Composite Wool – Natural Product Antimicrobial Textiles

S. Aljohani* and J.H.Johnston**

Victoria University of Wellington
School of Chemical and Physical Sciences
Wellington, New Zealand
*sami.aljohani@vuw.ac.nz
**jim.johnston@vuw.ac.nz

ABSTRACT

The growth of microbes on textiles adversely affects the user and damages the textile. This can be controlled by antimicrobial finishing of the textile using specific antimicrobial agents. In this work, we present a novel use of natural New Zealand Manuka honey (MH) and the practical use of the active ingredient methylglyoxal (MGO) in the honey as natural antimicrobial agents in wool fibres and textiles. The maximum amount of MH and MGO that were incorporated into the wool fibres were 15.36 mg/g and 215 mg/g respectively. The absorption and diffusion rates of MGO into wool fibres were highly dependent on the initial concentration, temperature and diffusivity. The MGO-Textile composites exhibited a zone of inhibition against *Staphylococcus aureus (S.aureus)* and *Escherichia coli (E.coli)* microbes.

Keywords: Methylglyoxal, Manuka honey, antimicrobial activity, textiles, wool, zone of inhibition.

1 INTRODUCTION

Wool textile products are widely used in apparel, furnishings and floor coverings. They also find use in medical, and healthcare applications such as bandages and masks which sometimes can be a medium to support microbial contamination. This is mainly due to the large surface area of the fibres and their ability to retain moisture. Synthetic fibres often generate more resistance to microorganisms than natural textiles due to their hydrophobicity [1]. However, as a protein fibre, natural textiles provide a hospitable host for the growth of microorganisms. This growth on textile surfaces increases the potential risk of infection, and transmission of microbial diseases [2]. The properties of natural fibres such as resilience and comfort attributes increase the demand on enhancing its functionalities and hygienic textile products.

Recently, antimicrobial finishing and functionalising of textiles have become extremely important to impart particular agents with antimicrobial properties into textiles. A number of studies of such antimicrobial fibre composites have reported for synthetic fibres and textiles, and to a lesser extent for natural fibres, notably cotton [3]. The wool textile fabrics have been successfully functionalized with nanogold as a novel colourant for the high value apparel and rug markets, and with nanosilver for antimicrobial apparel, furnishing fabrics and carpets [4]. The resulting nanosilver-wool fibres exhibit high antimicrobial activity against *S.aureus* and *E.coli microbes*. This silver-based antimicrobial treatment is very effective and depending on the actual applications, EPA approval may be required. It is therefore interesting to explore the use of naturally compounds that exhibit antimicrobial activity we present here.

Manuka honey (MH) made from pollen collected by bees fom the native Manuka trees (*Leptospermum scoparium*) from New Zealand is well known for its natural antibacterial medicinal properties and marketed and sold extensively under this attribute. The antibacterial activity is collectively attributed to the high level of sugar, moderately low pH, the presence of hydrogen peroxide, and non-peroxide compounds [5]–[7]. Methyl glyoxal (MGO), a highly reactive α-ketoaldehyde, has been identified as a non-peroxide antibacterial compound in this honey, which has been linked to its sought after and effective antimicrobial activity [8, 9]. An example of the commercial benefit of MH, Comvita® ,one of the largest producers of MH captures the antibacterial property of MH in various products such as wound gel, wound dressings and skin cream..

The minimum inhibitory concentrations (MIC) of MGO in MH and the isolated MGO compound have been reported ranging between 1.1 and 1.8 mM, which attracted attention for potential applications [10]. Several studies addressed the biological activity of MH and MGO in the form of wound dressings [11], polymer-based complexes [12] and poly vinyl alcohol (PVA) fibres [13]. Also, the coatingof cotton textile fibres with MGO and MH has been reported [14]. However, in this method, the textile was immersed into MH and MGO solutions for a short period of time and then dried and tested for their antimicrobial property. This coating is unlikely to have any durable bonding of the MH or MGO to the textile fibres and would readily wash off. A determination of the actual extent of uptake of MH or MGO has not been reported.

Here we present a brief introduction to our more detailed study [15] on the incorporation of MH and MGO into wool fibres and wool textiles and an assessment of the antimicrobial effectivness of the treated materials.

2 RESULTS AND DISCUSSION

2.1. The uptake analysis

We have innovatively incorporated MH and similarly MGO into wool fibres in loose top form, yarn and in finished fabrics. The extent of uptake of MH and MGO from solutions at different temperatures and concentrations has been characterised by High Performance Liquid Chromatography (HPLC). Both MH solutions (w/v) and isolated synthetic compound (MGO) solutions were studied. However, the higher viscosity of honey limited measuring the uptake up to 3 mM of MGO concentration in MH UMF10+ manuka honey.

The dDerivatisation of 1,2-dicarbonyl compounds with ortho-phenylenediamine and subsequent analysis of the corresponding chinoxalines using HPLC with UV detection at 312 nm is a well established method for the quantification of these compounds [10]. This method was used with some minor modifications to quantify the amount of MGO in solutions used in the uptake and in MH. Also, an external standard calibration was carried out.

All different forms of wool were treated with MGO and MH solutions. The loose top fibres showed a relatively higher absorption rate due to the larger accessible surface area. At room temperature, the diffusion rate was very slow resulting in minimal uptake of MH or MGO after one day's treatment. However the use of higher temperatures ranging between 50 and 80 °C significantly accelerated the uptake and diffusion rate into the fibres, enhancing the total amount of MGO alone and in MH solutions, absorbed by the wool. This is consistent with the generally long times, up to 14 days that are required for dye uptake and colour levelness to be achieved at higher temperatures. These treated samples showed a negligible leaching of the MGO on washing.

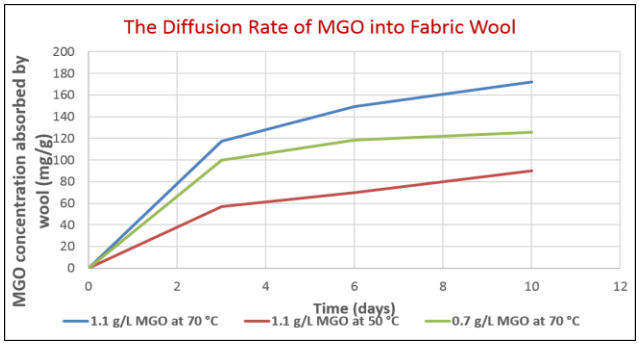

Figure 1: The effects of temperature and concentrations on the diffusion rate into textile fibres

Figure 1 shows how the uptake and diffusion of MGO into wool fabric increases from 90 mg/g (MGO/wool) at 50 ºC to 176 mg/g at 70 ºC. There is also an increase in the uptake as the concentration of MGO in solution is increased from 0.7 g/L to 1.1 g/L at 70 ºC. The MGO molecules are considered to diffuse intercellularly between the cuticles

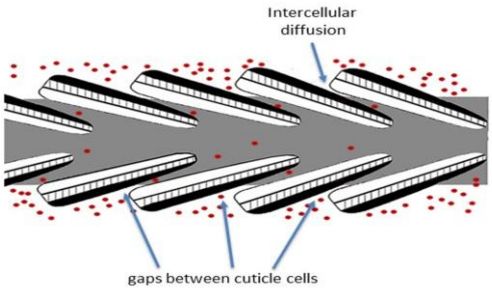

Figure 2: an illustration of the MGO diffusion pathway into wool fibres.

into the bulk fibre (Figure 2). The diffusion and hence the uptake is essentially first order (exponential) in nature and therefore dependent on the initial concentration of MGO and the temperature. Long times are required to reach satutation uptake under a particular set of conditions.

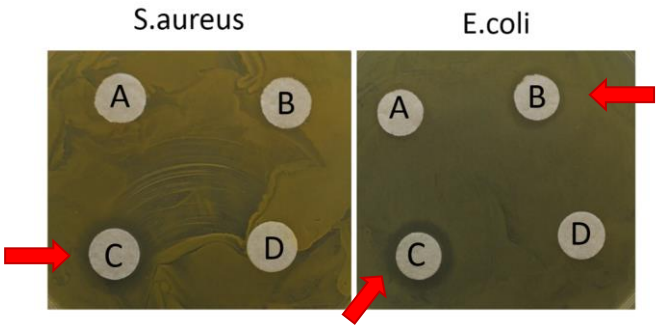

Figure 3: The zone of inhibition of paper coated with MGO solutions at: A) 6.9 mM B) 13.9 mM C) 27.8 mM D) 3.47 mM against S.aureus and E.coli.

2.2. Antimicrobial performance of MGO-Wool composites

The coated MH and MGO samples were tested against gram-negative *E.coli* and gram-positive *S.aureus* at varying MGO concentrations. The MGO-Wool sample 0.4 g/g, after determining the uptake, exhibited 371 mm of a clearly inhibition zone area toward *S.aureus* and non for *E.coli*.

In a related study, the uptake of MGO into paper substrates was studied similarly. Here the paper sheet was immersed into MGO solutions at various concentrations for 10 minutes then let to dry at room temperature before testing their antimicrobial performance. This treatment resulted in a significant zone of inhibition largely against *S.aureus* (Figure 3). It was observed that washing the treated paper samples with water prior to this antimicrobial

testing resulted in a reduction of the zone of inhibition size. This demonstrates that the MGO is the weakly bonded to the paper fibres by this immersiuon method. The binding of MGO to the wool fibres using the methodology at higher temoperatures described above, produced and treated wool fibre product that showed negligible leaching of the MGO from the wool on washing. Hence this is the preferred application method.

3 CONCLUSION

In this study, the innovative development of a wool composite with the natural antimicrobial agent MGO alone or as a component of MH is reported. The uptake of MGO by textile fibres was characterised and the treated MGO wool product showed excellent durability against leaching in water, suggestuing the MGO is chemically bound to the wool fibres. . The MG-wool composite produced zones of inhibition when tested against Gram-positive *S.aureus* and Gram-negative *E.coli* bacteria. This suggests possible applications in use MH and the active ingredient MGO in antimicrobial woollen apparel and medical textiles and bandages.

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