ANTIBACTERIAL PROPERTIES OF FUNCTIONAL POLYAMIDE 6.6

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Abstract:

*Polyamide 6.6 multifilaments are grafted with the monomer N-allyliminodiacetic acid for the purpose of removal of some heavy metal ions from their aqueous solutions by forming its metal chelate especially with Ag⁺ ion. Such a fibrous chelate-forming resin has been used with success due to its large surface area, which contains an important metal chelate-forming functional group, where metal ions are adsorbed or desorbed on its surface, and therefore having an improved adsorption and desorption capability. In previous work, chelate-forming fibre was characterised by ICP-AES analysis according to the digestion method by microwave. The antibacterial activity of the prepared fibre is investigated with *Escherichia coli* bacteria as reference, according to the zone of inhibition method in agar medium. The material used as reference without metal does not present any effect on *E. coli*. However, the chelate-forming fibres with Ag⁺ have a strong bactericidal effect, even with a low concentration of silver ions. These modified materials can be used as highly effective bactericidal composites that may be used in future applications for the production of antimicrobial textiles, papers or polymer materials.*

Keywords:

Functionalisation, Polyamide 6.6, N-Allyliminodiacetic acid, Silver ions, Antibacterial activity

1. Introduction

Recent literature has several interesting studies on the changes of the surfaces of polymers to overcome some of their weaknesses. In the polymeric age, it is essential to modify the properties of a polymer according to tailor-made specifications designed for target applications [1-3]. These materials have significant potential: fibres having bactericidal activity, being fireproof and anti-stain and so on [4].

The antibacterial functionalisation of textile fibres has become a very interesting method to develop textiles with protection against microorganisms, protect users against the spread of nosocomial infections, slow the formation of pathogenic biofilms and eliminate sources of infection, and hence their interest in the textile industry. Polyamide 6.6 is a fibre that finds its use in many areas including clothing, furniture and other technical uses.

Silver, both as a metal and in ionic form, exhibits strong cytotoxicity towards a broad range of microorganisms, and its use as an antibacterial agent is well known [5]. Indeed, silver has an oligodynamic effect, that is, silver is capable of causing a bacteriostatic (growth inhibition) or a bactericidal (antibacterial) impact [6,7]. Research indicates that silver is also effective in purification systems for disinfecting water and air [8,9].

In the above context, the association of Ag with fibre represents an interesting approach to developing a new fibre that might find a variety of new applications.

In this work, we wish to report a comparative study on the antibacterial effects of a fibre containing silver ion prepared through a variety of simple chemical strategies.

A detailed characterisation of these new materials and the determination of their antibacterial activity using *Escherichia coli* are presented in this article.

2. Experimental

2.1. Materials

Functional Polyamide 6-6 [10] was disintegrated and washed with acid solution before use.

Solution of silver nitrate (AgNO₃) was purchased from Aldrich and used as received. All the chemicals were analytically pure.

2.2. ICP analysis

In order to determine the amount of the metal captured by the chelate-forming fibre, according to the digestion method by microwave, we applied ISO 11185 standards, and the samples were leached in acid solution. The solution was analysed by ICP-AES (ULTIMA 2, JobinYvan).

2.3. Preparation of the chelate-forming fibre

The commercial solutions of AgNO₃ were further diluted to obtain 120, 60 and 25 ppm standards solutions of Ag⁺ with pH

value 5. The pH was further adjusted by the addition of 0.1 N solution of NaOH during dilution process. Batch experiments were carried out in different glass bottles at room temperature ($25 \pm 1^\circ\text{C}$) using 250 mg of pretreated grafted polyamide in 100 ml standard solution of different concentrations (scheme 1) with stirring for 1 h. The concentrations of chelating metal ions were determined using ICP-AES after digestion in microwave by acid solution according to ISO 11185 standards.

2.4. Assessment of antibacterial activity of PA-ALD/Ag⁺

The testing method used for the assessment of antibacterial activity was based on a standard methodology [11] usually adopted for textiles to verify antimicrobial additives included in the fibre structure.

3. Results and discussion

3.1. Characterisation of chelating fibre

The chemical analysis of the chelate-forming fibre is intended to determine the quantity of metal fixed by the functional fibre of Polyamide 6-6. We carried out the digestion method by microwave: 100 mg of chelate-forming fibre are introduced into 10 ml of HCl/HNO₃ solution 50:50 (v/v). After mineralisation; solutions were filtered and analysed by ICP-AES. Depending on the concentration of the initial solution of silver ions, we obtained around 20, 50 and 100 ppm of metal contents. These fibres were evaluated for an antibacterial barrier.

3.2. Evaluation of the antibacterial activity

The study of antibacterial activity of the chelate-forming fibre was done according to the zone of inhibition protocol method (presence or absence of the inhibition zone). We used *E. coli* bacteria as reference. In this study, we used the agar diffusion method with the disc maintained at 37°C for 1 day. The required nutrient agar medium was prepared by mixing peptone (5.0 g), beef extract (3.0 g) and sodium chloride (5.0 g) in 1000 ml of distilled water and the pH was adjusted to 7.0. Finally, agar (15.0 g) was added to the solution. Sterilisation of agar medium was performed in a conical flask at 120°C for 35 min. This medium was transferred into sterilised Petri dishes under a laminar air flow chamber.

After solidification of the media, *E. coli* (50 μl) from overnight incubated culture were spread on the solid surface of the media. Over this inoculated Petri dish, small pieces (50 mg) of chelate-forming fibres (20, 50 and 100 ppm of silver ions) and non-chelate-forming fibre used as control (blank) were distributed and incubated for 48 h at 37°C in an incubation chamber. The experiment was performed in triplicate; figure 1 illustrates these results.

In the picture, we observe that the fibre used as control (non-chelating fibre: 0 ppm) does not present a zone of inhibition: we see the growth of microorganisms on the fibre. However, the chelating fibres with different rates of silver ion (20, 50 and 100 ppm) presented a zone of inhibition (transparent zone).

Scheme 1: The chelate-forming fibre of polyamide 6.6 with silver ion

Figure 1. Zone of inhibition tests of polyamide non-chelating fibre (0 ppm) and chelating fibre with different concentrations (20, 50 and 100 ppm).

We note that the diameter of the zone of inhibition is very clear and important for 50 and 100 ppm; for this purpose, we have reduced the concentration of silver ions to 20 ppm that showed excellent activity against microorganisms.

This result is promising on its own because this material can be used as an antibacterial additive textile, which could have potential use for painting hospital environments, for example.

4. Conclusions

The present work relates to the preparation of functionalised polyamide fibres by chemical grafting and the use of these fibres in the extraction of dissolved metal ions. In this study, the chelate-forming fibre is evaluated for antibacterial activity.

Significant antibacterial activities have been demonstrated in chelating fibre samples against *E. coli* tested bacteria through the action of silver ion. The antibacterial activities could be reached with low concentrations of silver ions in the sample.

The Ag concentration can be controlled using different synthesis methods and/or different substrates. According to these results, it rightly shows that this chelate-forming fibre can find interesting applications such as clinical wound healing, biofilms and the coating of biomedical materials. For these reasons, we will extend this work to other ions or NPs (Zn^{2+} , Cu^{2+} ...), and also be make studies with other types of bacteria.

References

- [1] Bhattacharya, A., Misrab, B.N., (2004). Grafting a versatile means to modify polymers techniques, factors and applications, *Progress in Polymer Science*, 29, 767–814
- [2] Yoshikimi, U., Koichi K., Yoshito, L., (1998). Surface modification of polymers by grafting, *Advances in Polymer Science*, 137, 1-39
- [3] Baran, T. A., Hamarat, B. S., (2007). Immobilization of urease using glycidyl methacrylate grafted nylon-6 membranes, *Process Biochemistry*, 42(3), 439-443
- [4] Xie, W., Xu, P., Wang, W., Liu, Q., (2002). Preparation and antibacterial activity of a water-soluble chitosan derivative, *Carbohydrate Polymer*, 50, 35–40.
- [5] Makhoulouf, C., Marais, S., Roudesli, S., (2007). Graft copolymerization of acrylic acid onto polyamide fibers, *Applied Surface Science*, 253, 5521–5528.
- [6] Wang, P.H., Pan, C.Y., (2000). Polymer metal composite microspheres preparation and characterization of poly(St-co-AN)Ni microspheres, *European Polymer Journal*, 36, 2297-2300.
- [7] Louati, M., El-Achari, A., Ghenaim, A., Caze, C., (1999). Graft copolymerization of polyester fibers with a fluorinecontaining monomer, *Textile Research Journal*, 69, 381–387.
- [8] Sondi, I., Salopek-Sondi, B., (2004). Silver nanoparticles as antimicrobial agent: a case study on *E. coli* as a model for gram-negative bacteria, *Journal of Colloid and Interface Science*, 275, 177–82.
- [9] Zhang, S., Fu, R., Wu, D., Xu, W., Ye, Q., Chen Z., (2004). Preparation and characterization of antibacterial silver-dispersed activated carbon aerogels. *Carbone*, 42, 3209-3219.
- [10] El Bouchti, M., Hannache, H., Cherkaoui, O., Functional polyamide 6.6 fiber with n-allyliminodiacetic acid: synthesis and characterization, *Journal of Advances in Polymer Technology*, 32(4), 2013 Doi 10.1002/Adv.21378.
- [11] Textile fabrics - Determination of the antibacterial activity - Agar diffusion plate test, 2005, NF EN ISO 20645.