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Effect of biofibers addition on the structure and properties of soy protein composite films

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Abstract

Soy protein isolate (SPI) has garnered researchers' attention due to its abundance, cost-effectiveness, excellent biocompatibility, hemo-compatibility, and biodegradability. However, SPI faces limitations in application due to poor processability and weak mechanical strength. Substantial efforts have been made to address these challenges. In this preliminary study, glycerol and biofibers were added to SPI to improve the mechanical properties and film forming, and glyoxal was employed to crosslink SPI molecules. The microstructure and mechanical properties of the resulting SPI/composite films were evaluated. A 15% addition of glycerol proved sufficient for good film formation. Among the biofibers, short SF microfibers were the most effective in enhancing breaking strength, while TEMPO-oxidized CNF (cellulose nanofiber) excelled among CNFs. Crosslinking with glyoxal significantly enhanced the mechanical properties, with the type of biofiber minimally affecting the mechanical properties of the crosslinked SPI composite films.

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Introduction

The by-products of the soy oil industry, soy protein, are available in three forms: defatted soy flour/flakes, soy protein concentrate (SPC), and soy protein isolate (SPI). Among these, soy protein isolate (SPI) stands out with a high protein content (85-90%) due to the removal of oil, carbohydrates, insoluble carbohydrates, and dietary fibers (Preece *et al*., 2017). SPI, characterized by abundance, cost-effectiveness, excellent biocompatibility, hemo-compatibility, and biodegradability (Wang *et al*., 2021; Zhao *et al*., 2018), has been extensively researched in various forms such as emulsion, fiber, film, hydrogel, and sponge, not only in the food industry but also in cosmetics and medical fields.

Despite its advantageous properties, soy protein faces limitations in applications due to poor processability and weak mechanical strength resulting from weak interactions between hydrophilic groups in the soy protein chains. Therefore, to improve the film formability and flexibility as well as mechanical properties of SPI, glycerol and CNF have been used, respectively (González *et al*., 2019; Qin *et al*., 2019).

However, in case of glycerol, too high amount has been used [>25% on the weight (o.w.f) of SPI] to deteriorate pure SPI's properties (González *et al*., 2019; Qin *et al*., 2019). In case of CNF, although there were many kinds of CNFs, a comparative study on the effect of type of CNF on SPI has not been conducted (Martelli-Tosi *et al*., 2018; Qin *et al*., 2019; Wang *et al*., 2022).

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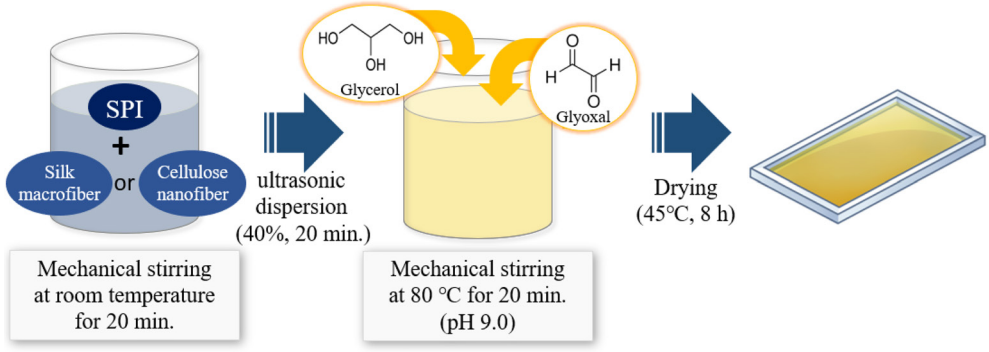

Scheme 1. Illustration of the preparation process of the SPI composite films.

In a meanwhile, a short silk fiber can be prepared relatively easily than silk nanofiber because it can be obtained by grinding degummed silk fiber with optimized condition. However, it has not been used to improve the mechanical properties of SPI.

Therefore, in this study, as a preliminary study on the improvement of mechanical properties of SPI composite film, low amount of glycerol (15% or less) was added to SPI. Also, three kinds of CNF (TEMPO–oxidized, Mechanical grinded, and Enzymatic hydrolyzed) and short SF microfiber were added. Finally, glyoxal was used to crosslink SPI molecule to enhance mechanical properties of SPI. Finally, mechanical properties and microstructure of SPI composite films were examined.

Materials and Methods

Materials

Soy protein isolate powder was purchased from Korea Beauty & Healthcare Co., Ltd (Natural Box Soy Protein Isolate Powder, Seoul, Republic of Korea). Sodium hydroxide and glycerol were purchased from Duksan Pure Chemicals Co., Ltd (Ansan, Republic of Korea). Glyoxal was purchased from Daejung Chemicals & Metals Co., Ltd (Siheung, Republic of Korea). Three types of CNF (TEMPO–oxidized, Mechanical grinded, and Enzymatic hydrolyzed) were provided by the Korea Textile Machinery Convergence Research Institute (Gyeongsan, Republic of Korea). *Bombyx mori* Baekokjam silkworm cocoons were supplied by the National Institute of Agricultural Science (Wanju, Republic of Korea). The silkworm cocoons were dried for 4 hours at a high temperature (90 °C) to kill the pupa.

Preparation of the SF microfiber

Non-protein components of silkworm cocoons were removed using the reported method (Kim and Um, 2014). The silkworm cocoons were degummed using purified water at 120 °C for 10 minutes in an autoclave (JSAT-65, JSR, Gongju, Republic of Korea). The cocoon-to-water ratio was 1:50 (w/v). Subsequently, the degummed SF fibers (fibroin) were rapidly frozen by immersion in liquid nitrogen and ground using a universal cutting mill (PULVERISETTE 19, FRITSCH, Germany) at the Korea Institute of Industrial Technology (Ansan, Republic of Korea). Finally, the short SF microfibers were obtained by filtering with a 200 μm mesh. Finally, the short SF microfibers were obtained by filtering with a 200-μm mesh.

Preparation of soy protein composite films with nano- and micro-scale biofibers

Scheme 1 illustrated the preparation process of the SPI composite films. The 5 wt% SPI suspension was combined with the three types of CNF or SF (0, 3, 5, 7, and 10% on the weight of SPI), followed by magnetic stirring for 20 min. All mixtures were homogenized in an Ultrasonic Processor (VC 750, Sonics & Materials, Inc., Newtown, USA) for 20 min. Next, glycerol (0, 5, 10, 15, and 25% on the weight (o.w.f) of SPI) was added as a plasticizer, and 5% glyoxal on the weight of SPI was added as a cross-linker. The pH was adjusted to 9.0±0.2 using a 10% (w/v) NaOH solution, and the mixtures were agitated at 80°C for 20 min. A 10 ml SPI mixed solution was poured into a 40-mm × 95-mm silicon mold and solidified at 45°C for 8 hours in the drying oven (HQ-FDO 260, Coretech Korea Co., Anyang, Republic of Korea) to obtain the SPI composite films. Table 1 showed the sample codes used in this study.

Measurement and characterization

SPI film images were captured with a digital camera (iPhone 13 Mini, Apple Inc., Cupertino, CA, USA).

Table. 1. The sample codes used in this study.

Sample code	SPI (%)	Glycerol (%)	CNF (%)	SF microfiber (%)	Glyoxal (%)
SPI	100	0	0	0	0
T-CNF	0	0	TEMPO–oxidized CNF 100%	0	0
M-CNF	0	0	Mechanical grinded CNF 100%	0	0
E-CNF	0	0	Enzymatic hydrolyzed CNF 100%	0	0
SF	0	0	0	100	0
SGC(15,25)	100	15, 25	0	0	0
SGC15SF(3~10)	100	15	0	3~10	0
SGC15T(3~10)	100	15	TEMPO–oxidized CNF 3~10%	0	0
SGC15M(3~10)	100	15	Mechanical grinded CNF 3~10%	0	0
SGC15E(3~10)	100	15	Enzymatic hydrolyzed CNF 3~10	0	0
SGC15O5T(3~10)	100	15	TEMPO–oxidized CNF 3~10%	0	5
SGC15O5M(3~10)	100	15	Mechanical grinded CNF 3~10%	0	5
SGC15O5E(3~10)	100	15	Enzymatic hydrolyzed CNF 3~10	0	5
SGC15O5SF(3~10)	100	15	0	3~10	5

The mechanical properties of the SPI composite films were assessed using a universal testing machine (OTT-003, Oriental TM, Ansan, Republic of Korea) with a 3-kgf load cell. The extension speed and gauge length were set at 10 mm/min and 30 mm, respectively. SPI composite films, preconditioned at 20°C and 65% RH for over 24 h, were cut into 50-mm × 5-mm pieces. Seven samples were tested for each condition, and the measurement results' average and standard deviation were calculated after excluding the highest and lowest values from the five results (Choi *et al*., 2023; Kim *et al*., 2022).

The molecular conformation of the SPI composite films was analyzed using Fourier transform infrared spectroscopy (FTIR; Nicolet 380, Thermo Fisher Scientific, Waltham, MA, USA) with the attenuated total reflection (ATR, Smart iTR ZnSe) method. The parameters were set at a scan range of 4000–650 cm^{-1}, 32 scans, and a resolution of 8 cm^{-1} (Bae and Um, 2022; Lee *et al*., 2021).

Results and Discussion

Effect of glycerol on the film forming of SPI composite

Plasticizer plays a crucial role in improving the processability and flexibility of SPI films (Rani and Kumar, 2019), and glycerol is the most widely used plasticizer in SPI films (Rani and Kumar, 2019). Therefore, in the present study, glycerol was added to SPI to improve its flexibility.

Fig. 1A-D presents digital images of SPI films manufactured with different amount of glycerol addition. In case of pure SPI film (i.e., no addition of glycerol), it is too brittle and was broken during the drying (Fig. 1A). This result is consistent with that of previous reports (Choi *et al*., 2011; Denavi *et al*., 2009). This brittleness was enhanced by adding glycerol and SPI film with 15% glycerol showed a good film formation without any crack or breakage of film (Fig. 1D). At 10% glycerol addition, the film does not cut smoothly (Fig. 1E). However, SPI film with 15% glycerol showed a smooth cut-out picture indicating the flexibility of SPI was quite improved by adding 15% glycerol (Fig. 1F). Also, Fig. 1G showed that the SPI film with 15% glycerol can be bent without breakage confirming the improvement of flexibility of this film.

The previous studies reported the flexibility of SPI film with 25% glycerol addition could be quite improved and they used 25% glycerol added SPI in their studies (González *et al*., 2019; Jensen *et al*., 2015; Martelli-Tosi *et al*., 2018; Qin *et al*., 2019). However, a use of too much amount of glycerol may deteriorate

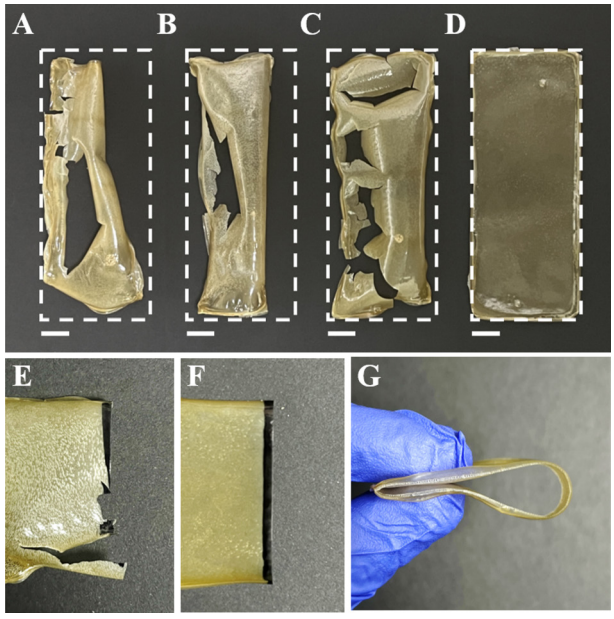

Fig. 1. Photographic images of (A-D) SPI films added with different amount of glycerol (A: 0, B: 5%, C: 10%, D: 15% on the weight of SPI), (E-F) cut-out pictures of SPI films added with glycerol (E: 10% and F:15% glycerol on the weight of SPI), and (G) bent SPI films added with 15% glycerol on the weight of SPI. The white bars in A-D represent 1-cm.

the advantage of pure SPI. As can be seen in Fig. 1, because the addition of 15% glycerol resulted in significant improvement of flexibility of SPI film, in the present study, we used SPI film with 15% glycerol in the subsequent study.

Effect of biofibers on the structure and properties of SPI composite films

In this study, CNFs and short SF microfiber were added to glycerin/SPI to improve its mechanical properties. Table 2 showed diameter and length of CNFs and short SF microfiber used in this study.

Fig. 2 showed mechanical properties of SPI composite film added with glycerol and biofibers. Because most previous studies used addition of 25% glycerol, SPI film with 25% glycerol was prepared for comparison. As the amount of addition of glycerol decreased from 25% to 15%, the breaking strength of SPI film increased two fold and the elongation at break decreased 2.2 fold. In case of addition of 15% glycerol, as the amount of biofibers addition increased to 10%, the breaking strength of biofiber/glycerol/SPI composition film increased. However, the degree of increasement of breaking strength depends on the type of biofiber added. That is, the addition of short SF microfiber was the most effective in increase of the breaking strength of SPI composite film among the biofibers. Also, TEMPO–oxidized CNF was the most effective in increase of breaking strength of SPI composite film among the CNFs.

On the other hand, TEMPO–oxidized CNF was the most effective in increase of elongation of SPI composite film. That is, as the amount of TEMPO–mediated oxidized CNF increased to 7%, the elongation at break of film increased from 21.8% to 42%. After the 7%, the elongation of film decreased. This might

Table. 2 Diameter and length of biofibers used in this study

	TEMPO–oxidized CNF	Mechanical grinded CNF	Enzymatic hydrolyzed CNF	SF microfiber
Diameter	5–10 nm	< 500 nm	< 30 nm	6-21 μm
Length	< 2 μm	< 300 μm	< 10 μm	< 200 μm

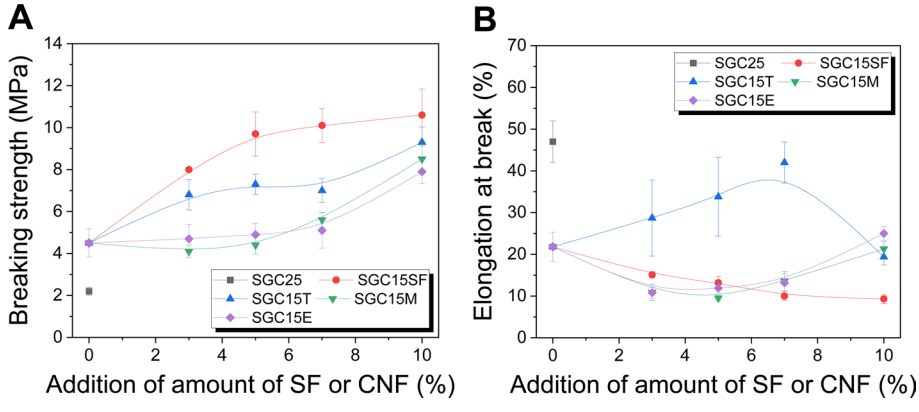

Fig. 2. (A) Breaking strength and (B) elongation at break of the biofiber/glycerol/SPI composite films.

be due to that the extra amount of TEMPO–oxidized CNF (i.e., 10%) reduces its contribution on the increase of elongation of SPI film. The other two types of CNF (mechanical grinded CNF and enzymatic hydrolyzed CNF) showed a similar effect on the elongation behavior of SPI composite film. That is, the elongation of SPI film decreased until 5% and then increased after that. On the other hand, the elongation of SPI added with short SF microfiber decreased constantly with increasing amount of SF microfiber.

It is noteworthy that the addition of short SF microfiber is more effective in increase of breaking strength of SPI composite film than the addition of CNFs. Considering the higher specific surface area and larger aspect ratio of CNF than the SF microfiber, this result was unexpected. Although the exact reason for it cannot elucidate in this study, it is assumed that a similar molecular structure of SF fibroin and SPI (i.e, the both are a protein polymer) favors the molecular interaction of SF and SPI and this might result in a better surface adhesion between the micro SF fiber and SPI matrix than the cellulose and SPI leading to a more increase of breaking strength of SPI film. The higher breaking strength of SPI composite film added with TEMPO-oxidized CNF than mechanical grinded CNF and enzymatic hydrolyzed CNF might be due to its fineness of fiber. That is, as can be seen in Table 2, TEMPO–oxidized CNF has a much lower diameter of fiber (5–10 nm) than mechanical grinded CNF (< 500 nm) and enzymatic hydrolyzed CNF (< 30 nm). The lower diameter of TEMPO–oxidized CNF result in the much higher specific surface area than mechanical grinded CNF and enzymatic hydrolyzed CNF resulting in more effect of increase of breaking strength. Kim *et al*., 2021 also reported similar results in their study. That is, the breaking strength of TEMPO-oxidized CNF film was higher than that of enzymatic hydrolyzed CNF film.

Fig. 3 showed ATR-FTIR spectra of SPI composite films. SPI showed IR absorptions at 1630 cm^{-1}, 1530 cm^{-1}, and 1235 cm^{-1} attributed to amide bond of SPI (Bai *et al*., 2023). The short SF fiber showed absorptions at 1620 cm^{-1}, 1515 cm^{-1}, and 1230 cm^{-1} attributed to β-sheet crystallite (Kim *et al*., 2023; Lee *et al*., 2022; Um *et al*., 2001). CNFs showed IR absorptions at 1160 cm^{-1} (C_1-O–C_5 stretching), 1110 cm^{-1}, 1025 cm^{-1} (C_1–O–C_4 stretching) (Poyraz *et al*., 2017; Wang *et al*., 2020). Additionally, TEMPO-oxidized CNF showed broad absorption at 1600 cm^{-1} (carboxylic acid sodium salt group) (Huang *et al*., 2019). When 15% glycerol was added into SPI (i.e., SGC15), IR absorptions at

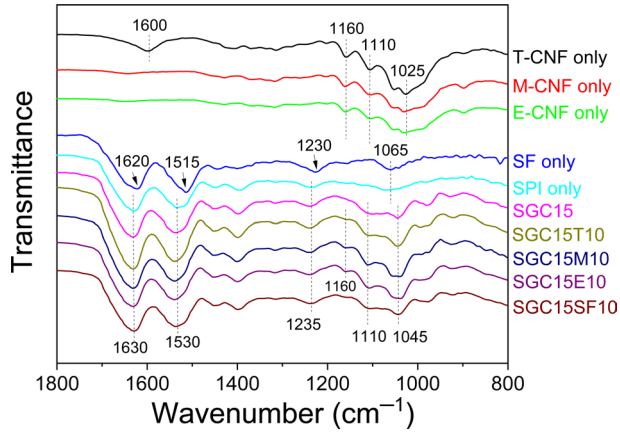

Fig. 3. ATR-FTIR spectra of SPI composite film added with glycerol, SF microfiber, and cellulose nanofiber.

1110 cm^{-1} and 1045 cm^{-1} due to secondary and primary alcohol group of glycerol, respectively were shown (Maquirriain *et al*., 2022). SPI composite films (SGC15, SGC15T10, SGC15M10, SGC15E10, and SGC15SF10) showed characteristics IR peaks of each polymer confirming all component polymers exist in the SPI composite film. No additional peak due to the specific interaction between the component polymers was appeared.

Effect of the glyoxal on the mechanical properties of SPI composite films

The various biofibers were added to improve mechanical properties of SPI film. The limited mechanical properties of SPI film might also be overcome by employing crosslinking agents to react with the reactive groups (e.g., –NH_2, –OH, –COOH, and –SH) in SPI (Choi *et al*., 2011). Glyoxal is a crosslinking agent with relatively low toxicity (Peles and Zilberman, 2012) and has been proven effective in enhancing mechanical properties of SPI (Choi et al., 2011; Peles and Zilberman, 2012). Therefore, in this present study, SPI/glycerol/biofibers mixtures were crosslinked by treating with glyoxal to enhance the mechanical properties of SPI.

Fig. 4 showed the effect of biofiber addition on the mechanical properties of SPI composite film crosslinked with glyoxal. Breaking strength of glycerol(15%)/SPI film (SGC15) was increased from 4.5 MPa to 8.5 MPa by crosslinking with glyoxal. With addition of biofibers the breaking strength was more increased to 11.6~12.3 MPa, as the amount of biofibers increased to 10%.

It is noteworthy that the type of biofiber did not affect the

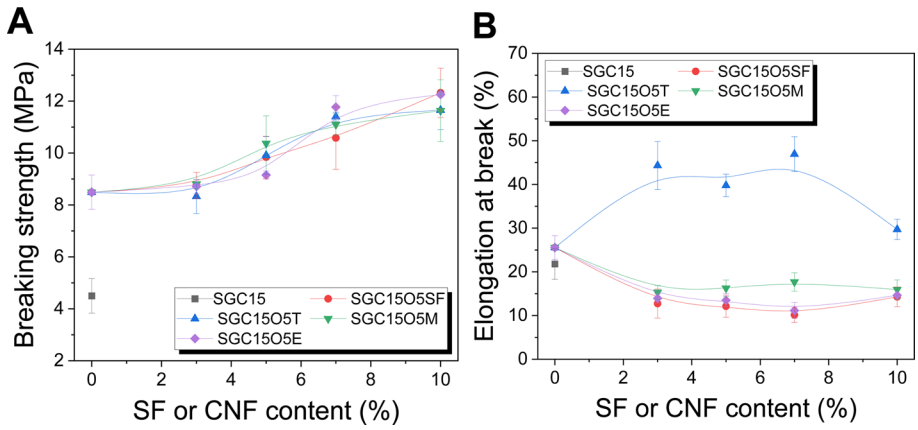

Fig. 4. Effect of amount of addition of biofibers on the (A) breaking strength and (B) elongation at break of SPI composite films crosslinked with glyoxal.

mechanical properties of SPI crosslinked with glyoxal. Although the exact reason cannot elucidate in this study, it is assumed that the effect of crosslinking on the breaking strength of crosslinked SPI composite films overwhelms the effect of the filler on the breaking strength of them.

A different result was shown in elongation at break of crosslinked SPI composite film. That is, as the amount of SF microfiber, mechanical grinded CNF, and enzymatic hydrolyzed CNF increased, the elongation at break decreased. On the other hand, in case of TEMPO–oxidized CNF, the elongation at break of crosslinked SPI composite film increased significantly to 39.8~46.9% until 3% addition and it kept constant until 7%. After that, it decreased to 29.7%. This indicates that TEMPO–oxidized CNF contributes the improvement of elongation of crosslinked SPI composite film. Although the exact reason for these results cannot elucidate in this study, it is assumed that the effect of crosslinking is more dominant in breaking strength of SPI composite films than the type of biofiber addition and that the fiber diameter of bio-filler (i.e., biofiber) strongly affects the elongation of SPI films.

To exmaine the introduction of crosslink among the mixed components (i.e., glycerol and SPI) and biofibers (SF microfibers and CNF) and their structural change by treating with glyoxal, FTIR measurement was conducted on the glyoxal treated SPI composite films and the results was shown in Fig. 5. Compared to untreated SPI/glycerol compoiste film (SGC15), glyoxal treated SPI/glycerol compoiste films (SGC15O5, SGC15O5T10, SGC15O5M10, SGC15O5E10, and SGC15O5SF10) showed a new weak IR absorption peak at 1160 cm^{-1}. This peak is attributed to C-O-C stretching by ester group resulted from

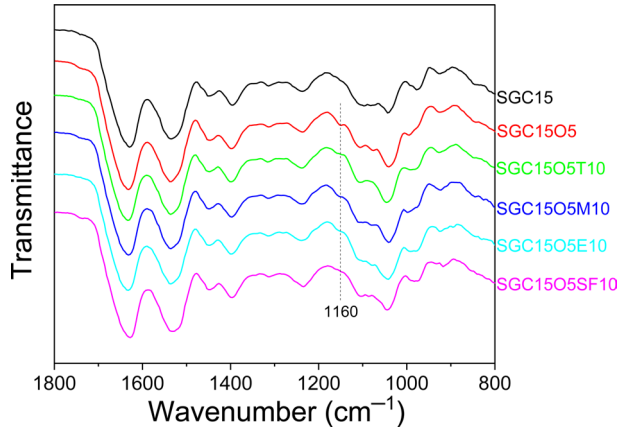

Fig. 5. ATR-FTIR spectra of SPI/glycerol/biofiber composite films crosslinked with glyoxal.

crosslinking reaction of glyoxal and SPI. That is, Wu *et al*., 2021 reported that –NH_2 and –COOH groups of SPI and –OH of glyoxal react result in crosslink between SPI molecules. All glyoxal treated SPI/glycerol compoiste films showed the IR absorption peak at 1160 cm^{-1} indicating SPI molecules were crosslinked.

Conclusions

In the present study, glycerol and biofibers (short silk fiber and CNF) were added to soy protein isolate (SPI), and glyoxal was used to crosslink SPI molecules to enhance mechanical properties of SPI film. The addition of 15% glycerol significantly improved film forming ability and flexibility. Among biofibers, short SF microfibers were the most effective in enhancing

breaking strength, while TEMPO-oxidized CNF excelled among CNFs. Notably, TEMPO-oxidized CNF significantly improved elongation at break until 7%, beyond which its impact diminished.

Glyoxal crosslinking effectively addressed the mechanical properties of SPI. Breaking strength of glycerol(15%)/SPI film (SGC15) increased from 4.5 MPa to 8.5 MPa with glyoxal, reaching 11.6-12.3 MPa with biofiber additions. The type of biofiber minimally affected the mechanical properties of the crosslinked SPI composite films, suggesting the dominant role of crosslinking.

This study, at a preliminary stage, presents research findings at a basic level for SPI/composite films. However, these initial results can serve as foundational data for developing high-performance SPI/composite films using biofibers in the future. To apply SPI materials in areas such as food packaging and biodegradable films, further systematic research is deemed necessary.

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