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Anti-bacterial performance of colloidal silver-treated laminate wood flooring

Sumin Kim, Hyun-Joong Kim*

Laboratory of Adhesion & Bio-Composites, Major in Environmental Materials Science, Seoul National University, Seoul 151-921, Republic of Korea

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Abstract

In this study both the anti-bacterial properties and strength of cockroach avoidance of laminate wood floorings containing colloidal silver is evaluated. The laminate wood flooring manufactured with the overlay added with resin containing colloidal silver ion showed an antibacterial activity of up to 98.9%. For colloidal silver-treated, laminate wood flooring, the relative avoidance rate was $87 \pm 1\%$. With colloidal silver treatment onto the surface of the laminate wood flooring, using melamine-formaldehyde resin for overlay paper impregnation, laminate wood flooring was developed as an environmentally friendly material for residential application.

Keywords: Laminate wood flooring; Low pressure melamine; Colloidal silver; Anti-bacterial property; Cockroach avoidance

1. Introduction

There are three types of wood flooring: laminate wood flooring, engineered flooring and solid wood flooring. The laminate wood flooring consists of high-density fiberboard (HDF) as the core material, while the engineered flooring consists of plywood with a thin fancy veneer bonded onto the face of the plywood using urea-formaldehyde and melamine-formaldehyde (MF) resins as hot-press adhesives (Kim and Kim, 2005a).

Interior fitment and furniture manufacturers are using more surfacing materials for decoration of fiberboard. This material is manufactured as uniform, flat panels that provide excellent surfaces for the application of coating materials. These coated panels are used in the construction of cabinets, furniture, paneling, kitchen worktops, and floorings in offices, educational institutions and houses. The purposes of coating fiberboard surfaces with decorative overlays are to suppress the absorption of water and humidity, and eliminate the release of formaldehyde. The performance of the coated panels is dependent on the quality of wood-based panel and the type of coating material (Nemli and Çolakoğlu, 2005; Sparkes, 1993; Hoag, 1993).

A laminate wood floor is a composite floor with either a chipboard or HDF core that is bonded to a film of wood-effect veneer and covered with a laminated surface. It is not to be confused with wood veneer flooring, which has a real wood veneer bonded on top; this is a different type of flooring altogether. Most wood laminate floors are simply a photographic representation of wood grain. Unlike wood veneers, laminates cannot be sanded or refurbished once any wear begins to show. Laminate wood flooring consists of four main components that are bonded together. A wear resistant, decorative surface made of resin-based MF resin and aluminum oxide. This material is bonded to a moisture resistant, wood composition based core. A balancing backing is bonded to the underside of the core. On the top is a clear cap sheet of aluminum oxide, which provides the protection and stain resistance (Kim and Kim, 2005b).

It has been known that among metallic elements, heavy metals such as silver, zinc, copper, mercury, tin, lead, bismuth, cadmium, chromium, and thallium possess antibacterial properties and the exchange with these metals imparts antibacterial activity to the zeolites (Top and Ülkü, 2004).

*Corresponding author. Tel.: +82 2 880 4784; fax: +82 2 873 2318.
E-mail addresses: sumin44@snu.ac.kr (S. Kim), hjokim@snu.ac.kr (H.-J. Kim).

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Silver has a long history of use in medicine as an antimicrobial agent. Silver ions have been found to have antibacterial effects on some microbes. Several studies have demonstrated that silver ions are selectively toxic for prokaryotic microorganisms, with little effect on eukaryotic cells (Park and Jang, 2003; Spadaro and Becker, 1976; Webster et al., 1981; Marino et al., 1974).

Silver exhibits good anti-bacterial properties and in recent years has been used in a variety of medical applications ranging from wound dressings to urinary catheters. The anti-bacterial activity of silver is dependent on the balance between the activity of the Ag^+ ions which kill bacteria and the total amount of silver released from the coating, which if too high results in cytotoxicity The anti-bacterial activity of silver is dependent on the silver cation (Ag^+), which binds strongly to electron donor groups on biological molecules containing sulfur, oxygen or nitrogen. The silver ions act by displacing other essential metal ions such as Ca^{2+} or Zn^+ (Betts et al., 2005; Dowling et al., 2001, 2003; Zhao and Stevens, 1998).

Colloidal silver consists of a very fine particle suspension of the metal in water. When silver particles are suspended and evenly dispersed throughout a solution, all the particles are microscopic and are electrically charged with a positive potential. This suspension can be prepared by the following electrical method. Pass 12–30 V through two silver electrodes in mineralized (salt) water for 2–3 min per glass. Put a small light bulb in the circuit. If the bulb lights, the water is conducting and the process has started. If not, add a little salt to disinfect any unhygienic water as is done while camping. The highest quality colloidal silver is produced by the electro-colloidal/non-chemical method. The silver particles and water have been completely "colloided" and evenly dispersed and held in suspension by an electrical current sent through the combination. This process is the only known method to create a truly homogeneous, i.e., evenly distributed, solution containing super-fine silver particles in the range of 0.005–0.015 μm in diameter, suspended in water, without the need of any chemical, stabilizer, dye, or other ingredients (Becker, 1985).

Researchers in the past have used various methods, including physical and chemical modification of the material surface, to try and prevent bacterial adhesion and slime production on materials. Bridgett et al. (1993) tested bacterial adhesion to cerebrospinal fluid shunts coated with a hydrogel material that created a more hydrophilic surface. This coating, although effective in reducing bacterial adhesion, was difficult to apply uniformly. Silver-impregnated cuffs on catheters have been another approach based on the anti-microbial activity of silver ions (Maki et al., 1988). However, this approach is limited by the degradation of the cuff resulting in the loss of the coated silver ions and thereby the antimicrobial activity (Raad 1998; Baveja et al., 2004).

In this study, we manufactured anti-bacterial, laminate wood flooring with colloidal silver, and then tested its anti-bacterial properties and examined the strength of its cockroach avoidance.

2. Experimental

2.1. Colloidal silver treatment onto the laminate wood flooring

Despite the importance of functionality for laminate wood flooring, it should not be the only factor under consideration. When it is approached without thinking about the production line and equipment, many obstacles can arise during manufacturing despite high functionality. Many anti-bacterial substances have been developed and are applied into different products. However, because most anti-bacterial substances are ceramic powder, it is very difficult to apply on surface of laminate wood flooring. Even though the point of interest is the line where the antibacterial material is applied, each aspect such as low pressure melamine (LPM) impregnation line, melamine faced board (MFB) line, or processing line poses its own problems. For example, when resin in flour form is applied in the impregnation line, it affects the resin composition or precipitate. In the case of the MFB line, it can affect the lifecycle of the cowl or hot pressure condition. When the flour type is applied on a surface, the product brightness or color is affected. In this situation the issue is to find the method able to grant the antibacterial ability while minimizing these problems. In this study, liquid type resin was used for an antibacterial material, i.e., colloidal silver. Colloidal silver comes in neutral pH (7–7.5) and clear so that there is no problem in applying colloidal silver in resin. However, consideration needs to be given to the concentration of colloidal silver, which was very low at 20 ppm in this study. Even the amount of colloidal silver, at 3%, was very low in respect to the amount of melamine resin. As shown in Table 1, 3% "water" added for the preparation of MF resin for impregnation of overlay paper for surface coating was substituted with 3% colloidal silver. Usually, water is added into MF resin when various additives such as hardening agent, plasticizer and release agent are incorporated into MF resin to control viscosity. And then, these are stirred together for 1 h. The colloidal silver was liquid state like water. We added the colloidal silver instead of water.

At 20 ppm, Ag^+ concentration was measured using an inductively coupled, plasma-optical emission spectrometer (Optima 3000DV model, Perkin Elmer Ltd., USA). The following principle is used for this spectrometer. The magnetic field induced by a coil where high frequency current is flowing excites the outermost electrons of neutral atoms using the plasma coupled to the magnetic field as the energy source. Chemical analysis is performed using scattered radiation emitted by this magnetic field. Quantitative analysis is done using spectral peaks, i.e., spectral intensity. Based on this analysis, the silver ion content in the colloidal silver-impregnated paper was 0.006 g m^{-2}.

Table 1
Composition of melamine-formaldehyde resin for impregnation

Composition	Amount added (g)
Melamine-formaldehyde resin	100
Hardening agent	0.82
Plasticizer	2
Release agent	0.4
Water or colloidal silver	3

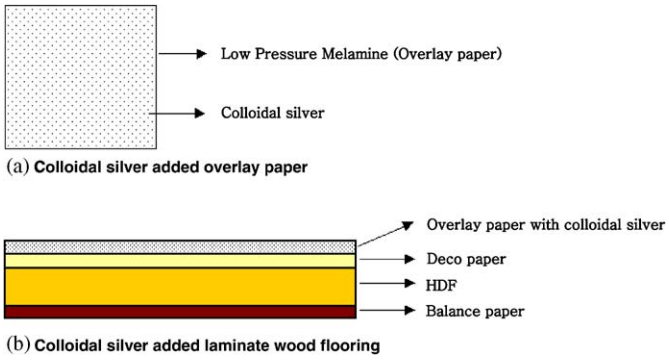

Fig. 1. Application of colloidal silver.

Surface treatment was used to apply the colloidal silver showing antibacterial activity only when contacting the laminate wood flooring. The overlay paper layer was the layer forming the surface of the laminate wood flooring. As shown in Fig. 1, the laminate flooring was composed of four layers: overlay paper, deco-paper, high density fiber-board (HDF), and balance paper. Colloidal silver was applied to resin in the overlay paper layer. To manufacture antibacterial treated laminate flooring by colloidal silver, these three MF resin impregnated papers and HDF were pressed all together as shown in Fig. 1 at 180 °C for 30 s. It can also be applied to the balance paper layer when antibacterial activity is needed in overall laminate flooring.

2.2. Anti-bacterial test

The method of microbial growth inhibition in sanitized, plastic products was examined. The sanitation processing was done to prevent secondary infection by inhibiting microbial growth in plastic products. For anti-bacterial function test, *Escherichia coli* and *Pseudomonas aeruginosa* were used as the bacilli. Table 2 presents the characteristics of these two bacilli. After the bacilli were inoculated to the surface of the colloidal silver-treated, laminate wood flooring and the non-treated, wood laminate flooring, some inoculation liquid was placed between each flooring. Then, the cultured bacteria were extracted. The rate of bacillus reduction between the colloidal silver-treated, laminate flooring and the non-treated, laminate wood flooring was calculated by measuring the number of bacteria present in this cultured bacterial solution in order to quantify the degree of sterilization in the plastic. The sample was a square shape of 40×40 mm in length and width (thickness was 8 mm). The number of test samples was three pairs. The size of the control sample was the same as that of the test sample. Eight and eighteen millimeter laminate wood floorings, the former is a standard apartment room size while the latter is for a school classroom, for the non-treated were tested.

2.2.1. Preparation of culture media

To prepare culture media, the broth medium consisted of 10.0 g of peptone (Bacto-Peptone or Thiotone), 5.0 g of beef broth (Beef Extract), 5.0 g of NaCl (pure or grade 1) and 100 ml of distilled water. These materials were placed in a beaker and dissolved thoroughly and the pH of the solution was adjusted to 6.8 using NaOH. Ten milliliter of the solution was aliquoted into 125×17 mm test tubes, which were placed in a high pressure sterilizer to sterilize at 1055 g cm^{-2} steam pressure and 120 ± 2 °C for 20 min. Agar medium having the same components as the above beef broth was placed at 15 g, heated to dissolve thoroughly, and the pH was adjusted to 7.0–7.2. The sample was placed into test tubes with 15 ml each or into flasks with 100 ml each and sterilized in the high pressure sterilizer at 1055 g cm^{-2} and 120 ± 2 °C for 20 min.

2.2.2. Manipulation

The broth medium containing the bacteria was cultured for 24 h so that the number of bacteria produced in the colloidal silver-treated, laminate flooring and in the non-treated, laminate flooring was $1–2 \times 10^6$ at inoculation time '0'. The medium was then diluted, and 0.1 ml of this diluted sample was used as the inoculation source. Physiological saline solution was used for dilution. Each flooring sample was sterilized at high pressure.

When using bacilli, the bacteria cultured for 24 h were shaken and left for 15 min before inoculation. Using a pipette, 0.1 ml of inoculation source for each sample was smeared onto the surfaces of each flooring sample.

Both the inoculated and non-inoculated, non-treated, laminate flooring sample were placed into a petri dish and washed with neutral solution (20 ml). After shaking for exactly 1 min, dilutions were continued. The diluted sample (0.2 ml) was taken and inoculated onto tryptone glucose extract, agar medium. At this time, the dilution ratios were 10^0, 10^1, and 10^2. The dilution solution was 0.85% NaCl sterilized in a high-pressure sterilizer. The inoculated, colloidal silver-treated, laminate wood flooring sample and the inoculated, non-treated, laminate wood flooring sample were both placed into sterilized plastic bags and placed in constant water bath for 25–24 h at 1 kg/cm^2. After culturing, the test and control samples were placed into petri dishes and washed with neutral solution (20 ml) or shaken slowly with neutral solution (4 ml). Sample (0.2 ml) was obtained from this solution and thinly inoculated into tryptone glucose extract, agar medium. The appropriate dilution ratios at this time were 10^0, 10^1, and 10^2. However, the control sample may have required

Table 2
Characteristics of *E. coli* and *Pseudomonas aeruginosa*

Bacilli	Characteristics
Escherichia coli	There are several million *E. Coli* in our gut aiding in digestion and producing some vitamins It can be pathogenic causing urethral infection, sepsis, meningitis, and scar infection External source *E. coli* causing infection in the gut is different from that indigenous to the gut. Virulent *E. Coli* causes diarrhea by producing enterotoxin
Pseudomonas aeruginosa	This cockroach strain is distributed widely in nature and some types are pathogenic in human and animals Although it does not reside in an animal host, it can infect an animal with weakened immunity Its infection can be fatal in severely burned patients, cancer patients going thorough immune treatment and patients with cystic fibrosis

different dilutions depending on the culturing period. Culturing was done in a plate medium at 37 °C for 48 h.

2.2.3. Quantitative analysis

The number of bacteria per ml of neutral solution or the rate of reduction between process and unprocessed foods was calculated. Conversion for dilution was needed.

$$\text{Reduction rate}\,(\%) = (A - B)/A \times 100, \quad (1)$$

where A is the number of bacteria 24 h later in unprocessed food and B the number of bacteria 24 h later in processed food.

The following criteria should be followed for effective testing:

- the number of colonies generated from the sample not inoculated should be "0";
- there should be a definite decrease in the number of bacteria generated from the control sample cultured for 24 h, in respect to the number of bacteria generated from the control sample at the inoculation time of "0".

2.3. Test for cockroach avoidance

2.3.1. Out line of test

(1) Set the standard specimen and test specimen in the test box.
(2) Set a wooden-made hiding place on the upper part of each specimen so that the cockroach can easily hide in it.
(3) Set some portion of cockroach imago group as standard strain, supply them with water and food, and then observe their hiding status.
(4) Cockroaches prefer dark places, so it is possible to observe the phobic rate of each specimen by observing the number of hidden cockroaches from each sample group. Thus, this quantification indicates the level of cockroach avoidance quantitatively.

The sample was a square shape of 100 × 100 mm in length and width (thickness was 80 mm). The number of N was 2 pairs. The size of the control sample was the same as that of the test sample.

2.3.2. Test device and materials

The test box dimensions were 340 mm (width) × 260 mm (length) × 110 mm (height) (Fig. 2). It was made of slippery glassy material with a plastic inner wall and a thin film on the cover allowing the standard strain cockroaches to breathe. The outer wall, however, was designed to block the light somewhat to prevent the standard strain cockroaches from going up. Two sets of hiding places of the standard strain cockroaches were made to check the environment accommodation and the avoidance rate of both colloidal silver-treated, laminate flooring and non-treated, laminate flooring. This hiding place was composed of two sets of thick paper of dimensions 100 mm (width) × 100 mm (length) × 35 mm (height). Then four holes for the entrance and exit of the standard strain cockroaches at four dimensions in the hiding place were made. For feeding of the standard strain cockroaches, solid type feed and watered cotton were prepared. The standard strain cockroach was the dominant species, *Blattella germanica*. Twenty imago individuals as standard strain cockroaches were set in each test box regardless of their sex.

2.4. Test condition and test

2.4.1. Environment accommodation test of standard strain cockroaches

When testing the standard strain cockroaches, the test box was stored at room temperature. The preliminary treatment of the specimen should be done within 60 ± 3 °C, at 168 h after treatment. We put 20 test standard strain cockroaches in each test box for 1 day. The non-treated, laminate wood flooring was laid on the floor of the hiding place and covered with a piece of paper board. This test followed the test method of FC-TM-401/402 from FITI testing & research institute, Korea (FC-TM, 2003).

2.4.2. Test on relative avoidance of standard strain cockroaches

(1) After the environment accommodation, we removed the hiding place and at the colloidal silver-treated, laminate flooring and the non-treated, laminate wood flooring.

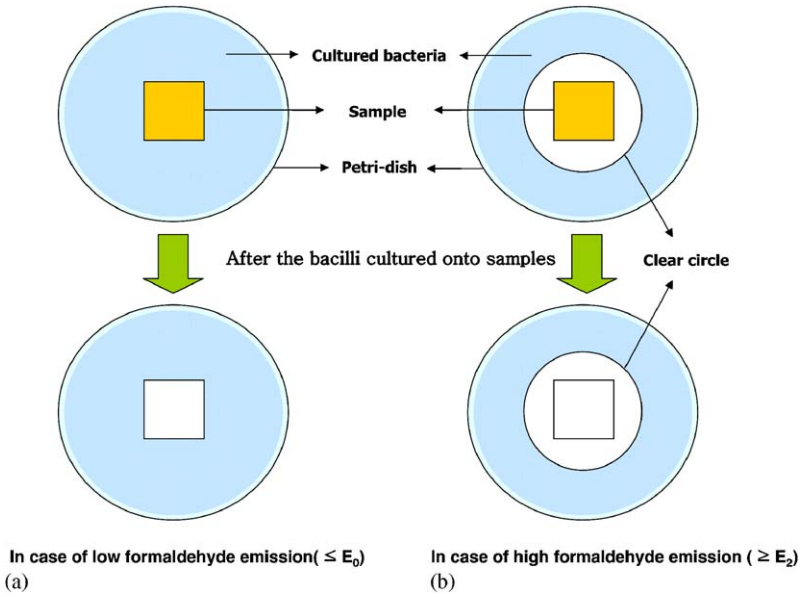

Fig. 2. Effect of formaldehyde emission during antibacterial testing.

(2) A day after set of (1), we removed the paper board of the colloidal silver-treated, laminate flooring and the non-treated, laminate wood flooring, and then observed the numbers of hidden standard strain cockroaches.
(3) Then, we exchanged the positions of the colloidal silver-treated, laminate wood flooring and the non-treated, laminate flooring and left them in a those position for a day, and then observed the numbers of hidden standard strain cockroaches as we done in the previous experiment.

3. Results and discussion

3.1. Anti-bacterial test

Before testing for the antibacterial activity of colloidal silver, antibacterial, laminate flooring, the antibacterial activity of the non-treated laminate was tested first. As shown in Table 2, the antibacterial effect was 25–35%, which was very low. The very low formaldehyde emission was notable. It is claimed that common laminate floorings have a high antibacterial activity probably because of high formaldehyde or VOC emission rate, creating a harsh environment for bacteria to survive in. In other words, the antibacterial activity results mainly from toxicity. Furthermore, in the report (Nemli et al., 2005), the MF coating on particleboard improved the decay resistance of the panel and they claimed formaldehyde in MF coating material is the reason. However, the low antibacterial activity in the non-treated, laminate flooring is due to a low rate of formaldehyde and VOC emission. In Fig. 2, which illustrates the effect of formaldehyde emission during antibacterial testing of laminate flooring, the laminate wood flooring with low-formaldehyde emission (under E_0) did not affect the bacteria surrounding the sample (clear circle), unlike the laminate flooring with high-formaldehyde emission (over E_2) where bacteria surrounding the sample flooring were killed due to high formaldehyde emission. This test demonstrated the effect of formaldehyde emitted by the sample flooring on the antibacterial test. The laminate wood flooring manufactured with the overlay added with resin containing colloidal silver ion showed an antibacterial activity up to 98.9%, as shown in Table 3. This antibacterial activity was not due to the formaldehyde effect but was a result of the pure silver ion. Fig. 3 shows the results of antibacterial testing done in petri dishes. About 65–75% of the bacteria were remaining in the ordinary laminate. In contrast, almost no bacteria are seen in the dish containing the silver ion, antibacterial, laminate wood flooring sample.

As single celled creatures called bacteria use a common type of enzyme or 'chemical lung'' for their oxygen metabolism, the presence of colloidal silver cripples the enzyme, therefore causing the organism to suffocate. Any and all bacteria are therefore killed within a 6-min time frame, without causing any adverse effect on the surrounding tissue cells. Becker also states in his book: ''Positive silver kills all types of bacteria'' (Becker, 1985). This is exiting, because no other single antibiotic works against all types of bacteria. Positive silver, however, offers several advantages over previous forms. There are no ions besides silver to burden side tissues. It works against all types of bacteria and viruses. ''To carry the fight to fungal infection, one must firstly understand fungal growths. A fungus is a series of single cells that have small tubes of the material from which the cell is made, which stretches between the cell walls. Whether more linear in its spread and expand mode or, alternatively, more like a fabric in its reproductive

Table 3
Results of antibacterial testing on the non-treated, laminate wood flooring and silver ion-treated, antibacterial, laminate wood flooring

Items tests	Samples	Initial concentration (CFU/40p)	Concentration after 24 h (CFU/40 p)	Rate of bacterial reduction (%)
Antibacterial test for *Escherichia coli*	BLANK	388	1141	
	Non-treated laminate wood flooring (8 mm)	388	730	36
	Non-treated Laminate wood flooring (18 mm)	388	814	28.7
	Silver ion-treated antibacterial laminate wood flooring	388	12	98.9
Antibacterial test for *Pseudomonas aeruginosa*	BLANK	378	1128	
	Non-treated laminate wood flooring (8 mm)	378	836	25.9
	Non-treated Laminate wood flooring (18 mm)	378	802	28.9
	Silver ion-treated antibacterial laminate wood flooring	378	22	98.0

The numbers of bacteria in the cultures were calculated by multiplying by the dilution ratio.
BLANK: no sample placed; CFU: colony forming unit; 40 p: 0.04 ml.

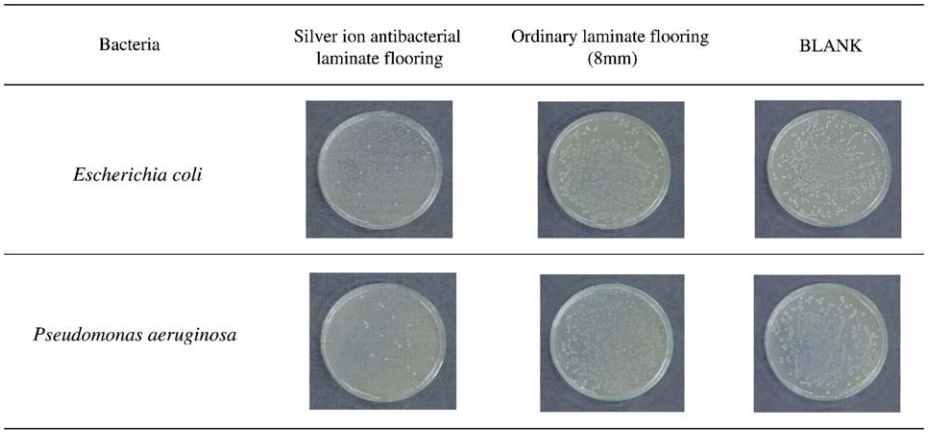

Fig. 3. Comparison of antibacterial activity in petri dishes.

mode, a fungus will still exhibit that characteristic of any single-celled bacterium, that particular type of chemical lung, which is almost completely and permanently disabled by the presence of colloidal silver in the body, as the silver suffocates the organism."

The mechanism of inhibitory action of silver ions on microorganisms is partially known. It is believed that DNA loses its replication ability and cellular proteins become inactivated on Ag+ treatment (Feng et al., 2000). In addition, it was also shown that Ag+ binds to functional groups of proteins, resulting in protein denaturation (Spadaro et al., 1974). The obvious question is how nanosize silver particles act as biocidal material against bacteria. There are reports in the literature that show that electrostatic attraction between negatively charged bacterial cells and positively charged nanoparticles is crucial for the activity of nanoparticles as bactericidal materials (Stoimenov et al., 2002; Hamouda and Baker, 2000; Sondi and Salopek-Sondib, 2004).

Consequently, we blended MF resin and colloidal silver. Though anti-bacterial function is enhanced, productivities of MF resin-impregnated papers and laminate floorings are not obstructed by adding colloidal silver. It is merely added like the water that is used when MF resin is produced because it is colorless and transparent liquid state colloidal silver.

3.2. Test of cockroach avoidance

The relative avoidance rate was tested in the colloidal silver-treated, laminate flooring and the non-treated, laminate wood flooring by placing a sample of flooring in a box. For the colloidal silver-treated, laminate wood flooring, the relative avoidance rate was 88.2% and 86.0%, before and after environmental treatment, respectively, as shown in Table 4. This test was repeated 4 times. The relative avoidance rate was higher than 80%. A higher antipest activity was shown with colloidal silver antibacterial treatment.

Fig. 4 shows the results of relative avoidance by cockroaches. As shown in the figures, the incidence rate of cockroaches in the colloidal silver-treated, laminate wood flooring was relatively lower than that in the non-treated, laminate wood flooring. Considering the fact that

Table 4
Relative cockroach avoidance rate in silver ion antibacterial-treated, laminate wood flooring

Category	Test box								Relative avoidance rate (%)
	A		B		C		D		
	First time	Second time	First time	Second time	First time	Second time	First time	Second time	
Silver ion-treated laminate wood flooring									
Before environmental treatment									
After adaptation	17	18	19	19	16	16	17	17	88.2
Upon confirmation of avoidance	2	2	1	1	2	3	3	2	
After environmental treatment									
After adaptation	17	18	18	18	19	17	16	16	86
Upon confirmation of avoidance	3	2	2	1	1	3	3	4	

(a) Relative phobic rate (Control-Left, Sample-Right) (b) Relative phobic rate (Sample-Left, Control-Right)

Fig. 4. Comparison of relative avoidance rate in the non-treated, laminate flooring and silver ion-treated, laminate flooring after environmental treatment.

the acryl box used for testing was small, this significant difference confirms a significantly high antipest activity in silver ion, antibacterial, laminate flooring.

The antibacterial activity exhibited by silver ion on microbes, such as preventing bacterial activity, damaging the cell membrane, and destroying bacterial cells by changing the protein composition inside bacterial cells, also affected pests such as cockroaches. According to Feng's report (Feng et al., 2000), although the mechanism of antimicrobial effects of silver is still not fully understood, two mechanisms were suggested. (1) As a reaction against the denaturation effects of silver ions, DNA molecules become condensed and lose their replication abilities and (2) silver ions interact with thiol groups in protein, which induce the inactivation of the bacterial proteins. More detail mechanism of this test of cockroach avoidance, biological approach is necessary.

4. Conclusion

We blended MF resin and colloidal silver to manufacture environmentally friendly laminate wood flooring for enhancing anti-bacterial function. The productivities of the MF resin-impregnated papers and laminate wood floorings were not obstructed by adding colloidal silver. It was merely added like the water that is used when MF resin is produced because it is colorless and transparent liquid state silver ion (colloidal silver). The laminate wood flooring manufactured with the overlay added with resin containing colloidal silver ion showed an antibacterial activity of up to 98.9%. This antibacterial activity was not due to the formaldehyde effect but resulted from the pure silver ion.

In the test of cockroach avoidance, for the silver ion-treated, laminate wood flooring, the relative avoidance rate was 88.2% and 86.0%, before and after environmental treatment, respectively. With colloidal silver treatment onto the surface of the laminate flooring, using MF resin for overlay paper impregnation, we have found an environmentally friendly, interior material. Under this framework, our data facilitate further study and insight into the biological effects of colloidal silver. In particular to obtain an explanation for cockroach avoidance of colloidal silver impregnated surfaces.

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