/cm. Based on the microscope images most of the fibrous material was equal or shorter than 100 µm, but there were also significantly longer fibers, as presented in Figure 2. The fibers were stiff and sharp-pointed, but there were also round-shaped fines. The FiberLab™ analyzer detected fibers with length around 100 µm (number average 80 µm and weight average 110 µm). However, long fibers were excluded from this analysis, although they were obviously present.

3.2. Film Formation

Films were prepared successfully from all the formulations, except for formulation 17. Adding 0.25% calcium chloride to a mixture of feathers, ethanolamine, and sodium alginate had a detrimental impact on colloidal stability, and deposits were formed during mixing. Therefore, this formulation was omitted.

Based on visual evaluation of the films prepared from formulations containing feathers and a single plasticizer (Table 1), ethanolamine and maleic acid produced the most uniform films. Other films had drying-induced patterning, but even these were intact and more uniform than the films prepared without any additives. Figure 3 presents films without additives and films from formulations 1 - 6. The films without additives were fragile and difficult to handle.

Figure 4 shows examples of films from formulations with two or three additives. Formulations with either ethanolamine or maleic acid together with formamide
Figure 3. Film without additives and films from formulations 1 - 6 with single plasticizers.

(such as 12 and 19, respectively) resulted in uniform films as did ethanolamine together with sodium alginate (formulation 16) and citric acid, although adding both ethanolamine and formamide resulted in a slight discoloration. Films with both maleic and citric acids with sodium hypophosphite monohydrate (formulation 21) indicated some patterning formed during drying. Most of the films with ethanolamine, sodium alginate, and metal salt (e.g. formulation 23) demonstrated drying-induced shrinkage and slight unevenness. Visually films from
formulation 18 with glyoxal were the most striking with a strong orange color due to an exothermic reaction between ethanolamine and glyoxal.

3.3. Film Structure

All the films were opaque caused by the light scattering in a porous film structure. SEM images prepared from films without additives and from formulations 8, 12, and 16 support this (Figure 5). These images show a structure consisting of randomly oriented and relatively stiff and flat fibers covered with a network of fine fibrillated and non-fibrous material on the side in contact with the PET film. No significant differences were observed between the film surfaces or in the crosscuts. Some of the additives must have concentrated within the structure and on the top side of the films.

3.4. Mechanical Properties

Films without any additives were too fragile for mechanical testing. This was in
Figure 5. Surface and crosscut SEM images (magnification 500×) of films without additives and films with 30% ethanolamine (formulation 8), 30% ethanolamine and 5% formamide (formulation 12), and 30% ethanolamine and 5% sodium alginate (formulations 16).

In line with previous studies indicating that keratin films even with a small amount of plasticizer (0.05 and 0.1 g of glycerol/g of keratin) were too brittle for testing [38]. In this study, all the films with plasticizers could be measured for the tensile properties. However, bonding between the fibers may dominate the mechanical properties of such porous films.

Figure 6 shows the tensile properties for the films with a single plasticizer. These films were stronger and more brittle than those prepared previously from thermoplastic feathers and keratin (tensile strength 5.2 - 5.9 MPa [18], and strength 0.3 MPa and Young’s modulus 25 MPa [9]). The mechanical properties, except of strain at break, were similar to the carboxymethylated keratin films by Schrooyen et al. [38], but weaker than the grafted feather films without and with 30% glycerol (206.3 and 55.7 MPa, respectively) [19]. With 20% plasticizer the strongest films were obtained with sorbitol, 1,4-butandiol and ethanolamine, while the films containing maleic acid, urea, and glycerol were among the weakest. Studies with keratin films have shown that increased glycerol concentration decreases the tensile strength and the elastic modulus, while the strain at break increases [9] [38]. Increasing concentration of 1,4-butandiol decreased strength in this study. However, little effect was observed with ethanolamine, and strength increased with an increasing concentration of maleic acid. Maleic acid must have promoted bonding at the fiber-fiber contacts. Maleic acid is known to improve adhesion in polymer-fiber composites [39].

Increasing the amount of maleic acid increased also the strain at break. 1,4-butandiol, glycerol, sorbitol, and urea yielded similar results, while ethanolamine provided slightly lower strain at break, albeit the differences were small.
Figure 6. Tensile strength, strain at break, and Young’s modulus for films with a single plasticizer.

In general, the level of maximum strain was low. 20% urea, glycerol and maleic acid, and 30% 1,4-butanediol resulted in a low elastic modulus. The highest stiffness was obtained with ethanolamine and 30% maleic acid.

Results from formulations with a blend of ethanolamine and maleic acid together with formamide are presented in Figure 7. At 30% concentration of ethanolamine and maleic acid the impact of formamide was logical. Tensile strength decreased and strain at break increased. Elastic modulus decreased indicating
increased elasticity at small deformations. At 20% concentration of ethanolamine more formamide was needed to obtain a plasticizing effect. Formamide together with other plasticizers can provide formulations with good plasticizing effect and improved stability of the plasticizing system. According to Yan et al. formamide can act as a solvent and prevent phase separation [32]. Such a synergistic effect was observed when the properties of films with 20% ethanolamine and 10% formamide were compared to those with only 30% ethanolamine.

**Figure 7** shows the impact of other additives on the tensile properties. Together
Figure 8. Tensile strength, strain at break, and Young’s modulus for films with ethanolamine and maleic acid together with citric acid and NaPO$_4$H$_2$H$_2$O, glyoxal, and sodium alginate.
with 30% concentration of ethanolamine both citric acid and glyoxal decreased all the tensile properties. Additional heat treatment of films with ethanolamine and citric acid had only a minor impact (formulations 14 and 15). According to a previous study with thermoplastic feather films already a 5% concentration of citric acid can lead to overcrosslinking indicated by a decrease in all the three tensile properties [18]. Citric acid together with 30% maleic acid, on the other hand, increased strain at break, which indicates more of a plasticizing effect (formulation 21). Glyoxal reacted readily with ethanolamine, but this did not increase strength (formulation 18). Using ethanolamine and sodium alginate together (formulations 17 and 26) improved the tensile properties. However, even in this case, the strain at break was less than 6%.

The impact of crosslinking sodium alginate in the formulations with metal ions was tested. Besides a low amount of CaCl₂, also other metal cations (Fe³⁺ and Mg²⁺) were tested. As presented in Figure 9, CaCl₂ increased both tensile strength and elastic modulus. However, strain at break was again less than 6%. A similar impact was reached with a higher concentration of MgCl₂·6H₂O together with 5% sodium alginate. However, with 10% sodium alginate doubling the amount of MgCl₂·6H₂O actually decreased tensile strength and elastic modulus, which is likely due to a negative impact of a high addition level of salt on the overall stability of the feather suspension. CaCl₂ decreased the strain at break. These results are in-line with the previous results indicating that calcium ions crosslink sodium alginate more efficiently than, for example, magnesium ions [40].

4. Conclusions

An aqueous suspension was prepared from chicken feathers using an alkaline pretreatment followed by a three-stage mechanical milling process consisting of crushing, grinding, and microfluidization. The purpose was to avoid costs and complexity related to extraction of keratin or chemical modification of feathers. The resulting suspension consisted of stiff and sharp-pointed fibers together with a smaller number of more round-shaped fines.

Films were cast from the feather suspension. Films without additives were of poor quality and fragile. Film formation was improved with additives, and visually the best films were obtained with 20% - 30% of ethanolamine and maleic acid as the single additive or together with formamide, sodium alginate, and citric acid. The films were opaque and porous, which would limit their feasibility for applications where transparency and good barrier properties are essential.

The strongest films were obtained with maleic acid, sorbitol, and ethanolamine as the single additive. Adding formamide together with ethanolamine and maleic acid resulted in a plasticizing effect. Sodium alginate as a secondary additive with ethanolamine had a positive impact on tensile strength with and without cationic metal ions used to crosslink sodium alginate. However, the modest impact of additives on the tensile properties, especially strain at break, can be
explained by the dominating role of the porous film structure with stiff fibers and low reactivity of the fiber surfaces towards the additives.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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