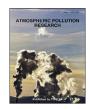
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Short Communication

A facile and cost-effective method for removal of indoor airborne psychrotrophic bacterial and fungal flora based on silver and zinc oxide nanoparticles decorated on fibrous air filter

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ABSTRACT

In tropical countries, food and agricultural crops need to be kept cool to reduce spoilage and quality losses. Airborne psychrotrophic bacteria and fungi can cause adverse effects on food quality and consumers' health safety. The present study aimed to present a facile and cost-effective approach to remove airborne microbes from indoor air by employing silver (Ag) and zinc oxide (ZnO) to decorate fibrous air filters. A water-based anti-germ solution containing Ag/ZnO nanoparticles was first prepared using highspeed homogenization. Second, a commercially available washable non-woven air filter (thickness 50 mm) was coated by aerosol generated from the mixture using spray coating process. This facile method successfully led to homogeneous coating of active nanomaterials on the filter's surface as unveiled by scanning electron microscope (SEM) with energy dispersive X-ray spectroscopy (EDX). On laboratory scale, the Ag/ZnO air filter was shown to exhibit antibacterial effectiveness against Staphylococcus aureus and Escherichia coli under contact mode following an antibacterial standard method (AATCC 147-2011). Finally, the Ag/ZnO filter was assembled into a commercial air filtration system $(670 \times 820 \times 1420 \text{ mm})$ containing two UVA light lamps (365 nm). The Ag/ZnO air-filtration unit was placed in a 45-m³ cold storage room (4–5 °C) for evaluation of airborne psychrotrophic microbial reduction efficiency. The developed Ag/ZnO air filter reduced the airborne psychrotrophic germs concentrations by ~50% and its efficiency increased to ~70% when combined with UVA illumination. Based on these results, a simple and low-cost ZnO/Ag air filter was successfully introduced as an effective strategy for removal of psychrotrophic microbes from indoor air.

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1. Introduction

Nowadays, massive quantities of food are still lost due to spoilage on the journey to consumers (FAO, 2011; Kiaya, 2014), and especially wastage in tropical countries can regularly be as high as 40–50% (SPORE, 2011). The tropical weather and air microbiological pollution contribute to the safety and quality of food during both storage and production (Asefa et al., 2009; Kiaya, 2014). Low

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Peer review under responsibility of Turkish National Committee for Air Pollution Research and Control. temperature storage is a common strategy to reduce food losses and suppress microbial growth. Nevertheless, growth of airborne psychrotrophic bacteria and fungi can still occur during cold storage even at -1 or 0 °C and spread rapidly from the infected to adjacent fruits (Bonaterra et al., 2003; Robiglio et al., 2011). Fungi predominate in refrigerated food spoilage over bacteria, where *Penicillium, Aspergillus, Mucor, Cladosporium, Botyrtis*, and *Acremonium* are found to be the predominant fungal genera in food storage refrigerators (Altunatmaz et al., 2012).

Chemical fungicides are commonly and widely utilized to reduce the spoilage in cold storage with reasonable effectiveness. However, public and personal concerns over health and environmental impact of fungicide disposal and residue levels on foods are

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growing. Alternatively, air purification could be one of the most promising technologies to prolong the storage life, increase food safety and protect the surrounding environment from microbial pollutants. Among technologies for air cleaning, air filters are widely used to remove pathogens from the air. Most of modern filters produce clean air similar to that obtained from HEPA filters (Kowalski and Bahnfleth, 1998, 2002). However, filters including HEPA have several limitations. Even though HEPA filter could remove pathogens from air via its fine porous structure, it cannot be cleaned and its replacement is costly. Furthermore, HEPA filters do not have the capability to kill airborne pathogens. Airborne microorganisms can be electrostatic and stick to the filter surface, and such contaminated surfaces can act as a reservoir of pathogenic microorganisms and may potentially induce biofilm formation (Donlan, 2001).

Recently, the biocidal properties of silver nanoparticles (AgNPs) have been applied extensively against a variety of bacteria and fungi (Kuuliala et al., 2015; Pandey and Ramontja, 2016; Quinteros et al., 2016; Zarpelon et al., 2016; Zhao and Stevens, 1998). It was reported that direct contact with silver ions at very low concentrations resulted in a significant decrease in microbial growth. Some researchers have ascribed cytotoxicity of silver to free radical generation, causing high oxidative stress, disturbing cell layer integrity and damaging protein/DNA binding (Bressan et al., 2013; Phrabhu and Poulose, 2012; Quinteros et al., 2016). Emerging functional materials such as zinc oxide (ZnO) nanoparticles have also been reported as antimicrobial agents for killing and reducing activity of numerous microorganisms (Azizi et al., 2016; Esparza-González et al., 2016; Hui et al., 2016).

While material characteristics played important roles (as described above), the practical usefulness of these antimicrobial agents also relied heavily on methods of use. As such, decorations of both Ag and ZnO nanoparticles onto various substrates have been investigated to achieve highly efficient and cost-effective antimicrobial tools. So far, many processes have been applied for metal or metal oxide coating such as plasma coating, spin coating, electrospinning, dip or spray coating. First, plasma coating provides an effective coating method for both organic and inorganic constituents, but requires a large investment of raw materials and instrumentation (Heimann, 2008; Zhao et al., 2006). Second, spin coating imparts unparalleled thickness homogeneity. However, the size of practical substrates is limited by the spinning devices (Spaid and Homsy, 1994). Third, electrospinning techniques have been applied for various surface coatings, but the process requires high voltage and unique precursor solution's characteristic (Dzenis, 2004; Li and Xia, 2004) for jet formation. Fourth, dip coating represents a facile, scalable and cost effective process (Brasjen et al., 2011). However, this method needs substrates to be in specific shapes such as flat surface or unidirectional rods. Finally, the spray coating process (Gilmore et al., 1999; Klinkov and Kosarev, 2006) could cover a large area and requires a lower amount of coating solution compared to dip coating and other methods. Being facile and scalable, the process could also be applied for various solutionbased precursors. For these reasons, the spray coating method was preferred as a highly practical technique to other coating processes.

In this work, the objective is to develop facile, cost-effective and

low maintenance Ag/ZnO air filters prepared by spray coating. A prototype is used to reduce airborne psychrotrophic bacteria and fungi inside a cold storage room at 4-5 °C. The physical and chemical characteristics of the developed fibrous air filter are studied by scanning electron microscope (SEM) with energy dispersive X-ray spectroscopy (EDX).

2. Experimental

2.1. Materials

Zinc oxide nanoparticles with specific surface area of 26.22 m²/g (ZnO, Nano Materials Technology Co., Ltd., Chonburi, Thailand), polyurethane dispersion (PU, Archroma Co., Ltd., Bangkok, Thailand) and silver-zeolite powder (Global Connections Public Co., Ltd., Samuthprakarn, Thailand) were of commercial grade and used as received. Pleated washable synthetic pre filters (PWS filter, Chaimitr Engineering International Co.,Ltd., Samutprakarn, Thailand) were used as received.

2.2. Instrumentation

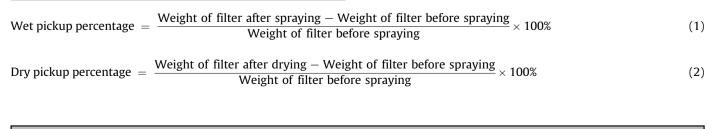
The morphologies of filter membranes were characterized by scanning electron microscopy (SEM, Hitachi model S3400N, 20 kV, working distance 5–7 mm; SEM EDX, 20 kV, working distance 10 mm). The spraying process was performed by using a mini-handheld spray gun (Full star industrial supply co. ltd, Chonburi, Thailand) equipped with an air compressor machine (Puma air compressor, model: XM-2525, Puma industrial Co.,Ltd., Taiwan). The air purification system (Fan filter unit, Chaimitr Engineering International Co., Ltd., Samutprakan, Thailand) was commercially available and modified for air treatment in cold storage room.

2.3. Anti-germ spraying solution preparation

Ag-zeolite powder (60 g) and ZnO (60 g) were suspended in deionized water (250 g) under magnetic stirring for 10 min. Subsequently, the mixture was homogenized by a high-speed homogenizer at 8000 rpm for 15 min. Next, PU (30 g) was added into the mixture followed by homogenization at 8000 rpm for 5 min.

2.4. Filter membrane preparation

First, the filter was placed inside a fume hood prior to spraying process. Then, the as-prepared antibacterial solution mixture (100 ml) was transferred into a spraying chamber equipped with the mini-handheld spray gun before applying 2 bar air pressure. Subsequently, with the spray gun outlets adjusted to 1 mm and with 30 cm distance between the spraying tip and filter, the mixture was sprayed onto the filter for 10 s per area with 3 repeats. The control and sprayed filters were weighed for wet pickup percentage calculation (Equation (1)). Finally, the sprayed filter was dried and cured in a hot air oven (Model: UFB 500, Becthai Bangkok Equipment & Chemical Co., Ltd., Thailand) at 100 °C for 60 min and at 130 °C for 30 min. The dry pickup percentage was calculated with the weight of the dried sample (Equation (2)).



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