**1. Introduction**

With the recent emergence of technical textiles, the focus in the textile industry is rapidly shifting from regular textile fibers to technical textile fibers. Poly(m-phenylene isophthalamide), m-aramid fibers (known as Nomex®) exhibit excellent heat resistance, chemical resistance, abrasion resistance, electrical insulation, and hygroscopic properties, and are widely industrially applicable.

In recent years, m-aramid fibers have been successfully commercially applied in safety work clothes, aircraft seats, arctic clothes, and aircraft blankets. However, m-aramid fibers do not inherently possess antimicrobial properties. For example, in the case of firefighter uniforms, microorganisms may proliferate due to exposure to moisture. Therefore, when the high-performance characteristics of m-aramid are combined with strong antimicrobial properties and durability that can be maintained even with repeated use, it is feasible for these fibers to become a material of choice.

Even though not all microorganisms are dangerous, some microorganisms (i.e., virus, fungi, and bacteria) can cause damage to fiber materials and induce human infections. In order to minimize the damage caused by these microorganisms, various methods such as spraying, padding, dipping, and nanocoating with inorganic antimicrobial materials have been developed for imparting antimicrobial properties to the fibers. However, these methods have the following disadvantages: (i) they induce deterioration of the physical properties of the fibers due to the application of external stimulation such as heat to induce antimicrobial activity, (ii) the antimicrobial activity is non-continuous, and (iii) contamination by microorganisms is possible.

Inorganic antimicrobial materials that are harmless to the human body and possess excellent stability (i.e., are non-volatile and do not readily decompose) have been widely popularized. Among the known inorganic antimicrobial materials, silver nanoparticles (AgNPs) have been the focus of continuous interest and research due to their desirable optical and conductive properties. For example, optical characteristics are used for biosensors and numerous assays, and the conductive properties are used for conductive inks and integrated into composites that enhance thermal and electrical...

**Effective surface attachment of Ag nanoparticles on fibers using glycidyltrimethylammonium chloride and improvement of antimicrobial properties†**

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Functional poly(m-phenylene isophthalamide), m-aramid (known as Nomex®) fibers with antimicrobial properties were prepared by applying quaternary ammonium salts (such as glycidyltrimethylammonium chloride (GTAC)) in combination with silver nanoparticles (AgNPs). The fibers treated by this simple process exhibited enhanced antimicrobial activity. In the coating process, the m-aramid fibers were immersed in a GTAC solution and reacted via the pad-dry-cure process. The GTAC-treated m-aramid fibers were then reacted with an Ag colloid solution at 40 °C for 90 min to prepare GTAC/AgNP-treated m-aramid fibers. Scanning electron microscopy was used to confirm the surface morphology of the m-aramid fibers treated with GTAC and AgNPs. Changes in the chemical composition before and after GTAC and AgNP treatment were analyzed by scanning electron microscopy with energy-dispersive X-ray spectroscopy. The tensile strength of the GTAC/AgNP-treated m-aramid fibers declined by about 3.5% compared to that of untreated m-aramid fibers. Durability of the AgNPs on the m-aramid fibers treated with GTAC/AgNPs was demonstrated through a washing-fastness test, indicating 76% retention after five washing cycles. The antimicrobial activity analysis showed that the synergic antimicrobial properties of the GTAC/AgNP-treated m-aramid fibers resulted in efficacy against P. aeruginosa.
knowledge, it is the properties of direct reaction of the GTAC/AgNPs were investigated. To verify attached into epoxy ring. In addition, this study introduced AgNPs directly trimethylammonium chloride (GTAC), which is containing an attachment on durability and antimicrobial e... Throughout these studies, we expect that AgNPs can be converting power against hundreds of harmful microorganisms. In order to treat the m-aramid fibers with quaternary ammonium salts, solutions of GTAC (a quaternary ammonium salt) were prepared at respective concentrations of 2, 4, 6, 8, 10, 20, 30, and 40 wt% using distilled water as a solvent. The pad-dry-cure process was used for preparation of the sample, and the specific method was as follows. The m-aramid fibers were immersed in 50 mL of GTAC solution prepared at each concentration for 30 min at ambient temperature without shaking. The treated samples were kept at 80 °C for 30 min for drying, followed by curing at 190 °C for 15 min. Finally, the treated samples were rinsed with distilled water to remove unreacted GTAC and dried under ambient conditions.

2.3 Silver nanoparticle-coated m-aramid fibers
The untreated and GTAC-treated m-aramid fibers were immersed in 15 mL of silver colloid and shaken in a water bath at 40 °C for 90 min at 110 rpm. After the coating process, the fibers were washed twice with distilled water and dried naturally.

2.4 Transmission electron microscopy (TEM)
A transmission electron microscope (H-7600, Hitachi, Tokyo, Japan) was used to observe the size and distribution of the AgNPs in the colloid. The acceleration voltage was 120 kV with a resolution of 0.32 nm.

2.5 Scanning electron microscope-energy dispersive X-ray spectroscopy (SEM-EDS) analysis
Analysis of the morphological changes of the antimicrobial m-aramid fibers and elemental analysis of the chemical composition of the sample surface were performed by using SEM-EDS (S-4100, Hitachi, Tokyo, Japan). The surface of the fibers was observed using SEM-EDS with an accelerating voltage of 15 kV. The samples were coated with platinum by sputtering before the examination. In order to compare the surface composition of the pure m-aramid fibers with that of the antimicrobial treated m-aramid fibers, each surface was observed at ×3k magnification using energy dispersive component analysis.
2.6 Washing-fastness test

For the washing-fastness test, the KS test method (K IOS 105-C01: 2007) was used to determine the durability of the AgNPs bound to the m-aramid fibers. This method involved the use of a cylinder made of glass or stainless steel with a diameter of (75 ± 5) mm, height of (125 ± 10) mm, and volume of (350 ± 50) mL. An aqueous detergent solution (5 g L⁻¹) was prepared by using AATCC 2003 standard reference liquid detergent without an optical brightener (Test fabrics Korea, Inc., Ansan, Korea). The prepared solution was placed in a cylinder at a solution/m-aramid fiber ratio of 50 : 1 and washed at a temperature of 40 ± 2 °C for 30 min at a rotation speed of 40 ± 2 min⁻¹ for 30 min. After the washing process, the test specimens were washed with cold water for 2 min and dried at room temperature.

2.7 Tensile strength

The mechanical properties of the m-aramid fibers treated with GTAC and AgNPs were evaluated using the ASTM D 7269 test method. Test specimens with a width of 100 ± 2 mm and a length of 150 mm were prepared in accordance with the test method before analysis. The tensile testing instrument (Model 3345, Instron Corp., Canton, MA, USA) was set at a holding distance of 75 ± 1 mm and a stretching speed of 300 ± 10 mm min⁻¹. The alignment line on the test specimen was grasped to touch the side surface of the front jaw, with a width of 25 mm at the top and bottom; the specimen was then placed in a straight line and the process was repeated at least five times.

2.8 Thermogravimetric analysis (TGA)

The thermal analysis data, including the TGA weight loss curve, were acquired using a TG-DTA (SDT Q600, TA Instruments, Delaware, USA) instrument to analyze the thermal properties of the untreated m-aramid fibers, GTAC, and the Ag colloid-treated m-aramid fibers. The tests were performed under N₂ atmosphere and the temperature was ramped to 600 °C at a heating rate of 10 °C min⁻¹.

2.9 Antimicrobial test

The strains used for the antimicrobial test were Escherichia coli O157:H7 (ATCC 43895) and Pseudomonas aeruginosa PAO1 (ATCC 15692) as Gram-negative bacteria and Staphylococcus aureus MRSA (ATCC BAA-1707) as Gram-positive bacteria. Luria-Bertani (LB; ATCC bioscience, NJ, USA), Nutrient Broth (NB; ATCC Medium, Sigma-Aldrich, USA), and Agar (Difco) were used for the growth of each strain. The cryo-preserved bacterium was placed into the liquid medium and was activated in an incubator for 24 hours at 37 °C. A 1 mL aliquot of the activated liquid medium was inoculated into a sample with dimensions of 1 × 1 cm². After 24 hours, the treated sample was added to 0.85% of physiological saline solution and vortexed, and the diluted solution (controlled by a decimal system) was used for the experiment. The antimicrobial activity was measured using the Disc Diffusion Method (Halo test) and the AATCC Test Method 100 (Contact Assay in Liquid).

Fig. 1 TEM image of AgNPs in the silver colloid solution.

3. Results and discussion

3.1 Surface analysis of GTAC and AgNP-treated samples

The distribution and size of the dispersed AgNPs in the silver colloid solution used for attaching the AgNPs to the surface of the m-aramid fibers were measured by using TEM and the data are presented in Fig. 1. The measured size of the AgNPs was generally found to be less than 20 nm, and the mean size of the AgNPs was observed to be about 11.49 ± 2 nm. The stabilized AgNPs were formed in solution, and most of the bare AgNPs were involved in the reaction. It is known that in water, aggregation of the AgNPs is hindered because the asymmetrical charge distribution on the metal nanoparticle surfaces can be minimized by dissociated water. The proposed experimental plan for inducing effective bonding of the AgNPs on the m-aramid fibers is illustrated in Scheme 1.

In order to compare the surface properties of the pure m-aramid fibers with those of the m-aramid fibers treated with antimicrobial materials, the morphology of each surface, as shown in Fig. 2, was observed at a magnification of ×3000 using a SEM instrument. The data indicated that the untreated m-aramid fibers had a relatively smooth surface (as in Fig. 2a). As
the concentration of the AgNPs increased, the surface of the treated m-aramid fibers gradually became rough. In particular, the surface of the m-aramid fibers treated with 10 wt% GTAC/AgNPs at a concentration of 30 000 ppm showed significant changes (as in Fig. 2d). Fig. 3 shows the SEM-EDS data for comparative analysis of the surface elemental composition of the untreated m-aramid fibers and the m-aramid fibers treated with GTAC and AgNPs. The profile of the m-aramid fibers
treated with GTAC/AgNPs (as in Fig. 3b) showed a peak of Ag from the AgNPs and a Cl peak from GTAC, as compared with that of the untreated m-aramid fibers. Based on the results in Fig. 2 and 3, it was found that GTAC and the AgNPs were successfully bound to the m-aramid fibers, and with increasing GTAC concentration, the AgNPs reacted with GTAC on the m-aramid fibers, leading to a rough surface. These results confirmed that the peak intensity was intensified by the increase of OH-groups at 3200 cm⁻¹ due to epoxy ring opening at the surface of m-aramid fibers by GTAC (ESI 1†).

Results of analysis of the reactivity of Ag (atomic%) towards the m-aramid fibers as a function of various factors are shown in Fig. 4. The change in the loading of Ag (atomic%) on the m-aramid fibers as a function of the GTAC concentration is presented in Fig. 4a. Increasing the concentration of GTAC enhanced the amount of Ag (atomic%) attached to the m-aramid fibers. Specifically, the loading of Ag (atomic%) increased sharply with the addition of up to 10 wt% GTAC, beyond which the increase in the loading of Ag (atomic%) was relatively slow. This is presumably because the number of amine groups (–NH₂) on the m-aramid fiber that can chemically react with GTAC was limited; thus, despite the increase in the concentration of GTAC, only a certain concentration of GTAC could react with the amine groups of the m-aramid fibers. Fig. 4b shows the loading of Ag (atomic%) on the m-aramid fibers with variation of the treatment temperature. The loading of AgNPs increased almost linearly with increasing treatment temperature. This is because at higher temperature, movement of the AgNPs in the treatment solution is enhanced, leading to greater migration to GTAC attached to the m-aramid fibers. The variation of the loading of Ag (atomic%) on the m-aramid fibers as a function of the concentration of the Ag colloid solution, is shown in Fig. 4c. As the concentration of the Ag colloid solution increased up to 30 000 ppm, the loading of AgNPs on the m-aramid fibers increased. The loading of Ag (atomic%) increased markedly in the concentration range of the Ag colloid solution of 0 to 10 000 ppm; however, the loading increased to a lesser extent in the range of 10 000 to 30 000 ppm. It is presumed that the GTAC-treated m-aramid fibers have a certain number of GTAC functional groups that are used to bind the AgNPs; thus, it is easy to bind AgNPs up to a certain concentration of the Ag colloid solution. However, the binding between GTAC and AgNPs approaches saturation at higher Ag colloid concentrations, thereby limiting attachment of additional AgNPs.

3.2 Physical and thermal properties

Table 1 shows the variation of the tensile strength of the m-aramid fibers before and after treatment with the GTAC/AgNPs. The tensile strength of the GTAC/AgNP-treated m-aramid fibers declined slightly (by about 3.5%) compared to that of the untreated m-aramid fibers. This means that there was no

| Control (treated with distilled water) | Treated with GTAC (2 wt%) | Treated with GTAC (10 wt%) | Treated with GTAC (10 wt%)/AgNPs (30 000 ppm) |
|--------------------------------------|--------------------------|---------------------------|-----------------------------------------------|
| 27.12 ± 0.42                         | 27.03 ± 0.98             | 26.62 ± 0.61              | 26.17 ± 0.61                                  |

![Fig. 4](image-url) Ag loading (atomic%) on GTAC/AgNP-treated m-aramid fibers with variation of the (a) GTAC concentration, (b) temperature, and (c) Ag colloid concentration.
significant change of the dynamic properties of the m-aramid fiber even after GTAC/AgNP treatment.

Thermograms of the untreated m-aramid fibers, GTAC-treated m-aramid fibers, and m-aramid fibers treated with GTAC and different concentrations of the Ag colloid are presented in Fig. 5. The initial degradation temperature (IDT) of the m-aramid fibers was 430 °C (as in Fig. 5a). On the other hand, the IDT of the GTAC-treated m-aramid fibers decreased slightly (423 °C). The thermal stability of the GTAC/AgNP-treated m-aramid fibers decreased compared to those of the m-aramid fibers treated with GTAC and also decreased as the concentration of AgNPs increased. The decrease in the IDT is thought to be due to the fact that GTAC, which has relatively poor thermal stability as compared with m-aramid fiber, adheres to the m-aramid fibers. It is also postulated that the metal particles (AgNPs) do not help to improve the thermal stability of the m-aramid fibers.

### 3.3 Durability and antimicrobial efficacy

Durability of the AgNPs attached to the GTAC-treated m-aramid fibers was evaluated after the washing test (Table 2). The durability is important because this property is required to maintain the superior functionality of the functional fibers. In the case of the m-aramid fibers used in this study, (as shown in Table 2) a retention ratio of about 76% Ag was obtained after five wash cycles. In previous studies, AgNPs attached to 3-MPTMS-treated cotton fibers exhibited 66% retention after five wash cycles, while m-aramid fibers treated with GTAC and AgNPs exhibited improved retention despite the use of different materials.27

As mentioned, the purpose of this study is to fabricate synthetic fibers with enhanced antimicrobial efficacy by simultaneously applying GTAC, a quaternary ammonium salt, and AgNPs, another antimicrobial agent. The antimicrobial properties of the untreated and treated m-aramid fibers were verified as shown in Fig. 6 and Table 3. Fig. 6 shows the results of the Halo test, a qualitative method used to visually observe the generation of the zone of inhibition around a sample by placing the sample on a microorganism-coated medium. In the case of the samples treated with AgNPs alone (/#3) and the combination of GTAC/AgNPs (/#4), inhibition bands appeared for the three types of bacteria. To further confirm the antimicrobial performance, a quantitative test method, the count reduction method, was applied (Table 3). In the case of E. coli and S. aureus, the bacteria were simultaneously inactivated in the samples treated with GTAC.

### Table 2 Washing-fastness of GTAC/AgNP-treated m-aramid fibers

| Washing (cycles) | Ag (atomic%) | Retention (%) |
|------------------|--------------|---------------|
| 0                | 1.19         | 100           |
| 1                | 1.13         | 94.96         |
| 2                | 1.07         | 89.92         |
| 3                | 0.99         | 83.19         |
| 4                | 0.91         | 76.47         |
| 5                | 0.90         | 75.63         |

### Table 3 Antimicrobial test results for native and treated m-aramid fibers

| Samples                          | E. coli$^a$   | S. aureus$^b$   | P. aeruginosa$^c$ |
|----------------------------------|---------------|-----------------|-------------------|
| Native m-aramid (#1)             |               |                 |                   |
| m-Aramid treated with GTAC (#2)$^d$ | 6.00 × 10²    | 7.15 × 10²      | 7.8 × 10⁴         |
| m-Aramid treated with AgNPs (#3)$^e$ | 0             | 1.44 × 10³      | 1.70 × 10³        |
| m-Aramid treated with GTAC/AgNPs (#4)$^f$ | 0             | 0               | 1.40 × 10²        |
| m-Aramid treated with GTAC/AgNPs after 5 washing cycles$^g$ | 0             | 0               | 0                 |

$^a$ Total bacteria: 4.8 × 10⁶ cfu per sample. $^b$ Total bacteria: 7.3 × 10⁶ cfu per sample. $^c$ Total bacteria: 9.6 × 10⁶ cfu per sample. $^d$ Sample treated with 10 wt% GTAC. $^e$ Sample treated with 10 wt% GTAC/30 000 ppm AgNPs. $^f$ Sample treated with 10 wt% GTAC/30 000 ppm AgNPs (Ag (atomic%), 1.19). $^g$ Ag (atomic%), 0.90.
(2), AgNPs (3), and GTAC/AgNPs (4). On the other hand, P. aeruginosa, classified as a relatively strong strain, was not inactivated with GTAC only (2) or the AgNPs (3), but was inactivated by the m-aramid fibers treated with GTAC/AgNPs (4). This means that the m-aramid fibers treated with GTAC/AgNPs exhibited increased antimicrobial activity against P. aeruginosa relative to the m-aramid fibers treated with GTAC or AgNPs alone.

4. Conclusion

GTAC and AgNPs were simultaneously and successfully attached to the surface of m-aramid fibers. The washing-fastness test proved that the durability of the AgNPs on the m-aramid fibers was relatively high even after five repeated washes. The GTAC- and GTAC/AgNP-treated m-aramid fibers exhibited slightly (just 3.5%) reduced tensile strength compared to the untreated m-aramid fibers. The GTAC/AgNP-treated m-aramid fibers exhibited enhanced antimicrobial properties compared to the m-aramid fibers treated with GTAC alone or with AgNPs alone. It can be concluded that m-aramid fibers treated with GTAC/AgNPs exhibit increased antimicrobial activity without changes of the physical properties of the fibers and can be used as a multifunctional fiber. Furthermore, m-aramid fibers can be widely applied in industrial textile fields, including medical textiles; thus, the treated fibers presented herein are considered promising.

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