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Development of durable antimicrobial surfaces containing silver- and zinc-ion–exchanged zeolites

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Abstract: The present work involves development of stable antimicrobial materials containing silver- and zinc-ion–exchanged zeolites. Faujasite X and Linde type A zeolites were synthesized, and following ion exchange with Ag^+ and Zn^{++} ions they were found to exhibit antimicrobial effects against bacteria (*E. coli*, *P. aeruginosa*, and *S. aureus*), yeast (*C. albicans* and *C. glabrata*), and fungi (*A. niger* and *P. expansum*). Zeolites-X and -A containing silver and zinc ions were then mixed with various coating materials, including paints and polypropylene, to develop antimicrobial composites. The long-term antimicrobial characteristics of zeolite-containing composite materials were investigated by inoculating selected microorganisms onto the surface of the materials. The results indicated that the higher the zeolite concentration present in the composite, the more long-term antimicrobial activity was achieved. Silver-ion–exchanged zeolites were more effective against bacterial and candidal species, while zinc zeolites exhibited noticeable antifungal properties. Materials manufactured with metal-ion–exchanged zeolites would prevent microbial growth on surfaces, reducing cross-contamination and infection risk as well as the microbial degradation of products.

Key words: Silver, zinc, zeolite, antimicrobial, coating, polypropylene, paint

1. Introduction

Microbial species including fungi, yeast, and bacteria can live almost anywhere on the earth, and some may be primary and opportunistic pathogens causing clinically important diseases in human beings, animals, and plants. In the early 1900s, infectious diseases were the most common cause of death worldwide (Cohen, 2000). Current technology is available to control pathogenic microbial flora under in vivo and in vitro conditions with use of antimicrobial agents such as antibiotics, antiseptics, disinfectants, and synthetic drugs. Over the last century the number of deaths originating from microbial infections has decreased considerably with the development of antimicrobial agents. On the other hand, microorganisms have developed resistance to certain antibiotics due to misuse and overuse (Parekh et al., 2005). The use of high-dose antibiotics resulted in microorganisms with acquired resistance such that the effectiveness of some of the available antibiotics has been invalidated (Gold and Moellering Jr, 1996; Walsh, 2000). Both gram-positive and gram-negative bacterial pathogens that develop drug resistance in hospitals compromise our ability to treat serious infections (Boucher et al., 2009; Rice, 2009; Chanda et al., 2013). This challenging and dynamic pattern of infectious diseases and the emergence of antibiotic resistance demands longer-term solutions (Taylor et al., 2002; Huh and Kwon, 2011). Toxicity, adverse drug reactions, and drug resistance have led scientists to develop novel and safer antimicrobial agents that are effective against most microorganisms.

Natural and manufactured surfaces provide a shelter for microorganisms where they can survive and proliferate. As they increase in number, they secrete extracellular matrix proteins which act as a barrier to external threats and make them 1000 times less sensitive to biocides and antimicrobials (Mah et al., 2003). Microbial contamination of surfaces, especially in the hospital environment, is the major cause of the spread of infection between patients (Scott and Bloomfield, 1990; Binder et al., 1999; Richards et al., 1999). Microbial species including human pathogens can easily adapt to the surface of various materials and survive more than 90 days (Neely and Maley, 2000). A possible solution for preventing surface contamination is the frequent use of disinfectants. However, they have adverse effects on the environment and may not be an economical approach (Siedenbiedel and Tiller, 2012).

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Therefore, developing novel, safe, and cost-effective antimicrobial surfaces that inhibit microbial growth is of considerable interest to scientists.

Zeolites are inorganic, nanoporous crystalline solids (Valdés et al., 2006; Sahner et al., 2008). The negatively charged aluminosilicate structure is balanced with exchangeable alkaline or alkaline earth metal cations. The cation exchange capacity of zeolites can be altered by the SiO_2:Al_2O_3 ratio of the framework. Zeolites are used in petrochemical industries (Ghobarkar et al., 1999); detergent production (Adams et al., 1995); aquaculture, agriculture, and horticulture (Rehakova et al., 2004); medical applications (Cerri et al., 2004); and water treatment (Wang and Peng, 2010). Recent studies have reported several types of zeolites with various ion exchange capacity and antimicrobial activity. Silver-, zinc-, and copper-exchanged natural zeolites have been investigated for their antibacterial activity (Top and Ülkü, 2004). Acrylic resin containing silver and zinc zeolites have an anticandidal effect against *Candida albicans* (Casemiro et al., 2008). In addition, insulated ducts containing silver zeolite installed in healthcare settings display a remarkable antifungal effect against *Aspergillus niger* (Tinteri et al., 2012).

The aim of this study was to synthesize silver- and zinc-ion–loaded antimicrobial zeolites (antibacterial, anticandidal, and antifungal) for the manufacture of durable antimicrobial composite surfaces using paints and polymers.

2. Materials and methods

2.1. Materials and reagents

The sodium aluminate, sodium hydroxide, and sodium metasilicate pentahydrate (Na_2O:SiO_2:$5H_2O$) and colloidal silica (Ludox:SiO_2:$5H_2O$) used for zeolite synthesis were obtained from Sigma–Aldrich. Steel plates, surface coating materials, and polypropylene (PP) surfaces were supplied by a leading appliance company (VESTEL; powder coating material, acrylic paint, and polyester paint). The silver nitrate ($AgNO_3$) and zinc chloride ($ZnCl_2$) used for the ion-exchange process were obtained from Sigma–Aldrich. The potato dextrose agar (PDA), Sabouraud dextrose agar (SDA), tryptic soy agar (TSA), Sabouraud dextrose broth (SDB), and tryptic soy broth (TSB) used in antimicrobial activity tests were purchased from Merck (Darmstadt, Germany). A 6-branch manifold filtration system and incubator shaking cabinet (CERTOMAT BS-T; Sartorius, Germany) were used during the zeolite synthesis and ion exchange processes.

2.2. Zeolite synthesis

During the study, 2 types of zeolites with different Si:Al ratios were synthesized as described previously by our group (Demirci et al., 2014). Synthesis gel formulas of the zeolites are listed in Table 1. Sodium metasilicate pentahydrate (SMS) and Ludox were used as the silica source, sodium aluminate was used as the aluminum source, and sodium hydroxide was the source of the balancing cation.

Chemicals were weighed and placed into polyethylene Erlenmeyer flasks. The required amounts of SMS or Ludox were put into Erlenmeyer flasks along with the required amount of water. Hydrothermal synthesis of zeolites took place in an oven at 90 °C for 3 days. At the end of the crystallization period zeolites were filtered using vacuum filtration and placed into an oven for 24 h at 90 °C (Hagiwara et al., 1988). Dried zeolite samples were ground using a mortar and pestle.

2.3. Ion-exchange processes of zeolites

Zeolite (80 g/L) samples were mixed with 1 M silver nitrate ($AgNO_3$) and 1 M zinc chloride ($ZnCl_2$) solutions individually. Mixtures were shaken at 200 rpm for 3 days in dark medium at room temperature. At the end of the incubation period, zeolites were filtered by vacuum filtration and put into an oven at 90 °C for 24 h. Dried zeolites were ground using a mortar and pestle.

2.4. Modified disk diffusion assay

The standard NCCLS disk diffusion assay (Lalitha, 2005) was modified and used to assess antimicrobial activity against each microorganism tested. Briefly 100 µL of suspensions containing 10^8 colony-forming unit (CFU)/mL bacteria, 10^6 CFU/mL yeast, and 10^4 spore/mL fungi were prepared from freshly grown cultures and spread on TSA, SDA, and PDA, respectively. The blank disks (6 mm in diameter) were wetted with 20 µL of sterile distilled water and impregnated with approximately 40 mg of metal-ion–loaded zeolite samples. Disks carrying zeolites were placed on inoculated plates. Sterile, distilled-water–impregnated blank disks were used as negative controls. Ofloxacin (5 µg/disk) and nystatin (100 U/disk) were used as positive controls for bacteria and fungi, respectively.

Table 1. Zeolite gel formulations.

Sample name	Synthesis gel formula	Silica source	Type of zeolite
Zeolite X	$4.64\ Na_2O$:Al_2O_3:$3.2\ SiO_2$:$400\ H_2O$	SMS	Faujasite X (FAU X)
Zeolite A	$2\ Na_2O$:Al_2O_3:$1.6\ SiO_2$:$200\ H_2O$	Ludox	Linde type A (LTA)

The inoculated plates were incubated for 24 h at 36 ± 1 °C for bacterial strains and 48 h for yeast strains and 72 h at 27 ± 1 °C for fungal species. Antimicrobial activity in the modified disk diffusion assay was evaluated by measuring the zone of inhibition against test microorganisms (Kalaycı et al., 2013). Each test was repeated at least twice.

2.5. Preparation of antimicrobial composites

Stainless steel plates (5 × 5 cm) were coated with a powder coating material used in the household appliance industry and polyester paint. The PP plates were painted with acrylic paint. Briefly, commercially-pure powder coating material was mixed with different concentrations of zeolite samples (7%, 10%, and 12% w/w). Then 100 mg of mixture was poured onto a metal plate and oven-dried at 120 °C for 1 h. In addition, polyester paints were mixed with zeolite samples (7%, 10%, and 12% w/w). Finally, different concentrations of zeolite samples (10%, 12%, and 15% w/w) were mixed with acrylic paint. Type X silver zeolites were used for the manufacture of antimicrobial PP surfaces. Zeolite samples [7% and 10% (w/w) ratio] were mixed with melted PP bulk until homogeneity was achieved in an extruder (200–220 °C).

2.6. Antimicrobial activity tests of prepared antimicrobial surfaces

The antimicrobial activity of the surface-modified samples was investigated for selected microorganisms (Table 2). Antimicrobial composite specimens painted with mixtures containing metal-ion–loaded zeolite samples were placed into sterile petri dishes. The modified surface of the sample was placed facing upward, and 1 mL of TSB, PDA, or SDB was poured onto the surface for bacteria, candida, and fungi respectively. Then 100 µL of suspensions containing 10^6 CFU/mL bacteria, 10^4 CFU/mL yeast, and 10^3 spore/mL fungi were added to the medium on the surface. The petri dishes were capped to prevent medium evaporation and incubated for 24 h at 36 ± 1 °C for bacterial strains and 48 h for yeast strains and for 7 days at 27 ± 1 °C for fungal species. Stainless steel and PP plates painted with commercial paints were used as negative controls. After incubation, a 100-µL sample was transferred into TSB, SDB, and PDB and serially diluted. From each dilution a 100-µL sample was plated on TSA, SDA, and PDA and cultured at the appropriate temperature to detect bacterial, yeast, and fungal growth, respectively. Surface-modified specimens were re-inoculated with selected microorganisms (bacteria, yeast, and fungi) at 15-day intervals for 1 year.

3. Results and discussion

Biocidal activities of the zeolite samples tested based on disk diffusion assay revealed that pure zeolites X and A did not have any antimicrobial activities, whereas both silver- and zinc-ion–exchanged zeolites exhibited remarkable inhibition zones around the samples for all tested microorganisms. The diameters of the inhibition zones are given in Table 2. The metal-ion–exchanged zeolites display variable antimicrobial activity against all microorganisms tested in the current study. Similar results have been reported in previous studies showing that Ag^+- and Zn^{+2}-ion–exchanged zeolites had inhibitory effects on several microbial species including bacteria, yeast, and fungi (Top and Ülkü, 2004; Kwakye-Awuah et al., 2008; Egger et al., 2009).

In the current study, composite materials containing various concentrations of silver and zinc zeolites are

Table 2. Antimicrobial effect of silver- (Ag^+) and zinc- (Zn^{2+}) loaded zeolites for microorganisms based on inhibition zones in disk diffusion assay.

	AgX	ZnX	AgA	ZnA	PC	NC
Escherichia coli	11 ± 2[a]	13 ± 2	12 ± 2	11 ± 2	28	0
Pseudomonas aeruginosa	12 ± 2	7 ± 1	12 ± 3	7 ± 1	24	0
Staphylococcus aureus	13 ± 1	14 ± 2	13 ± 2	14 ± 1	20	0
Candida albicans	33 ± 5	19 ± 2	29 ± 4	16 ± 1	20	0
Candida glabrata	35 ± 4	16 ± 3	34 ± 4	20 ± 2	18	0
Aspergillus niger	7 ± 2	36 ± 2	8 ± 1	36 ± 4	20	0
Penicillium expansum	12 ± 2	18 ± 1	17 ± 2	16 ± 2	18	0

AgX: silver zeolite type X; ZnX: zinc zeolite type X; AgA: silver zeolite type A; ZnA: zinc zeolite type A; PC: ofloxacin (5 µg/disk) and nystatin (100 U/disk) for bacteria, candida, and fungi, respectively; NC: pure zeolite type A and X samples.

[a] All values are expressed in millimeters.

examined for their antimicrobial activity and stability against the microorganisms tested. Holtz et al. (2012) added nanostructured silver vanadate to water-based paints and found it effective against methicillin-resistant *S. aureus* (MRSA), *Enterococcus faecalis*, *E. coli*, and *Salmonella enterica* (Holtz et al., 2012). Another study indicated that stainless steel surfaces coated with paints containing a silver and zinc zeolite showed profound antimicrobial activity against *Bacillus* spp. (Galeano et al., 2003). In addition, Zielecka et al. reported that the addition of copper and silver nanoparticles to architectural paint provided antimicrobial properties against fungi including *A. niger*, *Paecilomyces variotii*, *P. funiculosum*, *Chaetomium globosum* mixture, and the bacterial strain *P. aeruginosa* (Zielecka et al., 2011). However, little is known about the stability and durability of antimicrobial materials or their antimicrobial efficacy against a wide range of microorganisms including bacteria, yeast, and fungi.

In the present work, different surfaces coated with various paints containing silver- and zinc-ion–exchanged zeolites were evaluated for their antimicrobial activity. Zinc-ion–exchanged zeolite compositions were used for white powder coating materials and polyester paints; there were no noticeable changes in the color of composite materials. In addition, silver colored powder coating material, polyester paint, and acrylic paints were mixed with silver-ion–exchanged zeolites, and there were no noticeable change in these surfaces either. On the other hand, for the PP surfaces there was a remarkable change in surfaces containing silver zeolite. The color of PP surfaces changed to a brownish color after 7% and 10% silver-zeolite additions.

The stability of all surfaces coated with different paints, including various amounts of silver and zinc zeolites (7%–15%), is given in Table 3. Silver- and zinc-ion– embedded zeolite X (AgX and ZnX) exhibited longer antimicrobial activity in comparison to silver- or zinc-ion–embedded zeolite A (AgA or ZnA). This result could be a consequence of the high and sudden release rate of ions in the case of zeolite A (Weitkamp, 2000). Zeolite A releases loosely bound metal ions faster than zeolite X so that the remaining metal content may not be sufficient to inhibit microbial growth.

The inhibitory effect of steel plates treated with commercial and antimicrobial powder coating containing silver- and zinc-ion–exchanged zeolites is shown in Figure 1 and Table 3. According to the results, the addition of 7% silver-zeolite (w/w) to the powder coating was sufficient to provide antimicrobial activity against the tested strains for 45 days. Moreover, AgX (10%) and ZnA (10%) were found to be free of *A. niger* and *P. expansum* contamination up until day 120 of inoculation.

Polypropylene surfaces including 7% and 10% AgX, prepared by extrusion and thermoforming, displayed the shortest duration of antimicrobial activity among surfaces, as expected. Figure 2 showed *P. expansum* and *A. niger* growth on a pure PP surface and the inhibitory effect of AgX-PP composite against the fungal isolates. Although silver-zeolite–enhanced PP surfaces exhibited antimicrobial efficiency for at least 30 days, their maximum efficiency period was 60 days. This may be due to the rigid structure of the PP material. Metal ions bound to zeolite structures could not be released fast enough to inhibit microbial growth for longer periods on PP surfaces. Therefore, antimicrobial activity on the PP surfaces was relatively short-term in comparison to coated materials. Our data supports the findings of previous studies that reported that silver-, copper-, and zinc-ion–exchanged zeolite/polyurethane composites have antimicrobial effects against *E. coli,* methicillin-resistant *S. aureus*, *P. aeruginosa*, and *C. tropicalis* (Kamışoğlu et al., 2008; Kaali et al., 2011).

In the case of PP surfaces coated with acrylic paint containing ion-exchanged zeolites, microbial growth inhibition was observed for more than 200 days (Figure 3; Table 3). Durability of antimicrobial efficiency was found to be directly proportional to the concentration of zeolite samples in material surfaces. These results are consistent with those reported previously (Kaali et al., 2010). As the concentration of Ag zeolites increased, the stability of antimicrobial activity progressed in parallel.

In this study different concentrations of zeolite samples were selected for various materials. These concentrations can be altered according to the desired durability of the antimicrobial effect. However, using a concentration of silver-ion–exchanged zeolite below 7% is not appropriate as there is no significant antimicrobial effect below that point. Silver and zinc ions are released at a controlled rate and ensure long-term antimicrobial protection on surfaces. However, silver-ion–exchanged zeolites exhibited greater inhibitory effect against bacteria than zinc-ion–exchanged zeolites on all surfaces. These results may be explained by the low antibacterial effect of Zn zeolites as reported previously (Kim et al., 1998). On the other hand, Zn^{2+}-loaded zeolites displayed better antifungal effects than Ag^{+}-loaded zeolites in general. Similar findings have been reported in previous studies (Malachová et al., 2011; Demirci et al., 2014). As zinc, but not silver, is an essential element for fungal species and necessary for fungal metabolism, transportation of zinc into the cytoplasm is easier than transport of the silver ions (Baldrian, 2010). Although zinc is necessary for the fungal system, it can display biocidal activity at high concentrations. Therefore, zinc ions released from zeolite samples may have accumulated in fungal cells, resulting in greater fungicidal activity, with respect to silver ions.

Table 3. Durability of antimicrobial effect on surfaces containing metal-ion–loaded zeolites.

	Powder coating											
	AgX (7%)	AgX (10%)	AgX (12%)	ZnX (7%)	ZnX (10%)	ZnX (12%)	AgA (7%)	AgA (10%)	AgA (12%)	ZnA (7%)	ZnA (10%)	ZnA (12%)
Escherichia coli	75>[a]	105>	105>	45>	75>	105>	45>	75>	90>	45>	60>	90>
Pseudomonas aeruginosa	90>	105>	135>	60>	90>	105>	60>	90>	105>	60>	75>	105>
Staphylococcus aureus	45>	75>	105>	30>	60>	90>	45>	75>	90>	30>	60>	75>
Candida albicans	90>	105>	150>	75>	90>	120>	75>	90>	120>	75>	75>	105>
Candida glabrata	75>	105>	135>	45>	60>	105>	75>	90>	135>	60>	60>	105>
Aspergillus niger	90>	135>	180>	105>	150>	255>	75>	105>	135>	90>	135>	180>
Penicillium expansum	75>	120>	165>	75>	135>	215>	60>	105>	135>	75>	135>	180>
	Polyester paint											
	AgX (7%)	AgX (10%)	AgX (12%)	ZnX (7%)	ZnX (10%)	ZnX (12%)	AgA (7%)	AgA (10%)	AgA (12%)	ZnA (7%)	ZnA (10%)	ZnA (12%)
Escherichia coli	45>	75>	90>	30>	60>	75>	45>	60>	90>	30>	45>	75>
Pseudomonas aeruginosa	60>	90>	105>	45>	60>	90>	45>	75>	105>	30>	60>	75>
Staphylococcus aureus	60>	75>	105>	60>	60>	90>	60>	75>	90>	45>	60>	90>
Candida albicans	75>	90>	120>	60>	90>	90>	60>	90>	105>	60>	75>	90>
Candida glabrata	75>	90>	105>	60>	75>	90>	75>	90>	105>	60>	60>	75>
Aspergillus niger	45>	75>	105>	60>	75>	120>	45>	75>	90>	60>	75>	105>
Penicillium expansum	45>	90>	105>	60>	90>	105>	30>	75>	90>	45>	75>	90>
	Acrylic paint						Polypropylene surface					
	AgX (10%)	AgX (12%)	AgX (15%)	AgA (10%)	AgA (12%)	AgA (15%)	AgX (7%)	AgX (10%)				
Escherichia coli	60>	90>	225>	45>	75>	225>	30>	30>				
Pseudomonas aeruginosa	105>	120>	210>	75>	75>	210>	45>	45>				
Staphylococcus aureus	105>	105>	225>	45>	90>	195>	30>	45>				
Candida albicans	150>	165>	285>	60>	75>	240>	45>	60>				
Candida glabrata	150>	150>	270>	60>	75>	240>	45>	45>				
Aspergillus niger	135>	240>	360<	45>	60>	285>	30>	45>				
Penicillium expansum	45>	175>	360<	45>	135>	360<	30>	45>				

AgX: silver zeolite type X; ZnX: zinc zeolite type X; AgA: silver zeolite type A; ZnA: zinc zeolite type A.

[a] All values are expressed in days.

>: The day of microbial growth detection.

<: Existence of antimicrobial effect more than 360 days.

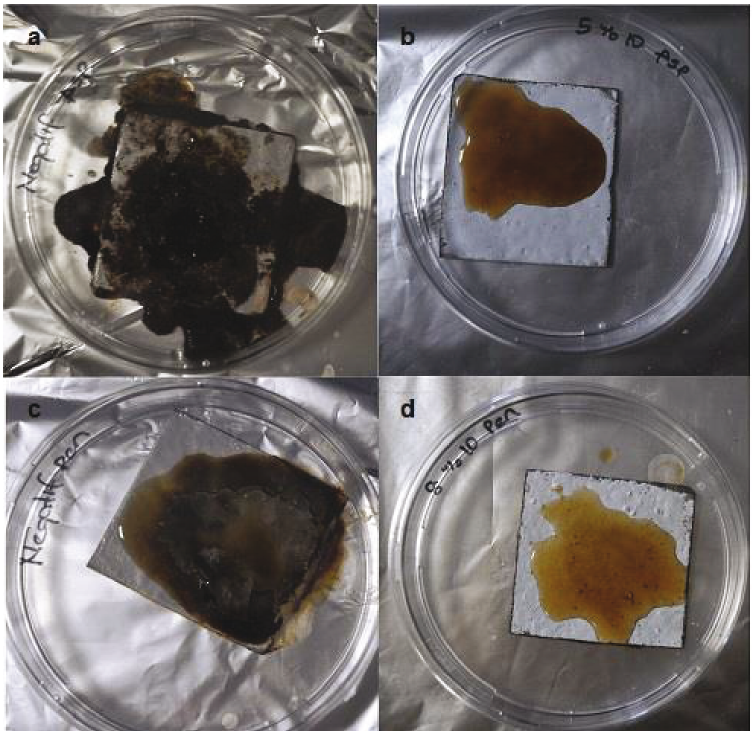

Figure 1. Zinc zeolite samples prevent microbial growth on powder-coated surfaces. Day 90 of *A. niger* contamination on a) commercial powder coating material and b) zinc-zeolite type-X –containing modified powder coating mixture coated surfaces (10% w/w). Day 90 of *P. expansum* contamination on c) commercial powder coating material and d) zinc-zeolite type-A–containing modified powder coating mixture coated surfaces (10% w/w).

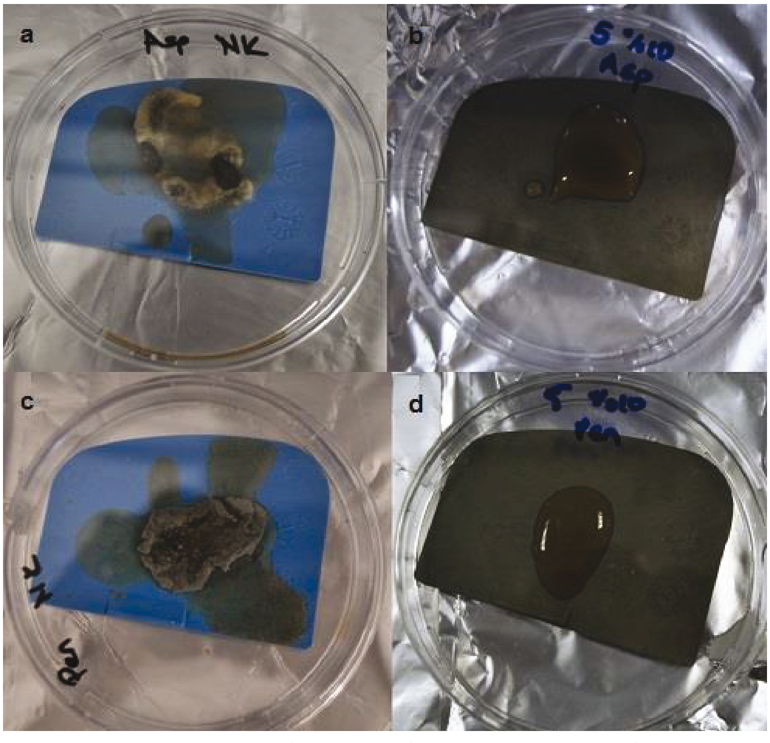

Figure 2. Silver zeolite containing polypropylene (PP) surfaces inhibit fungal growth. Day 30 of *A. niger* contamination on a) commercial and b) silver-zeolite type-X –containing PP surfaces (10% w/w). Day 30 of *P. expansum* contamination on c) commercial and d) silver-zeolite type-X–containing PP surfaces (10% w/w).

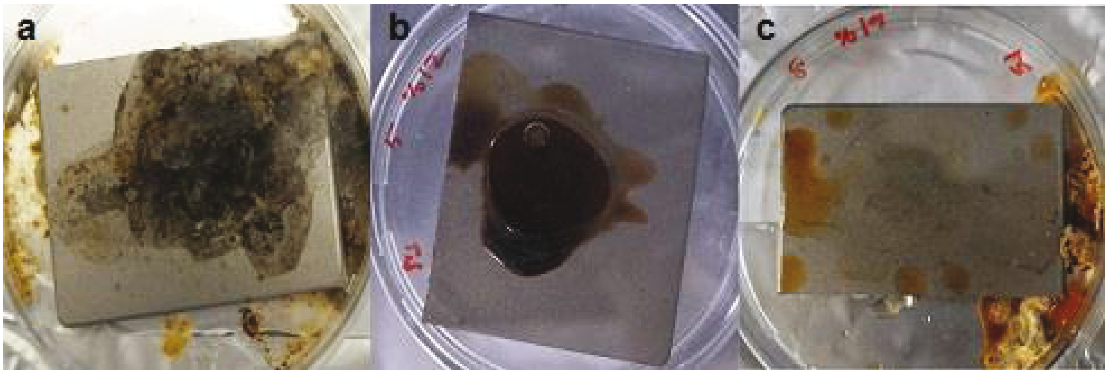

Figure 3. Day 225 of *A. niger* contamination on a) commercial and b) silver-zeolite type-X–containing (12% w/w) and c) silver-zeolite type-A–containing (15% w/w) acrylic paint coated PP surfaces.

4. Conclusion

Silver- and zinc-ion–exchanged zeolite formulations exhibit inhibitory effects against bacteria (*E. coli*, *P. aeruginosa*, and *S. aureus*), yeast (*C. albicans* and *C. glabrata*), and fungi (*A. niger* and *P. expansum*). Although Ag zeolites displayed significant antibacterial and anticandidal properties, Zn zeolites exhibited comparable antifungal characteristics. The current study indicates that tailoring ion-exchanged zeolite formulations for the desired antimicrobial efficacy and durability of the resulting composite is a promising approach for the development of novel antimicrobial materials. These materials would prevent microbial growth on their surfaces, reducing cross-contamination and infection risk as well as the microbial degradation of the products. Further studies are necessary to determine potential antimicrobial activity in cases involving binary ion exchange with silver and zinc ions acing together. In addition, investigations are needed to evaluate the antimicrobial efficacy of ion-exchanged zeolite formulations in other composite materials including plaster of paris, cement, ceramics, varnish, and grouting.

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