OLIVE OIL-ZnO/POLY (URETHANE) COMPOSITE NANOFIBERS AS PROPOSED FOOD PACKAGING MATRIX

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ABSTRACT

The aim of this work was to develop novel organic-inorganic hybrid nanofibrous packaging mat by means of simple electrospinning method. The fabricated packaging mats were characterized in detail using sophisticated techniques and tested against the widespread spoilage organisms. The introduced biodegradable packaging material displayed potent antimicrobial activity against *S. aureus* and *S. typhimurium.* The outcome of our investigation clearly suggests that the fabricated biodegradable nanofibrous mats are promising materials and can be used for packaging fresh/or processed foodstuff, meat and meat based products. Thus, we can conclude here that the prepared antimicrobial organic-inorganic hybrid nanofibers provide another attractive solution for packaging material to be utilized in meat and food packaging industries. The as-synthesized nanofibers seem to be capable to replace non-degradable films and overcome the recycling complexity. However, a detailed study is needed to investigate toxicity issues before this material can be reliably used.

Keywords – Food products, Packaging; Spoilage microbes; Biodegradable matrix; Olive oil

I. INTRODUCTION

Food spoilage is result of microbial activity in food matrix causing decomposition of carbohydrates and proteins [1, 2]. Meat and meat products are also frequently contaminated with *L. monocytogenes, S. typhimurium, S. enteritidis,* and *Y. enterocolitica* responsible for foodborne illnesses and in severe case deaths. Recent reports have shown that different types of food and environmental sources harbor bacteria that are resistant to one or more antimicrobial drugs [3, 4].The foodborne pathogens which were found to be resistant to a range of antibiotics are *E. coli, S. aureus, S. typhimurium, B. cereus, K. pneumoniae, Enterobacter* spp. [5]. Among the predominant bacteria involved in foodborne diseases; *S. aureus* is a leading cause of gastroenteritis resulting from the consumption of contaminated food. Likewise; *Salmonella* is a leading cause of bacterial food-borne diseases in both developed and developing countries [6]. Thus it is given to understand that additional measures should be used to ensure the safety of food products. In recent years, much weightage has been imparted on the safety of foods owing to cross contamination caused by spoilage food microorganisms. To prevent the development and spread of spoilage and pathogenic microorganisms via meat foodstuffs, antimicrobial packaging, especially based on the natural substance and biodegradable polymer could be a potential alternative. In the present investigation, we opted biocompatible, biodegradable poly(urethane) (PU) as a driving source to support the spinning progression of olive oil. PU is thermoplastic polymer having outstanding mechanical properties and water insolubility [7, 8]. PU possesses good barrier properties and oxygen permeability and more to the point it is an FDA approved polymer. Generally, chilled meat is packaged in polystyrene trays and wrapped in plastic polyvinyl chloride (PVC) film. However, these films impose recycling issues. The present study explores possibility of replacing PVC film with introduced biodegradable packaging material in order to get rid of microbial contamination and preserve characteristic features (such as color, taste etc.) of meat.

II. MATERIALS AND METHODS

Electrospinning of food packaging mats The fabrication of pristine PU and olive oil-ZnO /poly(urethane) hybrid mats was carried out by electrospinning. PU solution (10 wt%) was prepared by dissolving PU in DMF: THF (1:1 w/w) under magnetic stirring overnight. 5% weight% virgin olive oil was added into polymer solution and stirred for about 2 h at room temperature to get uniform olive/PU composite solution. For the preparation of olive oil-ZnO/poly(urethane) hybrid mat, 5 wt% of virgin olive oil and 3 wt% of ZnO were added together in the polymer solution and stirred for an hour prior to electrospinning. The obtained sol-gel was transferred to a 10 ml syringe (Shinchang medical Co. Ltd., Republic of Korea) fixed with a plastic micro-tip as the spinning head. A copper pin connected to high voltage power supply was inserted in the solution as a positive terminal whereas a ground iron drum covered by a polyethylene sheet served as counter electrode. The solution was kept in capillary by adjusting inclination angle. A voltage of 15 kV was applied to this solution. The distance between the syringe needle tip and collector was fixed at 10 cm. The mats were collected and dried in a vacuum oven overnight to remove the residual solvents.

Characterization

The XRD of pristine PU, and olive oil-ZnO poly(urethane) hybrid mats were recorded on X-ray diffractometer (D/MAX 2500, Rigaku Corporation, Tokyo, Japan) with copper Kα radiation (λ = 1.540 Å) over Bragg angles ranging from 10–80 degrees. The morphological features of pristine PU and olive oil-ZnO/PU hybrid mats were observed by a scanning electron microscope (SEM, S-7400, Hitachi High Technologies, Japan). To get insight into topographical, compositional and morphological view of hybrid nanofibers; TEM was taken. The chemical composition of pristine and olive oil-ZnO /poly(urethane) hybrid nanofibrous mats was analyzed by energy dispersive X-ray spectrometer (EDX) equipped with SEM apparatus.

Antibacterial activity

The bactericidal activity of pristine PU and olive oil-ZnO/PU hybrid nanofibers were tested using *S. aureus* and *S. typhimurium* as model organisms. Briefly, the inoculum was prepared from fresh overnight broth (TSB) cultures that were incubated at 37 °C. For antibacterial assay, the bacterial strains were first grown on agar medium and from the agar plates, fresh colonies were inoculated into broth (TSB-100 ml). Growth was monitored at every 4 h with UV-visible spectrophotometer (Shimadzu, UV-2550), till the optical density reached 0.1 at 600 nm. TSB broth was supplemented with known weights of pristine PU and oil/PU-ZnO composite mats respectively. All the flasks were incubated at 37 °C in a rotary shaker with shaking at 150 rpm. The growth rates and the bacterial concentrations were monitored by OD measurements as aforementioned at an interval of 4 h for 16 h. Furthermore, we also checked the changes in the pH and color of meat packed in the antimicrobial hybrid mat in the present study.

III. RESULTS

Figure 1 shows the SEM images of the pristine PU and organic-inorganic hybrid matrix. PU solutions containing 5 wt% of the polymer gives rise to well-defined nanofibers with diameters in the range of about 250–550 nm(±10 nm) as demonstrated in Figure 1a. Plain electrospun nanofibers obtained had a smooth surface, bread free morphology. We initially tried 10% of olive oil with respect to PU polymer, but this couldn't yield very well defined nanofibers (Fig. 1b). Then we gradually decreased concentration of oil. The 5% doping of olive oil in PU resulted good morphology (Fig. 1c). The incorporation of virgin olive oil into the spinning solution altered the fibrous morphology (Fig. 1b, c). From the images (Fig. 1b, c) merged fiber morphology is evident and this interconnection between fibers could be due to low volatility of olive oil. The shift from the non-bonded to point-bonded fiber morphology of mat significantly affects the mechanical strength and stability of mat, which is desirable for packaging material. Figure 1d shows the TEM image of the ZnO doped PU/Olive oil nanofibers after electrospinning. ZnO nanoparticles were observed as small particles and cluster in/on the surface of composite nanofibers. Figure 2 shows EDX analysis of the plain and olive oil/PU-ZnO nanofibers. The signals of carbon and oxygen were observed for pristine PU nanofibers (Fig. 2a). As revealed by the figure 2b, the fibers contained carbon, oxygen, and Zn. The presence of olive oil and ZnO NPs in the dispersed composite was further confirmed by XRD pattern (Fig. 3). The ZnO crystals in the nanofibers have hexagonal structure (JCPDS no. 891397). Thus XRD spectrum clearly indicated the presence of ZnO particles in the composite nanofibrous mats. The results of XRD are consistent with EDX.

IV. DISCUSSION

Electrospun nanofibers are rapidly replacing the conventional packaging materials such as glass, ceramics, metals, etc. due to significant interesting characteristics such as light weight, low cost and ease of processability etc. However, in spite of many interesting properties, their capability to resist deformation is lower as compared to metals and ceramics. To address the aforementioned shortcomings and to meet the increasing expectations of customers there have been extensive investigation of suitable packaging materials with desired qualities. In the present study we have designed a hybrid material which is made up of polymer as well as the ZnO nanoparticels. Nanoparticles have proportionally larger surface area and significant aspect ratio than their micro-scale counterparts, which promotes the development of mechanical and barrier properties. Thus, it is expected that the performance of designed mat could be improved for food packaging by adding ZnO nanoparticles. Moreover, the advantage of using PU as the polymer is that PUs has increased tensile strength and melting points making them more durable. Their resistance to degradation by water, oils, and solvents make them excellent for the replacement of plastics (Gary 2002; Kriegel et al., 2008). In summary, the results of our investigation suggest that the fabricated biodegradable nanofibrous mats are eco-friendly and could be used to replace PVC films for packaging fresh processed meat and meat based products. Some improvements of the properties or modifications of these materials can also be explored from the industrial application viewpoint.

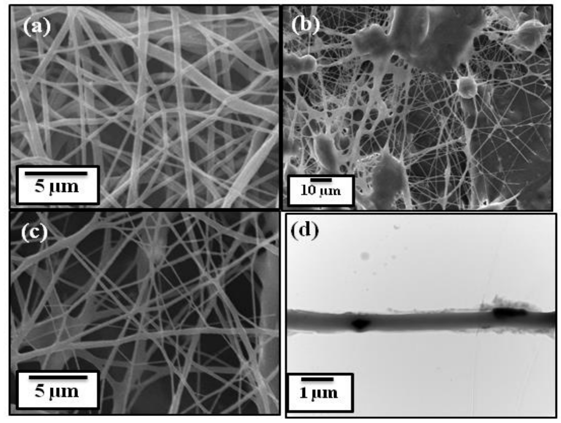

Fig. 1. SEM images of (a) pristine PU (b) Olive oil (10%)/PU (c) Olive oil (5%)/ZnO-PU (d) TEM (5%)/ZnO-PU hybrid nanofibers.

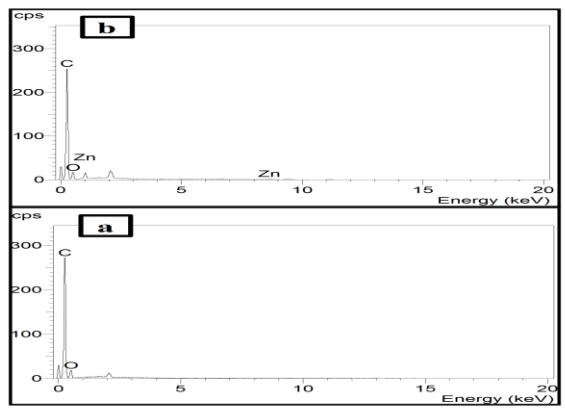

Fig. 2. EDX spectra of (a) pristine PU (b) hybrid food packaging mat

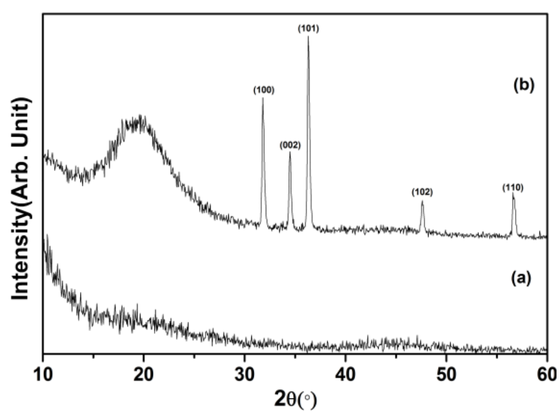

Fig. 3. XRD pattern of (a) pristine PU (b) hybrid food packaging mat

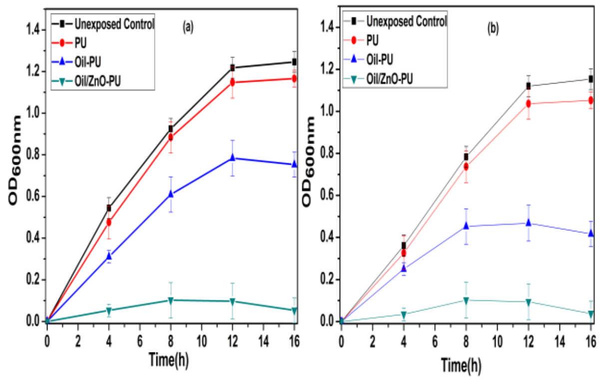

Fig. 4. Growth curve of (a) *S. aureus* and (b) *S. typhimurium* on hybrid packaging mat

V. CONCLUSION

In order to control growth and expansion of spoilage microbes by means of foodstuffs, antimicrobial packaging can serve as a potential alternative. The objective of this study was to develop a new class of antimicrobial hybrid packaging mat composed of biodegradable polyurethane supplemented with virgin olive oil and zinc oxide through electrospinning. Instead of mixing antimicrobial compounds directly with food, incorporation in packaging materials allows functional effect at food surfaces where microbial activity is localized. The nanofibers were characterized by SEM, EDX, XRD and TEM. The antibacterial activity was tested against two common foodborne pathogens viz., *S. aureus* and *S. typhimurium*. The present results indicated that incorporation of olive oil in polymeric nanofibers were able to inhibit growth of pathogens.

VI. FUTURE STUDY AND APPLICATIONS

It is undoubtedly explicable that key purpose of food packaging is to retain quality and safety of food and food by-products during storage and transportation and to increase shelf-life of food by preventing unfavorable conditions such as spoilage microorganisms, chemical contaminants, oxygen, moisture, etc. To attain these above mentioned functions, packaging materials provide physical protection and create proper physicochemical conditions for products that are essential for obtaining a satisfactory shelf life and maintaining food class and safety.

The as-spun proposed mat can be used as future antimicrobial packaging material, which potentially reduces contamination of food products.

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