Sayeda M. Ibrahim

Department of Radiation Chemistry, National Center for Radiation Research and Technology,
P.O. Box 29, Nasr City, Cairo, Egypt
E-mail: sayda.ibrahim@yahoo.com

Removal of Copper and Chromium Ions from Aqueous Solutions Using Hydrophilic Finished Textile Fabrics

Abstract
Modified textile fabrics were used to remove Cu^{+2} and Cr^{+3} ions from aqueous solutions. For this purpose, modified Nylon-6, polyester woven and knitted fabrics were prepared by means of coating their surface with a layer of aqueous solution of Carboxymethyl Cellulose (CMC) and Acrylic Acid (AAc) of 25 mm constant thickness. Radiation crosslinking of the coating layer was carried out by electron beam irradiation with a constant dose of 30 kGy. The morphology of the coated fabrics was examined by a scanning electron microscope (SEM), which indicated compatibility between the coating layer and fabric. Properties affiliated with hydrophilicity, especially water uptake and weight loss before and after several washing cycles, were determined. The effect of AAc concentration on the hydrophilic properties of the coated fabrics was studied. A considerable enhancement in water uptake was attained for nylon-6 by increasing the AAc content in the solution, followed by polyester woven and polyester knitted fabrics. The performances of the modified textile fabrics were evaluated in terms of the recovery of Cu^{+2} and Cr^{+3} from aqueous solution. The metal ion absorption efficiency of the modified textile fabrics was measured by UV-Spectrophotometer analysis and EDX. Parameters affecting the efficiency of these textile fabrics in the removal of metal ions from the aqueous solution, namely the concentration of AAc. and the immersion time, was studied. It was found that there is a marked increase in the recovery of metal ions when both the immersion time and concentration of AAc are increased. The results obtained showed that there is a good possibility of using such modified textile fabrics for the removal of some heavy metals, such as Cu and Cr.

Key words: *carboxy methyl cellulose, acrylic acid, hydrophiliity, electron beam.*

Introduction

A large amount of heavy metal ions are released by human activity (industries, transport, etc.) in the three compartments of the environment: air, water and ground. The main source of metal ion rejection in the aquatic environment is domestic water (As, Cr, Cu, Mn and Ni) [1]. Treatment methods traditionally used for the removal of metals from wastewater, including precipitation and ion-exchange, are unable to meet the new and more stringent requirements and /or are expensive [2].

Therefore there is a growing need for the development of new, innovative and cost effective methods for the removal of heavy metals from waste streams. Sorption and utilising low – cost filter materials would be an attractive option for small businesses, industries and municipalities to remove significant portions of the total metal concentrations to levels which will be less detrimental to public health and environmental quality [3]. Low-cost adsorbents are materials that generally require little processing and are abundant in nature, or are by-products or waste materials from other processes [4].

The has been an emergence of new polymer materials with intelligent properties is in answer to specific requirements for the application of these polymers in the removal of heavy metals from wastewater. Polyvinyl alcohol and carboxymethyl cellulose, which are considered as natural and biodegradable polymers, are a very significant class of polymers used in this application [5]. Moreover, natural and synthetic textiles show a lot of promise as materials used for heavy metal ion recovery. Since synthetic textiles are hydrophobic polymers, it is necessary to improve their hydrophilicity using several techniques, such as the surface coating technique.

A study was made to modify different textile fabrics, such as cotton, cotton / polyester blend and nylon-6 fabrics by surface coating with a constant thickness layer of 25 μm of aqueous solution of PVA and acrylic acid (AAc) [6].

In this context, a new material with desired properties can be prepared by applying several techniques, among which ionising radiation is the most convenient as it is free from pollution and easy to transport [7].

Moreover, the radiation grafting of specific monomers on polymer surfaces is a very interesting strategy for obtaining the properties desired for specific uses because it may change the surface activity without causing serious modifications of the mechanical polymers in a polymer [8, 9].

The graft copolymerisation of acrylamide (AAm) monomer onto polyethylene coated polypropylene (PE-co-PP) non-woven fabric was carried out by the mutual irradiation method[10].

Cellulosic materials containing various amounts of grafted polyacrylonitrile and poly(acrylic acid) molecules were used to remove Cd (II) and Cu (II) ions from aqueous solution [11].

In the present study, a coating solution of aqueous CMC containing acrylic acid in the presence of a cross-linking agent was prepared. The curing of these coated layers was carried out using electron beam irradiation. The textile fabrics modified, such Nylon, polyester woven and polyester knitted fabrics were used to remove such metal ions as Cu(II) and Cr(III) from the aqueous solution. Moreover, the efficiency of the uptake of heavy metal ions by the coated fabrics was measured by a UV- visible Spectrophotometer and EDX.

Experimental

Materials

The fabrics used were plain polyester woven and polyester knitted fabrics, which were kindly supplied by the Egyptian Company for Textile (Hosni and Bros.), Cairo, Egypt, and Nylon-6 was supplied by El-Nasr Company for Weaving and Net Fabrics (El-Shorbagy), Cairo, Egypt. All these fabrics were scoured and bleached, but were not subjected to any finishing processes. A laboratory grade acrylic acid monomer with a purity of 99%, from Mercke company, Germany, was used as received. CMC sodium salt (pure polymer) in the form of granules, M.Wt. 180, 000 was supplied by El- Nasr Pharmaceutical Chemical-Prolabo (Egypt), and N,N-Methylene bisacrylamide (MBAM), from Aldrich, Wisconsin, USA, was used as a cross-linking agent.

Coating preparation and irradiation

A known weight of CMC powder (concentration 5%) was dissolved in a known volume of distilled water. The coating solution was first prepared by dissolving the ratios of monomer (acrylic acid) required in the aqueous CMC solution with continuous stirring. This solution was then coated on different fabrics with a floating knife coater of 25 mm thickness. The surface coated fabrics were exposed to accelerated electrons using an electron beam accelerator of 1.5 MeV and 25 kW, made by High Voltage Engineering, USA, at the National Center for Radiation Research and Technology, Cairo, Egypt. The dose required was obtained by adjusting the energy parameters of the electron beam and conveyor speed. The dose used was 30 kGy. The coating percent of the coated fabrics was calculated according to Equation 1.

$$\text{Coating percent} = (W_2 - W_1)/W_1 \times 100 \quad (1)$$

Where W_1 is the initial weight of uncoated textile fabric, and W_2 is the final weight of textile fabrics after coating and several washings.

Water uptake measurements

Water uptake measurements were made using coated and dried samples of known weight immersed in distilled water for 24 h at 25 °C. The samples were removed, blotted on absorbent paper and quickly weighed. The water uptake percent is given by

$$\text{Water uptake in \%} = (W_2 - W_1)/W_1 \times 100 \quad (2)$$

where W_1 is the weight of the dry coated textile fabric, and W_2 the weight of the wet coated textile fabric after being immersed in water, taking into consideration $W_2 > W_1$.

Washing cycle measurements

Washing cycle measurements were carried out by weighting the coated sample, (W_1) then immersing it in a water bath at 40 °C for 10 min and finally drying it in a vacuum oven. This process was repeated 10, 15 and 25 times. After each of these washing times, the coated textile fabric was weighed (W_2). The weight loss is calculated by the following equation:

$$\text{Weight loss in \%} = (W_2 - W_1)/W_1 \times 100 \quad (3)$$

Scanning electron microscope (SEM) measurements

The surface morphology of the different fabrics before and after surface coating was examined by scanning electron microscopy (SEM; JEOL- JSM- 5400 scanning electron microscope – Japan) using an energy dispersive spectroscopy (EDS) X- ray spectrometer.

Determination of metal uptake

The metal uptake was determined by immersing the constant weight (W) of the fabric in different metal solutions of pH7 and constant concentration in mg/L until equilibrium. The remaining metal concentration in the solution was determined by a UV Spectrophotometer. The metal uptake was achieved by the batching method and determined as follows:

$$\text{Metal uptake in mg/g} = (C_o - C_1)/(100 \times W)$$

where C_o, C_1 are the initial and remaining concentrations of metal ions in mg/l (ppm), and W is the weight of the fabric in g. The constant 100 is because 10 ml of the metal ion solution was used instead of one litre, hence we divide it by 100.

Results and discussion

Coating percentage measurements

Coating is an advanced technique for surface and bulk modification of polymer properties. Radiation coating, especially by electron beam, is one of the most promising techniques due to its power fullness, easy attainability, high depth of penetration, and wide range of dose rates. Moreover, its high efficiency makes it appealing for a clean, simple and safe operation [12].

In this section, the coating of different types of textile fabrics such as nylon, polyester woven and knitted fabrics was carried out using a coated layer consisting of a hydrophilic polymer and monomer (CMC and AAc). ***Figure 1.a*** illustrates the effect of AAc concentration in the aqueous solution of CMC on the coating percent for modified fabrics.

From these data it was found that generally the coating percent of different textile fabrics increases with increasing AAc concentration, which was explained by the presence of free radicals available for eventual interaction with coating solution ingredients, namely the predominant radiation cross-linkable polyelectrolyte PAA and radiation degradable CMC(1) as well as with their radicals. Moreover, it was found that the value of the coating percent of all the fabrics is arranged in the following order:

Polyester knitted fabric > nylon-6 > polyester woven fabric

The less value of the coating percent for polyester woven fabric when compared to that of the knitted one is due to the difference in the spinning method which determines the extent of the penetration of the coated layer inside the woven fabric [13]. There is a similarity between nylon-6 and polyester knitted fabric in their nature which explains the high coating percent values obtained for them. The increase in the value of coating percent for polyester knitted fabric is due to direct interaction between the coated layer at the interface and the nature of polyester knitted fabric as a tricot, which has a plainwarp – knitted structure and therefore has larger capillary spaces into which the coating can penetrate [14].

Durability of coated layers towards the washing cycle

One of the important properties which had to be studied after the treatment of the fabrics was the durability of the coating layer towards washing cycles in the case of three acrylic acid concentration. Hence, the effect of several washing cycles on the durability of the coated fabrics was studied, the results of which are shown in ***Table 1***. From these results it was found that there is a decrease in the

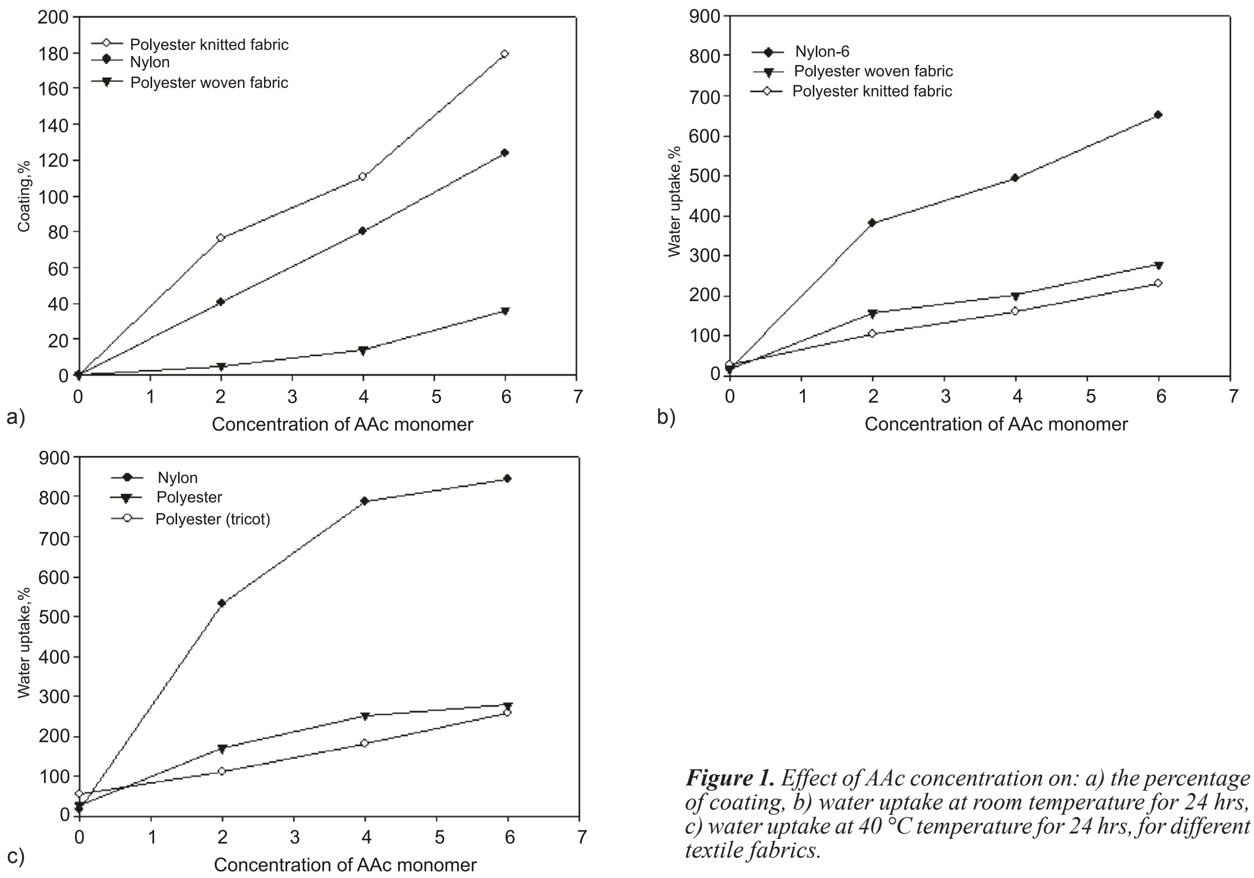

Figure 1. *Effect of AAc concentration on: a) the percentage of coating, b) water uptake at room temperature for 24 hrs, c) water uptake at 40 °C temperature for 24 hrs, for different textile fabrics.*

value of coating percentage for all the fabrics at all the AAc concentrations under investigation. Furthermore, the durability of the percent of coating for the fabrics is in the following order:

Polyester knitted Fabric > nylon-6 > polyester woven Fabric

These results again confirm the difference in the stability of the coated layer for the three fabrics under investigation. The higher the coating percent, the higher the durability.

The weight loss percent of the different coated fabrics was calculated, and the results of which are in ***Table 2***.

From these results it was found that for polyester knitted fabric an increase in AAc concentration in the coated layer does not affect the weight loss in %, which again indicates the stability of the coated layer on this fabric. However, in the case of polyester woven fabric and nylon-6, the weight loss decreases with an increase in AAc concentration in the coated layer, which indicates that a high stability of the coated layer is obtained at high AAc concentrations (e.g. 6%).

It would then be expected that the durability and stability of the coated layer of the three types of fabrics used in this work are due to the possible chemical linking of side chains of PAAc and eventually CMC with free radicals formed on the fabrics. This process may be facilitated considering that the solubility parameters of the polyester knitted fabrics 10.73 $(cal/cm^3)^{1/2}$ [15], CMC 11.4 $(cal/cm^3)^{1/2}$ and the final PAA of 12 $(cal/cm^3)^{1/2}$ [15] are close enough, indicating a possible compatibility between the three fabrics and coating layer ingredients. Under

Table 1. *Durability of coated layers towards washing cycle.*

Conc. AAc, %	Washing cycle	Coating, %		
		Nylon	Polyester (Tricot)	Polyester
2	Control	151.6	108.9	263.6
	1st	44.1	80.2	4.8
	2nd	40.8	77.6	4.6
	3rd	40.2	75.5	4.5
4	Control	302.9	157.9	218.9
	1st	85.6	127.4	15.8
	2nd	80.1	119.5	15.6
	3rd	78.9	110.7	15.3
6	Control	323.7	258.6	228.4
	1st	132.8	179.2	44.9
	2nd	123.9	179.2	37.4
	3rd	123.9	179.2	35.5

Table 2. *Weight loss in % of different textile fabrics.*

Conc. AAc, %	**Wight loss, %**		
	Nylon-6	**Polyester woven fabric**	**Polyester knitted fabrics**
2	73.4	98.2	30.6
4	73.9	93.0	29.9
6	61.7	84.4	30.7

these circumstances it may be expected that the chemical linking of strong hydrogen bond groups, namely OH groups from PAAc and CMC, in the three fabrics has taken place.

Water uptake measurements

The sorption and permeability of water is important for the comfort and wear properties of textiles where body contact or extreme humidity levels are encountered. Most water sorption and permeation occurs in the amorphous regions of semicrystalline fibers [16].

The dependence of the water uptake percent of different unmodified and modified textile fabrics on the concentration of AAc in the aqueous solution of CMC was studied, the results of which are shown in ***Figure 1.b*** (see page 101). The data obtained indicate that the initial unmodified polyester knitted fabric, polyester woven and nylon-6 fabric attained a water uptake percent value equal to 25.3, 16.7, and 15.7% respectively.

On the other hand, there is a marked increase in the water uptake percent value of all the coated fabrics. These values increase with an increase in AAc content in the coating solution, but at different rates. For example, an increase in AAc from 0 to 6% is accompanied by an increase in water uptake from 15.7% to 650.9, from 16.7% to 278% and from 25.3% to 230% for Nylon, polyester woven and polyester knitted fabrics, respectively. The rate of increase may be given in the following order for all the fabrics.

Nylon-6 > polyester woven fabric > polyester knitted fabric

It is noteworthy to observe here that the order of the increase in coating percent is contrary to the order of the increase in water uptake. That is to say that a higher coating in % means a lower water uptake in %, which was explained by the fact that the higher coating percent results from the complete absorption of the coated layer by the fabric, causing the blocking of the opened structure in the fabric, and hence the penetration of water is restricted.

The difference in the value of the percent of water uptake between the two polyester fabrics is due to the high penetration of the coated layer inside the interior of the knitted fabric, while in the case of woven fabric the absorption of the coated layer

Table 3. *Durability of water uptake in % of coated textile fabrics towards washing cycle.*

Conc. AAc, %	Washing cycle	Water uptake, %		
		Nylon	Polyester (Tricot)	Polyester
2	Control	379.6	103.4	156.0
	1st	379.1	102.0	150.0
	2nd	372.4	101.5	146.0
	3rd	371.0	101.5	146.0
4	Control	492.5	160.9	196.4
	1st	475.2	155.0	194.9
	2nd	469.9	155.0	194.6
	3rd	468.0	155.0	194.6
6	Control	650.9	230.0	278.0
	1st	601.9	215.0	260.0
	2nd	574.5	213.0	256.0
	3rd	570.0	213.0	256.0

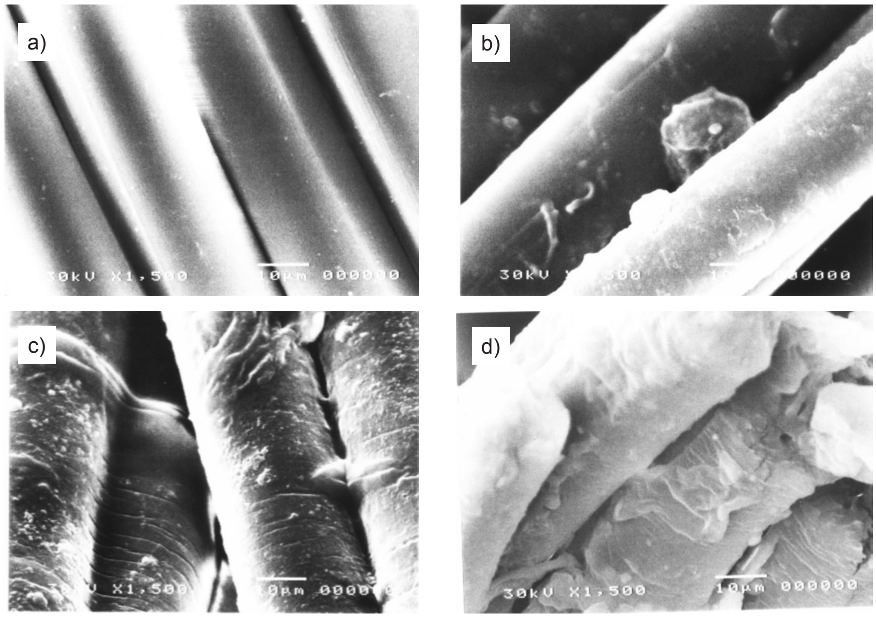

Figure 2. *SEM of modified and unmodified Nylon-6; a) unmodified nylon, modified with: b) 2% AAc, c) 4% AAc, d) 6 % AAc.*

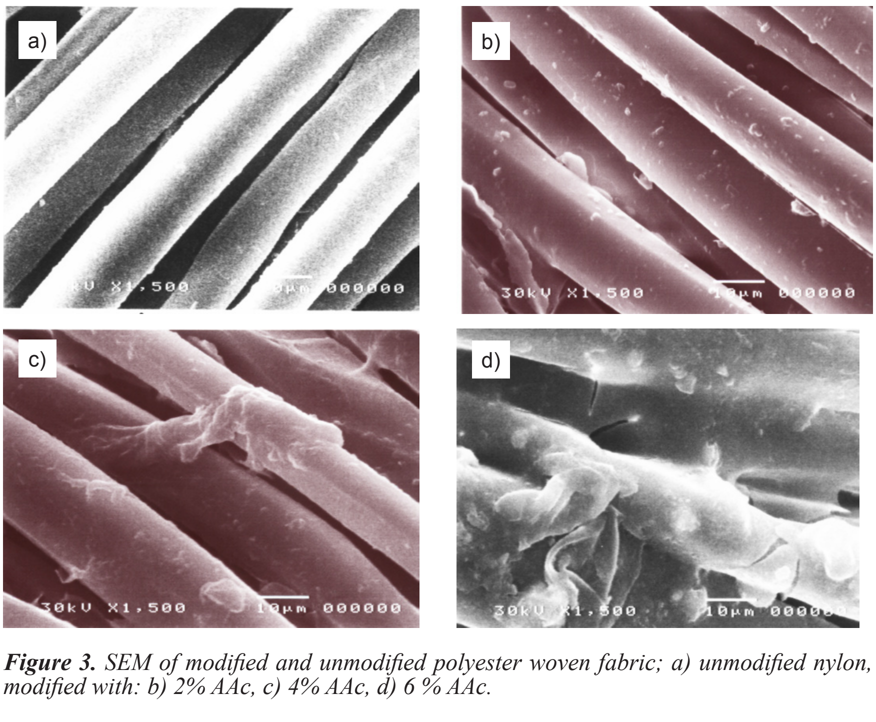

Figure 3. *SEM of modified and unmodified polyester woven fabric; a) unmodified nylon, modified with: b) 2% AAc, c) 4% AAc, d) 6 % AAc.*

is on the surface only, permitting high penetration of water inside the fabric.

The effect of temperature on the water uptake of the different textile fabrics was studied, the results of which are shown in ***Figure 1.c*** (see page 101). It was found that the water uptake increased with a rise in temperature to 40 °C and with an increase in the concentration of AAc content in the coating solution. This trend was due to the fact that increasing the temperature caused the opening of the coated layer structure on the textile fabrics, which facilitated the diffusion and absorption of water. Moreover, the temperature effect and water could lead to length and width shrinkage, leading to changes in capillary spaces in the fabrics.

Durability of the water uptake percent of the coated fabrics

It is important to investigate the durability of the water uptake in % of the coated fabrics by calculating the water uptake in % after several washing cycles until constant weight, the results of which are shown in ***Table 3***. It was found that there is a decrease in water uptake in % after three washing cycles with an increasing AAc concentration for all the coated fabrics under investigation. It is important to note that the biggest decrease was observed in the case of Nylon-6 fabric for an AAc concentration of 6%. For example, the water uptake of Nylon-6, polyester woven and polyester knitted fabric at 6% was found to be 651, 278, and 230, which then decreased to 570, 256, and 213, respectively. Moreover, there is a marked stability of the water uptake for all the coated fabrics after the second and third washing cycles. These results show that the absorption of water is not on the surface, but there is a strong chemical bond between components of the coated layer on the fabric and water.

Table 4. *Sorption of metals by different textile fabrics*

Conc. of AAc, %	Time, hrs	Amount of metal ion uptake, mg/L					
		Nylon-6		Polyester woven fabric		Polyester knitted fabric	
		Cu(II)	Cr(III)	Cu(II)	Cr(III)	Cu(II)	Cr(III)
2	24	16.4	20.8	10.0	12.0	11.0	13.0
	48	20.1	29.3	17.1	18.2	16.1	17.4
	72	20.1	29.3	17.1	18.2	16.1	17.4
4	24	25.5	28.4	18.1	24.0	15.4	17.8
	48	25.5	37.6	19.5	32.0	17.4	24.5
	72	25.5	37.6	19.5	32.0	17.4	24.5
6	24	29.7	38.4	22.4	25.4	17.4	18.3
	48	42.0	47.5	22.4	27.4	19.3	21.9
	72	50.5	51.6	22.4	27.4	19.3	21.9

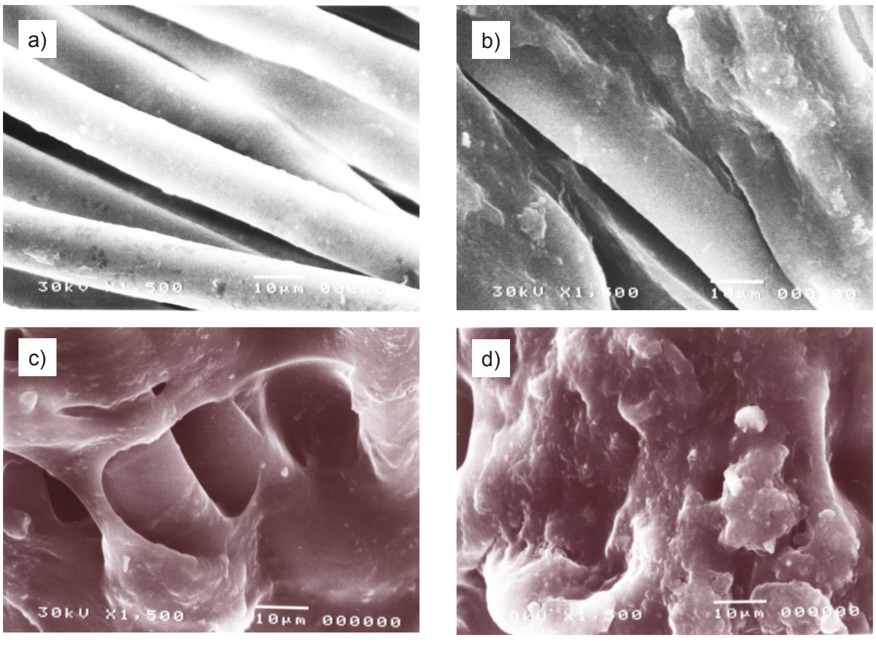

Figure 4. *SEM of modified and unmodified polyester knitted fabric; a) unmodified nylon, modified with: b) 2% AAc, c) 4% AAc, d) 6 % AAc.*

Surface morphology of the modified textile fabrics

The SEM technique was used to investigate the surface morphology of the different textile fabrics before and after they had been coated and cured by electron beam irradiation. The results obtained are shown in ***Figures 2, 3*** and ***4***. The surface of all the uncoated fabrics (***Figures 2, 3,*** and ***4.a***) looks smooth and distinct, while the coated fabric seems to be rough (***Figures 2, 3, 4.b, 4.c*** and ***4.d***). The fibers of coated fabrics are no longer smooth nor distinctly separate from each other, especially in the case of nylon-6 fabric. There is a clear change in the dimensions of fiber of the fabric after coating, resulting in the closeness and compactness of the weave in the fabric structure, as seen in ***Figures 2, 3, 4.b, 4.c*** and ***4.d***. This behaviour may be affiliated with the relative covering of fibers with chemically linked PAAc and CMC, which is of larger magnitude in the case of polyester knitted and nylon-6 than for polyester woven fabric, which again is a confirmation of the coating percent results discussed before. ***Figures 2, 3*** & ***4.d*** show clearly the complete covering of fabric with an active ingredient of coating solution containing 6% AAc, which is further confirmation of the result for the water uptake property previously discussed.

Sorption of heavy metal ions

In this work, the possibility of using modified coated fabric for the removal of heavy metal ions, such as copper (II) and chromium (III), was investigated.

The affinity towards these metal ions was studied using a UV –Visible Spectrophotometer, and the selectivity of specific metal ions or separate specific metal ions from aqueous solution was measured by EDX.

Determination of the percent of metal ion uptake using a UV-Visible Spectrophotometer

The percent of metal ion uptake was measured by UV-Visible Spectrophotometer, the results of which are shown in ***Table 4***. The results obtained by UV show that all the coated textile fabrics posses a higher affinity towards Cr^{3+} than Cu^{2+} , and that the affinity increases with increasing AAc concentration. This behaviour can be explained within the framework of the concept developed by Pearson concerning the softness and hardness of chemical species [17]. Pear-

Table 5. Selectivity of coated fabrics towards two metals in a mixture, determined by EDX.

Types of textile fabrics	Atomic % in coated fabrics	
	Cu(II)	Cr(III)
Nylon-6	42.83	57.17
Polyester woven fabric	45.63	54.37
Polyester knitted fabric	24.09	75.91

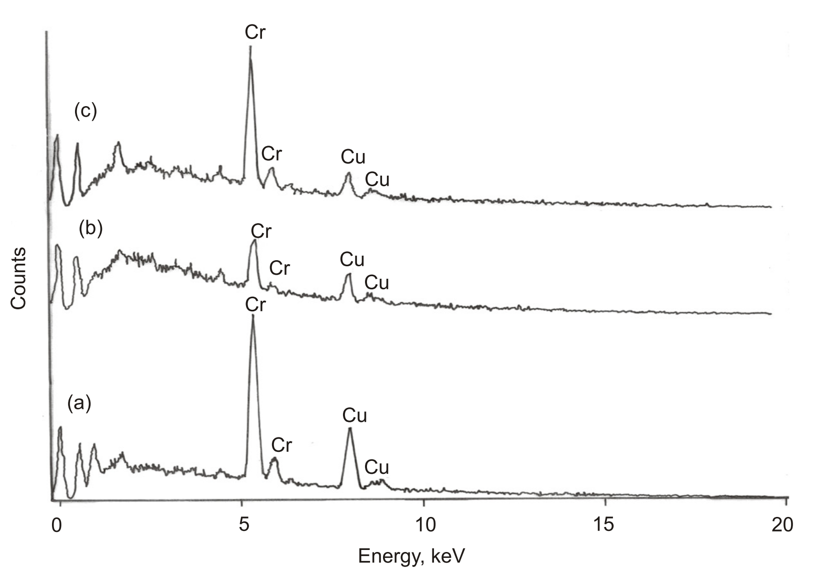

Figure 5. *EDX spectra for the uptake of different metal ions on (a) Nylon-6 (b) Polyester woven fabric (c) Polyester knitted fabric.*

son determined a scale of the hardness (or softeness) of a species according to its electronegativity, polarisability, size and charge (for ions). This theory is not quantitative but gives information about the ability of a species to be bound preferentially with another. Thus, a reagent which is defined as hard will preferably react with a hard reagent. A soft reagent will preferably react with a soft one. Cu^{2+} is defined as a soft acceptor because it is less oxydisable and, therefore, not very electropositive. On the other hand, Cr^{3+} is defined as a hard species.

This theory explains the high efficiency of Cr^{3+} complexation in a CMC/PAAc coated layer compared with Cu^{2+} [1].

Determination the selectivity of coated fabrics towards metal ions using EDX

The adsorption selectivity is an indispensable factor for appreciating the capacities of an adsorbent. With this property fabrics can be used to adsorb a specific metal ion or separate specific metal ions from a mixed metal ion solution. ***Figure 5*** and ***Table 5*** show results of the adsorption of metal ions onto Nylon-6, polyester woven and knitted fabrics from an equimolar solution of different metal ions at pH7. All the fabrics investigated have a better adsorption for Cr(III) than for Cu(II). These results can be explained by considering the valency of metals. The trivalent Cr(III) forms a more stable and stronger complex with functional groups of the coated layer, which indicates that the functional groups on the fabrics have a relatively stronger affinity for chromium ions than copper ions. The results obtained by EDX could be applied for the separation of Cr(III) in aqueous systems containing Cu(II). In general, it can be concluded that the selectivity of the investigated coated layer in fabrics towards different metals in a mixture depends mainly on the stability of the chelate and ionic valence of the chelated metal [18].

Conclusion

Different textile fabrics such as Nylon-6, Polyester woven and knitted fabrics were coated with a hydrophilic polymer and monomer (CMC and AAc) and then cured through electron beam irradiation. Firstly, the SEM confirmed the occurrence of complete adhesion between the textile fabrics and the coated layer. The hydrophilicity of the fabrics modified was examined in terms of the percent of water uptake. It was found that there is an improvement in the percent of the water uptake of the coated fabrics compared to that of the uncoated ones. The performance of the coated fabrics with respect to metal ions was evaluated by determining the removal of Cu(II) and Cr(III) ions from the aqueous solutions. It was found that these coated fabrics show a marked efficiency in the removal of heavy metal ions.

References

1. Lebrun L., Valle'e F., Alexandre B., Nguyen Q.T.; Desalination (2007) pp. 9-23.
2. Weber T.; Wastewater treatment, Met. Finish. Guidebook Directory 94(1), (1996) p. 734.
3. Mckay G., J Porter.F.; J.Chem. Technol. Biotechnol. 69, (1997) p. 309.
4. Bailey S. F., Olin J. M., Brika R. M., Adrain D. A.; Water Res. 33, (1998) p. 2469.
5. Guo J. H, Skinner G. W., Harcum W. W., Barnum. P. E.; Pharm. Sci. Technol. Today 16 (1998) p. 254.
6. Ibrahim M. S., El-Salmawi K. M., Ibrahim S. M.; Applied Surface Science 241, (2005) pp. 309-320.
7. Kyoung R. P., Young C. N.; Radiation Physics and Chemistry, 67(3-4), (2003) p. 361.
8. Anjum N., Gupta B., Riquet A. M.; J. Appl. Polym. Sci. 101, (2006) pp. 772-778.
9. Curti P. S., Moura M. R. D., Veiga W., Radovanovic E., Rubira A. F., Muniz E. C.; Appl. Surf. Sci. 245, (2005) pp. 223-233.
10. Ibrahim S. M., El-Salmawi K. M., El-Naggar A. A.; Journal of Applied Polymer Science, Vol. 102, Iss. 4, (2006) pp. 3240-3245.
11. Okieimen F. E., Sogbaike C. E., Ebhoaye J. E.; Separation and purification Technology, 44, (2005) pp. 85-89.
12. Morshedian M., Bagheri R.,Mirzataberi M., Moghadam M.; Master thesis , (2004).
13. Taskin C., Ozguney A.T., Gurkan P., Ozcelik G., Ozerdem A., Fibers & Textiles in Eastern Europe, Vol. 15, No. 1 (60), (2007) pp. 86-90.
14. Zhang C. H., Yang F. L., Wang W. J., Chen B.; Separation and Purification Technology, 61, (2008) pp. 276-286.
15. Rudin A.; The Elements of Polymer Science and Engineering, Academic Press, New York, USA, 1982.
16. Attia S. , M.Sc. Thesis, Ain Shams University, Cairo, 1999.
17. Pearson R. G.; Hard and soft acids and bases, Benchmark Papers in Inorganic chemistry, Dowden, Hutchinson&Ross, Inc., Strousburg, USA, (1973).
18. Manal F. Abou Taleb, Samia M. Elsigeny, and Mohamed M. Ibrahim; Radiation Physics and Chemistry 76 (2007) pp. 1612-1618.

Received 20.04.2009 Reviewed 26.02.2010