1. Introduction

Plasma, being highly reactive, has been used in surface modification of materials to improve surface adhesion, polymerization, sterilization, and etching [1]. Surface modifications are highly dependent on certain parameters being used to produce plasma i.e. gases, power source, treatment time, etc. In plasma treatment of fabrics, there has been numerous studies on mechanical impact [2], on hydrophilicity [2], adhesion [3] and on surface chemical breakdowns [4]. Perhaps the reason for improving fabrics nowadays is its wide applications in daily living for all kinds of people since time immemorial. Studies have proven the role of fabrics in transmission of many diseases by pathogenic microorganisms. Moreover, due to their chemical composition, fabrics are easily colonized by microorganisms causing unpleasant odor, discoloration, and weakening of fibers [4]. Cotton, a cellulosic natural fiber, provides an ideal environment for microbial growth due to its ability to retain oxygen and moisture attracting microorganisms [4]. Therefore, numerous methods have been used to enhance the antibacterial activity of cotton textile. One promising technique in antimicrobial coating of fabrics is metal-based nanoparticle impregnation. Metal-based nanoparticles such as silver, has the ability to penetrate the outer layer of microorganism due to their size and shape [5].

However, Mitchell et al. reported that increased cost associated with enhancing fabrics may be a reason for the limited adoption of technologies to improve fabrics [6]. In recent years, surface modification of textiles by plasma treatment has opened up new possibilities. Plasma processing of fabrics is a dry process offering more advantages than treatment using wet processes. Plasma treatment is also regarded as eco-friendly, and cost-effective as it reduces the consumption of water, energy and chemicals. This green process is proven to keep the original bulk properties of the material.

Cold plasmas or local non-thermal equilibrium plasmas are now being currently used for materials processing as they lessen the possible damages to any material. Cold plasmas are seen suitable for nanoscaled surface modification of fabrics since its bulk temperature is near room temperature. This is due to the atomic and molecular species being near-ambient while electrons are at high temperature [7].

As plasmas may be generated in low and atmospheric pressures, non-thermal plasma is generated by the Atmospheric Pressure Plasma Jet (APPJ), novel device used in this study. This device produces low-pressure plasma with temperatures from 25°C - 200°C that can be used for materials processing [8]. Single gases or mixture of gases flow through a pair of electrodes encapsulated by a glass tube. High voltage from a neon sign transformer is applied to the electrodes to form a glow discharge and with the flow of gases, the plasma discharge is plumed out of the nozzle.
This study aims to test the efficiency of the novel APPJ device in plasma treatment of pure cotton fabric for antibacterial applications.

2. Methodology

The deposition of silver nanoparticles was done using the atmospheric pressure plasma jet (APPJ) device (Fig. 1) adapted from the study of Deang et al. with modification [9]. A glow discharge was successfully ignited between the 5 mm electrode distance of two silver electrodes at 15 kV applied voltage from a 450W neon sign transformer. Silver electrodes were 99.95% pure with 1.5 mm diameter and 8 cm length. Flow rates of argon and oxygen gases were maintained at 15 LPM and 5 LPM respectively.

Pure cotton samples were placed 5 mm from the nozzle and were exposed at different treatment times of 1 min, 3 mins and 5 mins. A control sample (not exposed to plasma) was also prepared for characterization tests and antibacterial tests.

Optical emission spectroscopy (OES) was used to identify the species present in the plasma and measure the electron temperature. Scanning electron microscope (JEOL JSM – 6010LV/LA SEM)) was also utilized to obtain micrographs of the samples. Qualitative elemental analysis was done using Wavelength Dispersive X-Ray Fluorescence Spectroscopy (WDXRF).

Qualitative antibacterial test was done using a modified standard diffusion method as described by Pinho et al. [10]. Samples were tested against Gram-negative E. coli and Gram-positive S. aureus.

3. Results

A bright turbulent flow of purple discharge was produced in the APPJ device. OES analysis revealed that Argon peaks are more dominant over other species such as oxygen and silver (Fig. 2). Since a higher flow of argon was introduced in the APPJ device, the bright purple color can be attributed to this dominant species.

Silver in neutral states was observed at 328.07 nm and 338.29 nm while singly ionized silver peaks were observed at 800.54 nm and 825.47 nm. The presence of ionized silver suggests that silver sputtering occurred successfully in the plasma.

The electron temperature was measured at 0.51 eV (5,915.28 Kelvin) which is much greater than the gas temperature of an argon/oxygen atmospheric pressure plasma as reported by previous studies. It is important to note that the electron temperature must be much greater than the gas temperature to satisfy the condition of a cold plasma suitable for materials processing.

SEM micrographs of plasma-treated cotton samples showed the presence of silver particles sputtered on to cotton fibers (Fig. 3). It can be observed that fluorescent particles are in plasma-treated cotton samples. This fluorescence is typical for metallic contents viewed under a SEM as a result of the excitation of atoms due to the high energy from the electron beam. It can also be observed that more particle coating is present as treatment time increases. This is expected since longer exposure time allows for more particles to be sputtered on the textile. Analysis of elemental composition showed that plasma-treated cotton samples contain silver as evidenced by the silver peak. However, there is no silver peak detection for cotton sample treated for 1 min citing detection limit of instrument used (Fig. 4).

Antibacterial activity of samples with and without surface-immobilized silver nanoparticles was further
tested against Gram-negative bacterium *E. coli* and Gram-positive bacterium, *S. aureus* as test organisms. Zones of inhibition were evident around plasma-treated samples (Fig. 5). Generally, the antibacterial activity of surface-immobilized silver nanoparticles against both test organisms increased with longer treatment time (Fig. 6). Pure cotton fabric exposed to plasma for 5 mins exhibited the highest antibacterial activity and inhibited bacterial growth up to 5.8 mm and 5.2 mm against *E. coli* and *S. aureus*, respectively. The presence of these inhibition zones suggests that the plasma-treated cotton samples with sputtered silver particles inhibited the growth of bacteria. The diameter of the zone of inhibition correlates to the sensitivity of the test organism to silver nanoparticles; a greater diameter indicates higher susceptibility of the organism to the presence of silver nanoparticles.

The specific mechanism of action by which the silver particles acted as antibacterial agents is not determined in this study. Though in previous studies, silver in nanoparticle size only has the capability to act as antibacterial agent due to size, and physico-chemical properties that may disrupt and destroy bacterial cells [5].

### 4. Conclusion

The viability of the APPJ device used in this study was proven as plasma containing argon, oxygen and silver was successfully generated. This approach produced surface-immobilized silver nanoparticles on cotton fabrics as revealed by various analyses. Antibacterial results showed that inhibition zones increased with longer exposure to plasma against both test organisms.

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