Effect of Cationic Polyacrylamide and Antibacterial Nanosilver on Bank-note Paper Properties

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Bank-notes have been an integral part of buying and selling for many years. There are about 7 billion Bank-note leaves in Iran that have been in circulation in a 5-year period. This amount of Bank-notes and the frequency with which they are touched can promote transmission of many pathogenic factors, especially bacteria. Based on scientific research results, Bank-notes are seriously contaminated and must be considered a potential danger to society. In this study, for antibacterial Bank-note paper production, 100 ppm of Nanosilver was used as an antibacterial agent accompanied by cationic polyacrylamide at the 0, 0.3, 0.5, 0.7, 1 percent (based on O.D. pulp) as a retention aid of Nanosilver particles on cotton fibers. Then standard handsheets (90 g/m^2) were made from the above-mentioned pulps and tested for physical and mechanical properties. Also, for antibacterial tests of handsheets, *Escherichia coli* and *Staphylococcus aureus* were used as Gram-negative and Gram-positive bacteria, respectively. The results showed that handsheets strengths decreased with increasing of Nanosilver absorption. But antibacterial properties of handsheets increased by increasing of Nanosilver absorption so that the most antibacterial properties for handsheets were achieved at the 1% level of retention aid addition.

Keywords: Cationic polyacrylamide; Nanosilver; Antibacterial paper; Escherichia coli; Staphylococcus bacteria

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INTRODUCTION

Bank-notes have been a part of commercial exchanges from the long past. Perhaps no other object has been in such frequent daily use within societies. But there has been an unfortunate lack of proper maintenance of Bank-notes, which are a likely cause of many hygienic problems (Pinner and Teutsch 1996). Studies concerned with the recognition of pathogenic factors affecting Bank-notes have shown that there are Gram-positive *Staphylococcus aureus* and Gram-negative *Escherichia coli* on 13% of coins and 42% of Bank-notes in the US (Abrams and Waterman 1972). Also, investigations in China and Germany have shown that most microbes can remain viable on Bank-notes for long periods and feed on the fatty materials that have accumulated on them (Zhang 1984). On the other hand, studies of Bank-note contamination in Egypt have shown that

65% of the Bank-notes were contaminated with intestinal bacteria (Dave 2005). *Escherichia coli* bacteria belong to a big and various family of bacteria, some species of which cause diseases such as diarrhea, urinary channel infections, and respiratory conditions. This type of bacteria is one of the important factor of traveler's diarrhea. It has been reported that *Escherichia coli* bacteria was observed on 60% of Bank-notes in butcheries, 10% in poultry shops, and 5% in fish food shops and confectioneries (Zarei *et al*. 2008). The other pathologic factor is the *Staphylococcus aureus* family. The toxin of this bacteria causes vertigo, diarrhea, vomiting, skin infection, marrow infection, and blood infection (Leyinson *et al.* 2000). It was further reported that 55% of Bank-notes in butcheries, 40% in ice-cream shops, 30% in bakeries, 25% in fast food establishments, and 10% in confectioneries were polluted with *Staphylococcus aureus* bacteria (Zarei *et al.* 2008). Due to pathogenic factors resistant against medications and antibiotics, it is difficult to effectively inhibit infectious diseases. Development of tolerance by bacteria against the available antibiotics is a serious problem affecting the general health. In his regard, nanotechnology opens new ways to fight and prevent diseases by confronting the problem at an atomic scale (Kermanshahi 2008).

Among the best nanomaterials with antimicrobial properties, nano-metals that show high chemical activities by surface crystallographic structure are especially significant. Among these materials are products incorporating of silver ions and silver-based compounds. Nanosilver particles have antibacterial, antifungal, and antiviral properties, and antibacterial surfaces can be created by adding small amounts of this material; even a small quantity equates to many particles per unit area (Kermanshahi 2008). It has been found that a Nanosilver suspension was able to eliminate many of the recognized bacteria and even HIV viruses (Csreen *et al.* 2007). Nanosilver particles elliminate bacteria by closing up respiratory tracts and by affecting the metabolism and reproduction of the microorganism (Kelasen 2000).

An investigation was performed on two materials, chitosan and Nanosilver particles to produce filter paper (Imani *et al.* 2011). Nanosilver particles were applied as negatively charged layers over the fibers (LBL). It was found that with increasing numbers of the layers, there were increases antibacterial properties, so that bacteria growth at eight layers of the mentioned materials, as measured by a turbidity test, reached to zero. Also, the antibacterial property of papers was assessed against five types of bacteria. The effects of different antibacterial materials were different. Chitosan showed a better effect on *Basibs Sobtilitis* bacteria. By contrast, Nanosilver particles were more efficacious against *Staphylococcus aureus* and *Escherichia coli* (Imani *et al.* 2011). In another investigation to achieve better performance of silver particles, CFGI compound was produced on fibers pretreated with the chelating monomer glycidyl methacrylate-iminodi acetic acid (Gma-ida), and this treatment caused more bonding of Nanosilver particles to the fibers. Antibacterial performance was improved. With increasing concentration of Gma-ida solution, more silver was absorbed to fibers. Nanosilver with a particle size of 75 nm was said to have better performance (Chen and Chiang 2008). Also, Nanosilver particles with size of 25 nm and concentration of 25 and 50 ppm on the basis of ethanol were used for making antibacterial cloth fibers that were added to cloth before and after dyeing. When cloth was examined with an antibacterial test after dyeing,

it showed more efficacy. The suitable concentration of Nanosilver suspension to cloth treatment was reported as 50 ppm (Lee and Yeo, 2003).

Despite the fact that most infectious diseases can be transferred by touching biotic objects such as Bank-notes (Pinner and Teutsch 1996), no research has been reported about making antibacterial Bank-note paper. So the present study may be unique in its consideration of Nanosilver particles due to antibacterial properties, non-toxicity, and environmental friendly character as an antibacterial agent in Bank-note paper production.

EXPERIMENTAL

Materials

Industrial cotton pulp was provided with a Schopper-Riegeler (^{o}SR) value of 51, a consistency of 3.1%, and a pH of 7.2. Also, Nanosilver (with 4000 ppm concentration and 50 nano-meter particle size made by Rangdane Sharif company) and cationic polyacrylamide (with medium cationic charge with Farinret K325 trademark made by Degussa Co.) were used as antibacterial agent and retention aid, respectively. For antibacterial test of papers, the two bacteria including Gram-negative *Escherichia coli* ATCC: 25922 and Gram-positive *Staphylococcus aureus*, ATCC, which were provided by Dr. Ghahari of the medical diagnosis laboratory in Babolsar city, were used. Brain Heart Infusion Broth (BHIB) and Nutrient agar both made by Biolife company were used as liquid and solid culture medium, respectively.

Methods

The pulp consistency was decreased to 0.3% and stirred to homogenize it for 15 minutes. Cationic polyacrylamide (CPAM) with 1% concentration was added at the levels of 0, 0.3, 0.5, 0.7, and 1%, and then a constant level of 100 ppm nano silver was added to the pulp. Standard handsheets were made according to ISO 5269/1 and tested to evaluate physical and mechanical properties (Table 1). Surface fibers in Bank-note paper must be have good bonding so that they will not separate from the paper when the metal surface bearing the tacky ink is separated from it during printing. Hence, a wax test was carried out, in which the end part of a series of wax pieces is softened over a flame and put over the paper. The wax pieces are separated from the paper surface rapidly after 15 minutes. So the most highest number of wax (representing higher strength) that doesn't result in damage to the paper is accepted as wax test.

Table 1. Tests and Standards

Test	Paper basis weight	Tear strength	Folding strength	Smooth-ness	Breaking length	Ash content
Unit	gr/m²	mN	-	%	m	%
Test method	ISO 536:1976	ISO 1974	ISO 5626	ISO 5627	ISO 2493	ISO 2144:1997

Antibacterial properties of handsheets were also assessed with a turbidity test, using a Spectrophotometer 6300. Culture medium (BHIB) was placed in an autoclave (15 min at 121°C), and treated and untreated handsheets were sterilized in an oven (0.5 g, 2 h and 150°C) were sterilized. Sterilized papers were inserted into a tube containing sterilized culture; then 0.05 mL with 0.05% concentration of *Escherichia coli* and *Staphylococcus aureus* were added separately and put into a shaker incubator (160 rpm and 37°C) over one night. Controls were also run in parallel. All experiments were conducted in triplicate. Also, some pieces of paper were exposed on the solid cultures for evaluation of bacterial growth on papers by optical microscope.

Statistical Analysis

All results were analyzed by analysis of variance, and the Duncan test was used to compare of averages. SPSS software was used for all analyses.

RESULTS AND DISCUSSION

Ash Content

As can be seen in Fig. 1, ash content was increased with increasing CPAM level at a 95% confidence level. This can be due to amine groups on cationic polyacrylamide that join well to fiber, leading to some fine flocks of the fibers and also trapping and retaining the Nanosilver particles. This finding confirmed the results of Mokhtari (2011).

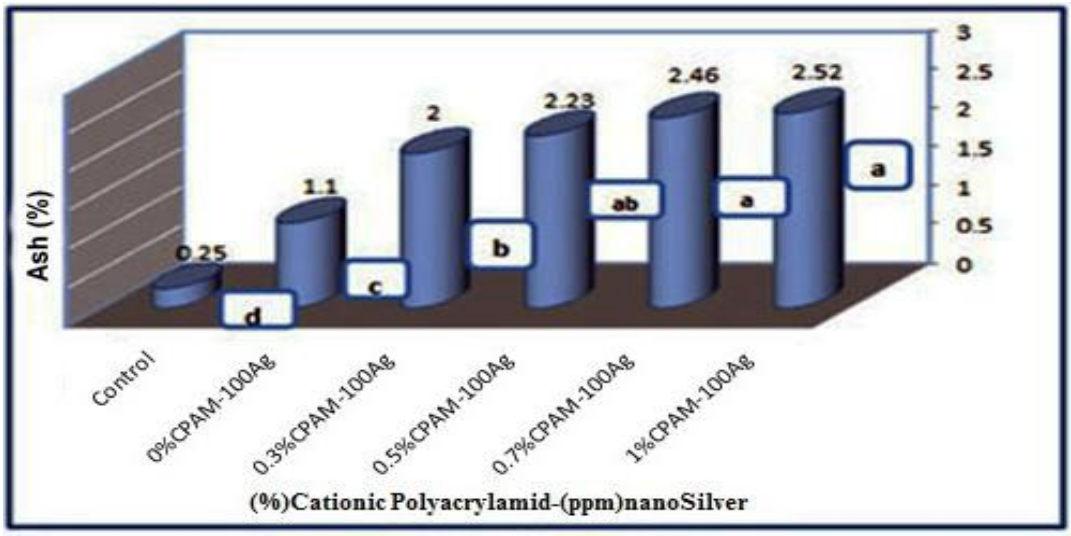

Fig. 1. Effect of CPAM and nano silver on ash content of Bank-note paper

Bulk Density

Effects of CPAM and Nanosilver addition on the bulk density of handsheets are shown in Fig. 2. It is clear that by increasing the CPAM level, the bulk density was decreased. This is probably due to fines retention between fibers. When dry strength agents such as polyacrylamide adheres to fibers, it forms bridges between fibers and increases bonding among the fibers (Scott 2006). The polyacrylamide, due to its cationic

charge, adsorbs fines and Nanosilver. Filling of empty space between fibers with Nanosilver may help account for the observed decline in caliper. Also, cationic polyacrylamide causes hydrogen bonding between fibers and prevents fines from being removed.

Due to Interactions of CPAM and Nanosilver, fines absorb more and by forming of hard and tiny flocks, caliper and subsequently bulk density of paper decrease. These results also confirmed the finding of Ebrahimi (2011).

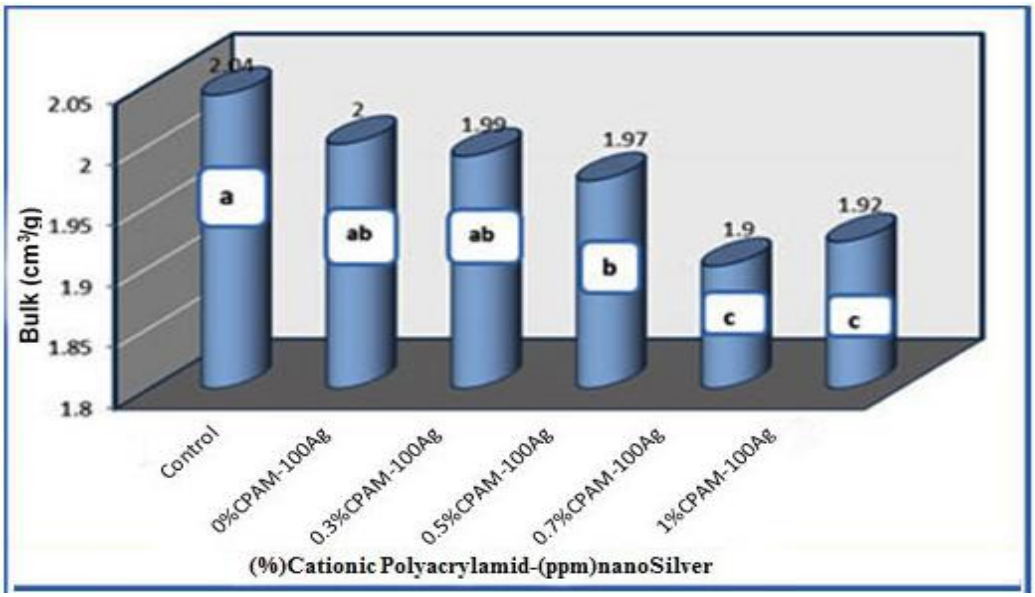

Fig. 2. Effect of CPAM and nano silver on bulk of Bank-note paper

Smoothness

Smoothness of paper surface is an important property for printed paper such as Bank-notes, which need more smoothness compared to other grades of paper (Scott 2006). As Bank-note paper is printed on both sides, there should be little difference between the two sides.

Figure 3 shows that by adding CPAM and Nanosilver particles, smoothness of the paper was increased compared to the control. It is worth noting that by adding of CPAM, smoothness appeared to be decreased, but the apparent effect was not statistically significant. Also, the difference between the two sides of paper is decreased by adding CPAM level that it might be related to more absorption of fines (Miyanishi, 1998). Smoothness difference between two sides of paper is decreased with an increasing addition of retention aid.

Wax Test for the Surface Strength of Paper

Surface fibers should be firmly bonded to the sheet when detaching steel printing plates and offset blankets from the surface, not separated or removed. One of the most common methods of determining of paper strength in the Z-direction is the wax test. The wax standards are numbered from 2A to 26A based on their adhesion power. In this study, the wax index was 10A for all samples and without meaningful difference between all by increasing in CPAM and Nanosilver levels(Fig. 4).

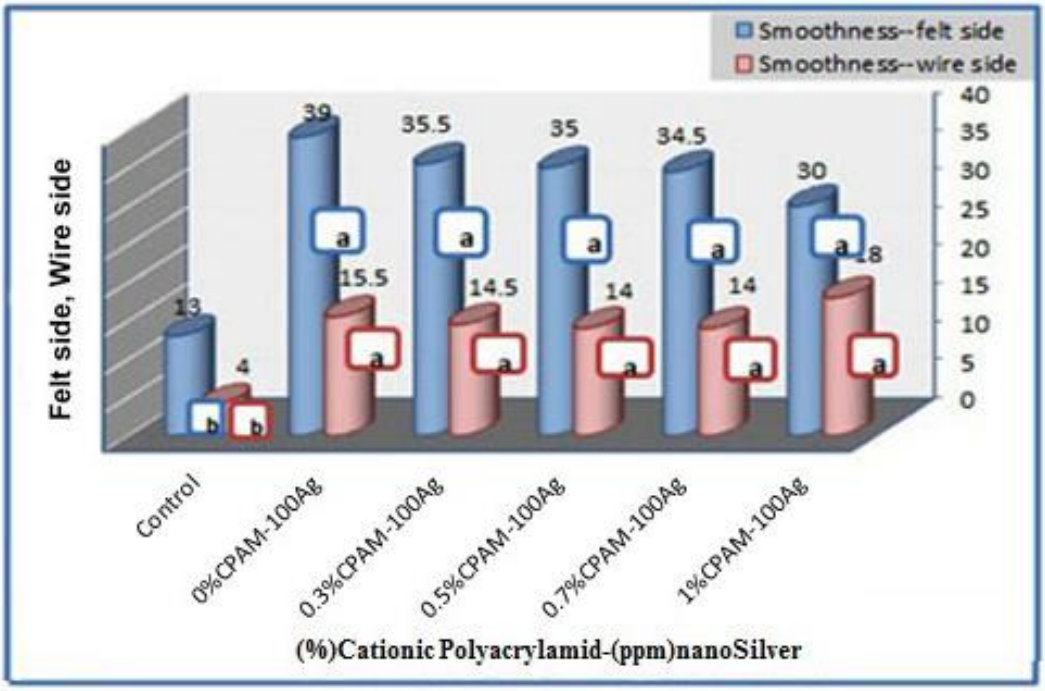

Fig. 3. Effect of CPAM and nano silver on smoothness of Bank-note paper

In evaluation of raw material efficacy in bank note paper production, Erfani (2011) reported a wax index of 16A. Due to use of a trimming refiner and also surface sizing in the final step of paper machine in Bank-note production, a value of 10A could be accepted for surface strength of handsheets.

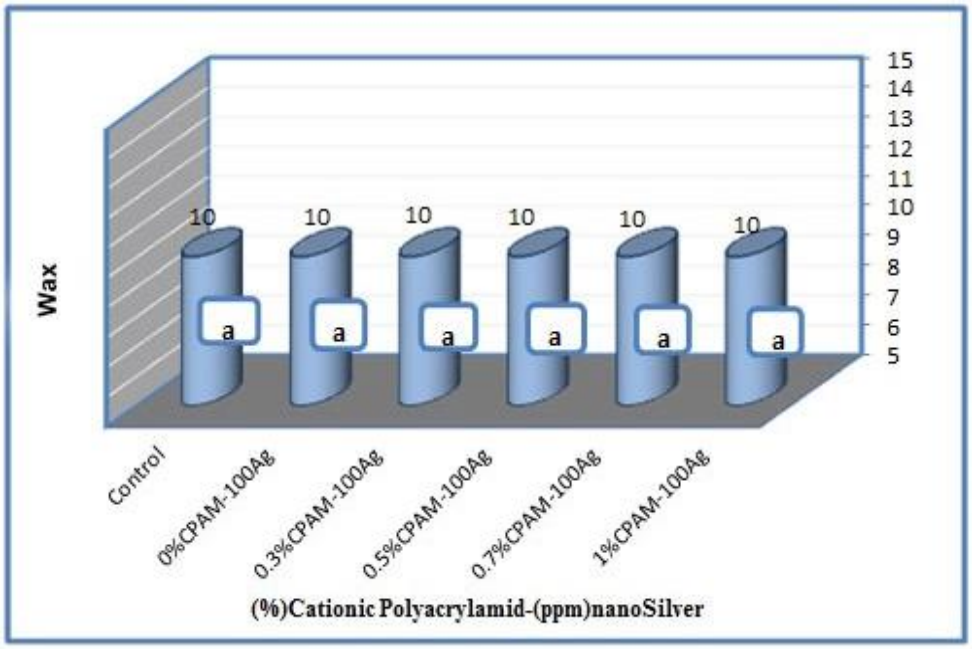

Fig. 4. Effect of CPAM and nano silver on wax index of Bank-note paper

Breaking Length

The most important factors affecting the breaking length of paper are the number and quality of bandings between fibers (Testova 2006). Tensile strength directly shows a key aspect of the durability and final functionality of many products such as Bank-note .

According to the Fig. 5, the breaking length decreased with the increase of CPAM level. Reduction of breaking length due to retention aids addition can be attributed, at least in part, to more absorption of nano silver and also increasing porosity or flocculated character of Bank-note paper (Roberts *et al* 1986). By increasing of bonding involving the fiber-CPAM-silver combination, fiber-fiber bonding decreased, resulting in a considerable decline in breaking length. Also, weak formation and flock formation decreases the breaking length. As tensile strength decreased even without retention aids, much of the decrease can be attributed to Nanosilver as a major factor (Imani 2011).

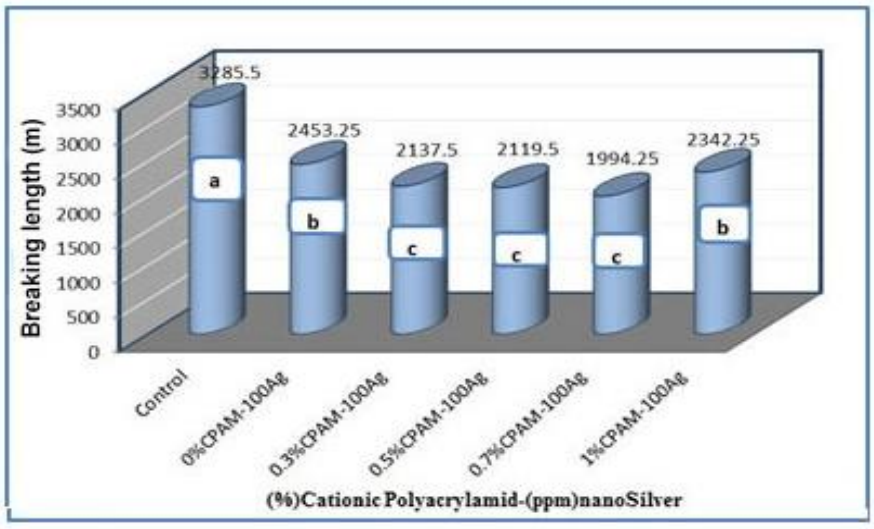

Fig. 5. Effect of CPAM and nano silver on breaking length of Bank-note paper

Tear Strength

Tear index depends on fiber length, individual fiber strength, and Relative Bonding Area (RBA). In the case of a sheet having low RBA, tear strength is more dependent on fiber-fiber bondings, whereas by increasing of bonding, individual fiber strength becomes crucial in tear index (Testova 2006). Tear index is one of the paper quality parameter that affects the runability of a paper machine.

As can be seen in Fig. 6, tear index was decreased with increasing consumption of retention aid. This can be related to increased retention of fines as well as decreasing in average fiber length. On the other hand, the presence of Nanosilver between fibers will tend to reduce fiber bonding, leading to a decrease in tear strength. These results confirmed Ebrahimi's report (2011), which indicated that by increasing filler retention on CMP fiber, tear strength was reduced.

Folding Endurance

This test involves a combination of tensile strength, elongation, flexibility, compatibility, shear stresses, and relative fiber length change (Erfani 2011). According to

Fig. 7, by adding CPAM to the furnish, along with the addition of Nanosilver, the folding endurance declined. This probably can be attributed to a lack of hydrogen bonding between cotton fibers. Addition of filler in paper results in a weakening of the fiber network, lack of long fiber in pulp suspension, and a higher level of fines in the paper; such changes are consistent with a decrease in folding endurance (Erfani 2011). As can be seen in Fig. 7, folding strength of handsheets was very low and there was considerable difference between the values obtained in the present work compared to the common level of Bank-note paper folds (about 5000). This difference probably can be attributed to the properties of the pulp that was provided before the trimming refiner. A trimming refiner causes fiber fibrillation and increases hydrogen bonds in paper machine before papermaking.

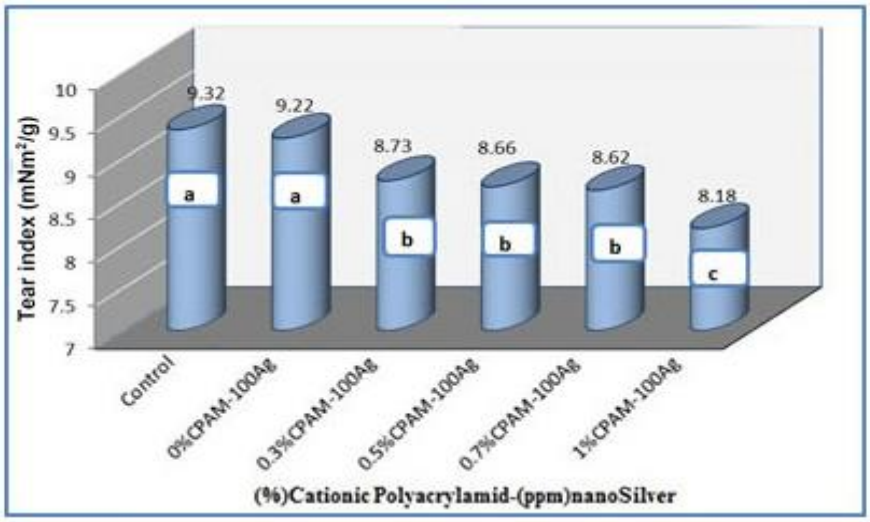

Fig. 6. Effect of CPAM and nano silver on tear index of Bank-note paper

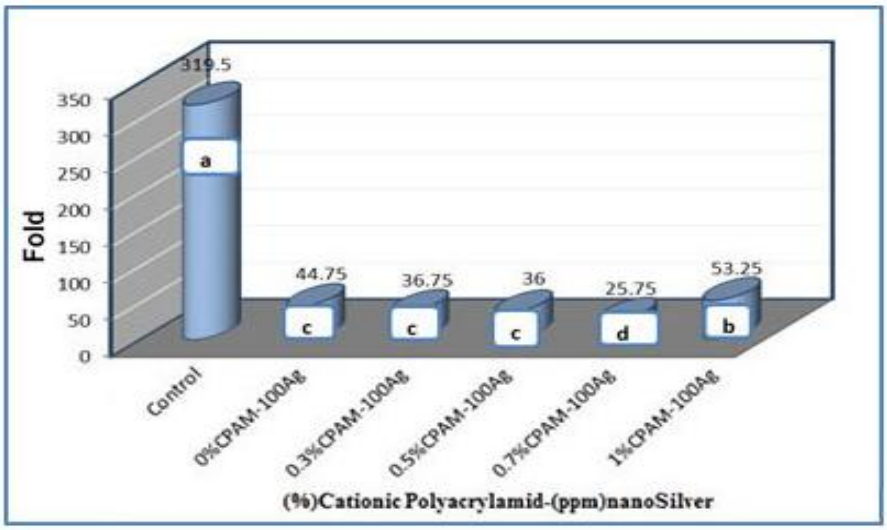

Fig. 7. Effect of CPAM and nano silver on folding strength of Bank-note paper

Biological Test of Bank-note paper

According to Fig. 8, for both types of bacteria *Staphylococcus aureus* and *Escherichia coli*, by increasing the amount of CPAM, the UV adsorption was reduced and the greatest reduction was obtained when using retention aid at the 1% level. This shows that higher adsorption of Nanosilver led to higher anti-microbial effects in handsheets. Also, turbidity in samples including *Staphylococcus aureus* was lower. As a rule, Gram-positive bacteria are more sensitive to Nanosilver particles than Gram-negative bacteria due to a difference in the cell walls. In Gram-positive bacteria, the cell wall is rich in mucopeptide component, while in Gram-negative ones the cell wall just contains a thin layer of mucopeptide and is primarily composed of lipoproteins and lipoprotein polysaccharides. Therefore, *Escherichia coli* shows more resistance to anti-bacterial materials (Tassou and Nychas1995). The present findings confirmed the other results reported by Sondi *et al.* (2004) and Giange *et al.* (2004). It is clear in Fig. 8 that treated paper showed higher resistance to bacteria than the reference samples.

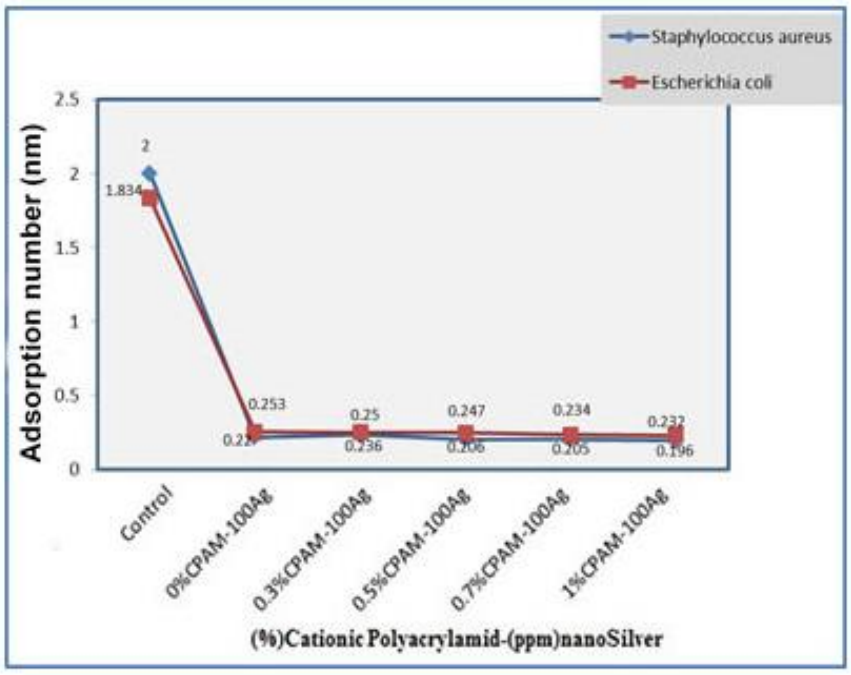

Fig. 8. Effect of CPAM and nano silver in Bank-note paper on turbidity of bacterial medium

Microscopic Investigation

Figure 9 shows the optical microscope images of papers on the solid culture in the vicinity of bacteria. As can be seen, *Esherichia coli* was able to grow more abundantly and closer to paper due to greater resistance to Nanosilver than *Staphylococcus aureus.* Therefore, the inhibition zone in *S. aureus* was larger. These images confirm the results of the previous section. The present findings are consistent with the results of Haji Mirza Baba *et al.* (2011). The results provide evidence of the reaction mechanism of Nanosilver with biological macromolecules, inhibition zone formation, and also the transformation of microorganism by conversion of SH bond of proteins that are present on the bacterial surfaces. These proteins give rise to a bulge in the outside of cell membranes and allow

passage of nutrients through the cell wall (Baumgartner and Cooper 1996; Sung Kaworn *et al.* 2007). In this mechanism, Nanosilver particles release silver ions over time. These ions convert SH bonds to SA-g bonds in the microorganism membrane during the substitution reaction. In this way, Nanosilver particles deactivate protein, reduce permeability of membrane, and finally cause the cell death (Tahan *et al*. 2006; Fengetal 2000; Stoimenovetal 2002).

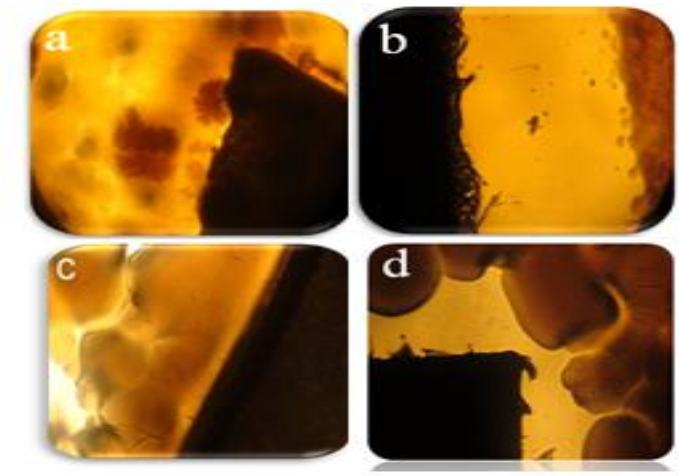

Fig. 9. Optical microscope image (magnification 4x) of treated paper with nano silver and control that tested with bacteria (a: control sample for *Staphylococcus aerus* growth; b: treated sample for *Staphylococcus aerus* growth; c: control sample for *Escherichia coli* growth; d: treated sample for *Escherichia coli* growth

CONCLUSIONS

At present, much attention is being given to the addition of nanoparticles to achieve essential interactions during the formation process of paper. Although usage of nanoparticles can provide a lot of advantages, it is very important to determine how best to use such products and how much of the nanoparticle is appropriate for the system. In fact, due to the change in pulp composition and even in the nanoparticles themselves (in terms of size and type), there are different interactions and changes in wet end chemistry. For this reason in spite of positive effect of nano particles, there is still a need for plenty of research in order to achieve better and different utilization of this system.

The present research was done to investigate the effect of poly-acrylamide and Nanosilver on physical and mechanical properties of paper pulp. Five levels of cationic poly-acryl-amide (0, 0.3, 0.5, 0.7, and 1%) were applied. Results showed that the best retention of Nanosilver as well as paper properties were obtained in at the 0.3% level of CPAM consumption.

Preservative properties of Nanosilver in recent decade have attracted much attention. In this research, Nanosilver used in paper pulp as an additive led to decreasing physical and mechanical properties of Bank-note paper, but in terms of biological resistance to bacteria, it was very effective. Anti-bacterial properties of paper were increased by applying of 1% Nanosilver. By adding of low levels of Nanosilver in pulp

suspension and then applying supplemental refining of the fiber furnish, both anti-bacterial and mechanical properties of Bank-note paper likely will be increased.

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