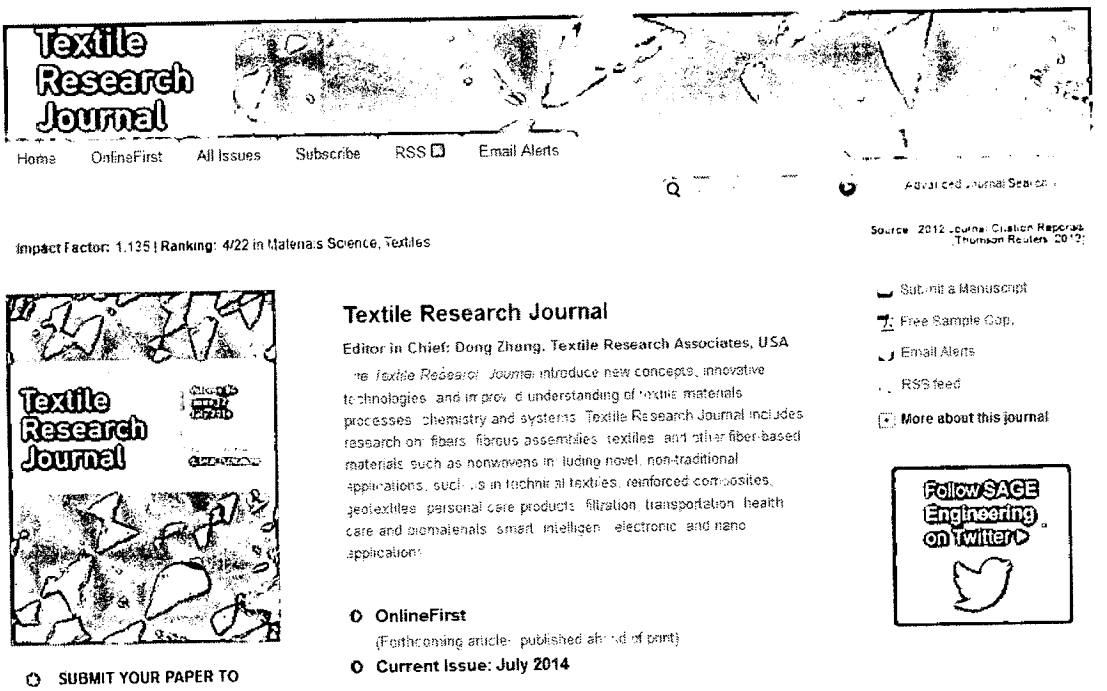
ภาคผนวก ก
ผลผลิตที่ได้จากโครงการ

ผลงานวิจัยอยู่ในระหว่างพิจารณาโดยผู้เชี่ยวชาญของวารสาร Textile Research Journal ซึ่งมีค่า mpact factor ปี 2012 อยู่ที่ 1.135 เป็นวารสารที่ก่อตั้งเมื่อปีค.ศ. 1930 ปัจจุบันถูกจัดอยู่ในลำดับที่ 4 จาก ำนวน 22 วารสารที่เผยแพร่งานวิจัยทางด้านสิ่งทอ



ภาคผนวก ข

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Preparation, characterization, and *in vitro* evaluation of antibacterial sol-gel coated cotton textiles with prolonged release of curcumin

Abstract

This study aims to develop a textile-based sol-gel release system with antibacterial properties intended for medical applications. Curcumin (a herbal compound extracted from *Curcuma longa* L.) was embedded in an acid-catalyzed silica xerogel coating on cotton textiles. The physical properties and anti-bacterial activity of the coated textiles were also evaluated. The release of curcumin was found to be purely diffusion-controlled as expressed by the Higuchi and the first-order models. Matrix dissolution release was ruled out by the absence of dissolved silica in the release medium. Results from nitrogen sorption analysis confirmed the presence of mesopores in the xerogel coating, and an increase in both the BET surface area and mesopore volume after the sol-gel treatment. Thicker xerogel coatings therefore provided greater porosity for curcumin entrapment, leading to retardation of the diffusion processes and consequently extended release profiles. The treated samples showed better anti-bacterial properties against *Staphylococcus aureus* than *Escherichia coli*. Overall, the developed sol-gel coated textiles demonstrate promising potential for use as wound dressing materials.

Keywords

Sol-gel, controlled release, cotton, curcumin, antibacterial properties

Introduction

Recently, sol-gel coating techniques have been employed by several research groups to improve the properties of, or add special features to, traditional textile products. One of the most widely-used sol-gel systems utilizes an alcoholic solution of alkoxysilane as a sol-gel precursor that is transformed into a porous solid network via hydrolysis and condensation reactions. To broaden the range of textile functionalities, direct modification of the sol-gel precursor or the uses of additives are two major approaches. The sol-gel coated fabrics have been demonstrated to acquire a wide range of additional functions according to the modification route: fabrics showing fire-retardant,[1] anti-static,[2,3] water/oil repellent,[2–4] insect-repellent,[5] anti-microbial,[6] and catalytic characteristics[7] have been produced. The additive initially introduced to the precursor solution during preparation may be leached out from the solidified sol-gel network through the matrix channels, or as a result of matrix dissolution when in contact with liquid media.[8] The possibility of gaining control over the sol-gel structure by varying parameters such as the type of precursor, temperature, pH, precursor:water ratio, and consequently tuning its release characteristics, have paved the way for the development of sol-gel controlled release systems.[9]

Since one important and rapidly growing area of the textile industry is the medical sector,[10] effort is currently being put into coating textiles with sol-gel technology for medical use, mainly as wound dressing materials.[11] The controlled-release principles

ıased on sol-gel systems have been applied to textiles by Mahltig *et al.* who developed ıntimicrobial sol-gel coated textiles with the use of silane precursors and four different ıiocides, i.e. inorganic biocide (silver metal and its nitrate salt), and organic biocides Octenidine and CTAB) on polyester/cotton.[6] More recent work has reported the use of ;ilver, copper, and HTAT for developing antimicrobial viscose textiles[12] and capsaicin chili pepper extract) for antimicrobial wool fabrics.[13] Due to the potential toxicity to ıumans, and environmental concerns about the use of some of these biocides, ıaturally-occurring and biocompatible compounds are preferable for use in wound ıressings and other medical textile applications. Curcumin (1,7-bis(4-hydroxy-3-ıethoxyphenyl)-1,6-heptadiene-3,5-dione) is a herbal compound found in turmeric (*Curcuma longa L.*). The WHO (World Health Organization) and FAO (Food and Agriculture Organization) have verified its safety as a natural food colorant (C.I. Natural Yellow 3).[14] As shown in Figure 1, curcumin exists in at least two tautomeric keto- and ənol forms, with equilibrium strongly favouring the enol form. The enol structure allows for inter- and intramolecular hydrogen bonding to occur. Pharmacological studies have ɔonfirmed that curcumin possesses anti-inflammatory, antifungal, antitumour, and antioxidant characteristics. It has been shown to promote wound healing in animals by ənhancing cell proliferation and effective free-radical scavenging.[15] It is believed that the methoxyl and hydroxyl groups are responsible for the microbial activity although the actual mechanism is not fully understood.[14] In light of the benefits of curcumin to wound healing, along with its ubiquitous availability and environmental compatibility, this herbal substance may be a good candidate for wound dressing applications.

[insert Figure 1.]

Although much research has dealt with sol-gel release systems, data on release characteristics when textiles are used as flexible, porous substrates is limited. More detailed examinations of structural, physical and controlled release properties are therefore needed. The originalities of this work lie in firstly the use of a biocompatible herbal extract for the development of controlled release sol-gel coated textiles, and secondly, the use of mathematical drug delivery models to describe the release kinetics and mechanism since it has been realized that controlled-release behaviors are sensitive to a variety of parameters, especially those concerning the nature of the delivery device.

The goal of this investigation was to prepare and evaluate the potential of sol-gel coating on cotton fabrics for tailoring the controlled release of curcumin. Other important factors relating to potential applications such as the appearance, weight, mechanical performance, and anti-bacterial properties of the material are also reported.

Experimental

Materials

A plain weave cotton fabric (mass per unit area of 76 g m^{-2}) having 108 yarns/inch in the warp and 88 yarns/inch in the weft direction was used. Curcumin (95% purity) was purchased from Changsha Kangrong Biological-Tech Co. Ltd. (Hunan, China). Tetraethyl orthosilicate (TEOS, >99%) and anhydrous sodium acetate (>99%) were supplied from Sigma–Aldrich (Germany). Analytical grade hydrochloric acid (35.4%)

vas obtained from Loba Chemie, Pvt. Ltd. (Mumbai, India). Absolute ethanol was ɔurchased from RCI Labscan Ltd. (Bangkok, Thailand). Non-ionic surfactant Sandoclean) was supplied by Clariant Ltd. (Bangkok, Thailand). Sodium hydroxide >99%) and glacial acetic acid were obtained from Merck (Darmstadt, Germany).

Preparation of sol-gel coated fabrics containing curcumin

The cotton fabrics were scoured using a solution containing 3 g/l non-ionic surfactant and 5 g/l NaOH at a liquor ratio of 30:1 at 90°C for 30 min. Silica sols were prepared by room-temperature mixing of TEOS, 0.01 M HCl, ethanol, and curcumin in various compositions according to Table 1. In the sample name, the notation C represents control samples (without TEOS) and T refers to treated samples (with TEOS). The numbers 10, 20 and 30 placed after T correspond to the amount of TEOS in millimoles. The numbers 30, 50, and 70 appearing after a hyphen represent the curcumin loading in milligrams. The total solution volume was fixed at 25 ml. All solutions were homogenized by sonication (VGT-2013HTD Portfolio) for 30 seconds giving clear, yellow solutions. Then, a 4cm × 10cm piece of cotton fabric was immersed in each solution for 10 minutes, taken out, and then padded at 2 kg cm^{-2} pressure (Labtec, Newave Lab. Equipments, Co., Ltd.). The % wet-pickups were calculated by measuring the fabric weights before, and immediately after padding. The treated fabrics were dried under ambient conditions for at least 48 h before final dry weights were recorded.

In vitro curcumin release study

A fabric sample size of 4cm × 5cm was used for the dissolution study. Prior to the experiment, all samples were immersed in 50 ml of ethanol and shaken in a water bath

shaker (Dye Plus DL-2003) for 1 h at 40°C, with a rotation speed of 60 rpm, to remove unbound curcumin. The release medium was 100 ml acetate buffer containing 3% v/v ethanol and 0.5% v/v Tween 80.[15] The sample was immersed in the release medium at 32±0.5°C in a closed glass vessel and shaken in the same water bath at 60 rpm. A 4 ml sample was collected, and this was replaced with fresh buffer solution of equal volume at 1, 2 or 4 h intervals (up to 24 h). The collected samples were kept refrigerated until analysis. The amount of released curcumin was determined from the absorbance values of the samples (UV-visible spectrophotometer, Perkin-Elmer Lambda 25) at 405 nm. All absorbance data points were in the linear range of the calibration curves with R^2 values of at least 0.98. To determine the actual amount of curcumin in each sample, curcumin extraction was done by shaking in release medium at 40°C, 60 rpm for 48 h, followed by 30-min sonication and mechanical squeezing. All experiments were done in triplicate.

Characterization of sample specific area and porosity

Nitrogen adsorption and desorption isotherms were obtained at 77K using a Quantachrome Autosorb iQ automated gas sorption system. The samples were outgassed at 80°C prior to measurement to remove all adsorbed water. Sample specific surface area was calculated based on the Brunauer-Emmett-Teller (BET) method in the relative pressure (*P/P0*) range of 0.05-0.30. Pore size distribution was analyzed from the desorption data using the conventional BJH method.

Morphological study

Surface morphology was examined using a scanning electron microscope (JEOL, JSM-5410LV) operating at an accelerating voltage of 15 kV and 100x magnification. All the samples were sputtered with gold prior to observation.

Mechanical testing

Testing of fabric mechanical properties was carried out using an Instron tensile tester (Instron 5567) according to ASTM D5035. A gauge length of 7.5 cm and a cross-head speed of 300 mm min^{-1} were used. An Elmendorf Tear Tester (Elmatear, James H. Heal & Co. Ltd) was used to determine the tear resistance of the samples following ASTM standard D 1424. All reported data were averaged across at least 5 measurements.

Antimicrobial testing

Samples were tested for their antibacterial activity according to AATCC Test Method 100-1999. All samples were sterilized by 15-min UV exposure on each side. Four circular swatches (4.8 cm in diameter) of the samples were inoculated with 1.0 ml of inoculums containing either 3.27×10^5 CFU of gram-positive *S. aureus* (ATCC 6538), or 5.25×10^5 CFU of gram-negative *E. coli* (ATCC 25922). After incubation (37°C, 24 h), the percentage reduction (*R*) in colony numbers in the treated samples compared to that of the untreated control was calculated using the following expression:

$$R(\%) = 100(A-B)/A$$

where *A* and *B* are numbers of bacteria recovered from the untreated and curcumin-treated fabric swatches, respectively, after inoculation and incubation.

Results and discussion

Sample weights and the calculation of related parameters: % wet pickup and % add-on

The parameter "*% wet pickup*" in textile finishing processes represents the amount of a finishing solution taken up by the fabric according to the following equation:[16]

$$\%wet\ pickup = \frac{wt.of\ wet\ fabric\ after\ pad - wt.of\ dry, untreated\ fabric \times 100}{wt.of\ dry, untreated\ fabric}$$

From Table 2, it can be seen that the % wet pickup values range from 73.4 to 86.9%. In general, the introduction of high-density TEOS increases the weight of a finishing solution and consequently the weight of the liquid carried by a cotton fabric. Normally, a higher % wet pickup implies an increase in the amount of chemicals deposited onto the fabric, which is described by another parameter, namely "*% add-on*". This is calculated as follows:[17]

$$\%\,add-on = \frac{wt.of\ dry, treated\ fabric - wt.of\ dry, untreated\ fabric \times 100}{wt.of\ dry, untreated\ fabric}$$

The % add-on values shown in Table 2 follow the same trend as % wet pickup, reflecting the weight gain of the fabrics due to the presence of both curcumin and silica coating.

Relationship between precursor concentration and percentage weight increase due to sol-gel coating

The percentage weight increase due to silica coating was calculated by excluding the adsorbed curcumin weight. As illustrated in Figure 2, the percentage weight increase was linearly dependent on the sol-gel precursor concentration. All data gave excellent correlation coefficients of 0.97-0.99, with a positive slope of 0.52. This suggests the possibility of estimating the final weight of the treated fabric if the amount of the precursor in the starting solution is known within the concentration range studied.

[insert Figure 2.]

Release profiles of curcumin from fabric samples

Figure 3(a) and (b) display the 24-h cumulative curcumin release profiles of the fabric samples (with three different curcumin loadings) as a function of immersion time and TEOS concentration. The cumulative release values were calculated as percentages based on the apparent surface area of the fabric (20 cm^2, from width × length). Although scoured cotton fabrics are generally good absorbents, their interactions with curcumin molecules involve weak intermolecular forces (van der Waals, hydrogen bonding), such that traditional curcumin-dyed cotton is prone to poor color fastness during laundering when no other chemical auxiliaries are applied. Consequently, samples without sol-gel treatment contained much lower adsorbed curcumin amounts at the beginning of the release experiment and thus released less curcumin than sol-gel treated samples. Sol-gel coating of cotton at high temperature[18] has been reported to improve color fastness since a porous sol-gel network, or xerogel, is formed which is capable of entrapping dye

molecules. According to our results, the use of more TEOS precursor results in a larger xerogel structure and therefore significantly higher amount of curcumin available for release. The silica coating on the fabric is assumed to be thicker from the increase in fabric weight with increasing TEOS concentration, as reported in Table 2. The experiment was carried out for 24 h at which point most of the release profiles were shown to level off.

[insert Figure 3.]

In Figure 4 cumulative curcumin releases, based on the total curcumin content, are plotted against immersion time (for brevity only samples containing 70 mg curcumin are shown). It was found that samples treated with TEOS generally release curcumin at lower percentages than the control samples. This again confirms that the presence of sol-gel porous coating results in enhanced curcumin entrapment compared to the untreated fabric surface, with 70-80%, and 50-60% release being observed for the controls and the sol-gel treated samples, respectively, after 24 h immersion. Curcumin release from samples with low TEOS content increases only marginally after 12 h, while samples with higher levels of TEOS show a more prolonged release profile.

[insert Figure 4.]

Release kinetics

The mechanisms of drug release from polymeric delivery systems can be broadly categorized into three major processes: diffusion-controlled, degradation-controlled, and swelling-controlled.[19] To date, it has proved impossible to derive a general mathematical

model that can describe the controlled release behaviors of all types of delivery systems. In some cases, these mathematical models are derived from theoretical analysis of the occurring process, while in other cases empirical equations have proved to be useful, at least for a comparison of different drug release profiles.[20,21] So far, several mathematical models have been reported to be applicable to sol-gel delivery materials, mostly those associated with diffusive release mechanisms. This suggests that different physicochemical phenomena underlie each specific case. These differences include the size and shape of the delivery device, the nature of the release substance and the interaction between the matrix and the release medium. In this work, the four models having found major applications in describing drug release profiles: the zero order, the first-order, the Higuchi, and the Hixson-Crowell models, were employed for the comparison of the release kinetics. The applicability of these models to our release system was also evaluated.

Zero order model. Zero order release kinetics refers to the process of constant drug release from a drug delivery device. The model is taken as the ideal delivery process and is represented by the following equation:[20]

$$Q_t = Q_0 + K_0 t$$

where Q_t is the amount of drug released in time t (expressed in %), Q_0 is the initial amount of drug in the solution (usually zero), and K_0 is the zero order release constant. Aughenbaugh *et al.* reported that the sol-gel delivery profiles may follow this model when the diffusive release is suppressed, such that the main release mechanism is

limited to surface degradation.[22] This type of release was also observed in sol-gel monoliths containing polysaccharides.[23]

First-order model. The first-order release model predicts a first-order dependence of drug release on the concentration gradient between the static liquid layer next to the solid surface (delivery device) and the bulk liquid (release medium). According to this model, there is no change in the solid surface area, i.e. no surface disintegration of the delivery device during the release process. The first-order release equation is:[9,20]

$$ln[1 - (Q_t/Q_\infty)] = -K_1 t$$

where Q_∞ is the cumulative amount of the substance released over an infinite time, and K_1 is the first-order release constant. As an example, this model renders a good fit for bupivacaine release from silica sol-gel granules, and vancomycin from silica sol-gel microspheres.[9]

Higuchi model. The Higuchi square root of time model can be used to describe the release of a solid drug from a porous matrix. Based on this model, the release process involves the penetration of the release medium, the dissolution of the drug, and leaching out of the drug through the interstitial channels or pores. The process is diffusion-controlled and dependent on the porosity and tortuosity of the release device:[20]

$$Q_t=[D\varepsilon/\tau(2A-\varepsilon C_s)C_s t]^{0.5}$$

The equation can be simplified as:

$$Q_t=K_2 t^{0.5}$$

From the above, D is the diffusion coefficient of the drug, ε is the porosity of the matrix, τ is the tortuosity factor of the capillary system, A is the total amount of the drug present in the matrix, and K_2 is the release rate constant. This model was found to be applicable to many sol-gel delivery systems, including sol-gel silica microspheres and monoliths.[9,23,24]

Hixson-Crowell model. The Hixson-Crowell cube root model describes the drug release from systems where the geometrical shape of the release matrix diminishes proportionally over time (erodible matrix). The release rate is limited by the matrix dissolution rate, not by the diffusion of drug through the polymeric matrix as shown in the following expression:[20]

$$Q_0^{1/3}-Q_t^{1/3}=K_3t$$

where K_3 is the rate constant. This model has been tested with sol-gel nanoparticle drug delivery systems but was found to be inadequate.[25]

How well a particular model can describe the experimental data is reflected in the correlation coefficient value (R^2), which should be close to 1. However, it should be noted that, in practice, there is difficulty in discriminating between competing models.[26,27]

The curcumin release data were plotted according to the aforementioned models for the first ten hours of release. According to the literature, at levels beyond 50% release significant deviations from the models may occur.[9] The rate constants and corresponding correlation coefficients of the fabric samples are listed in Table 3.

Interestingly, all four models can describe the release profiles of both sol-gel treated, and untreated samples at R^2>0.90 but overall, the quality of fit is slightly better for the Higuchi model and the first-order model (R^2>0.95). The rate constants for every model are consistent: the samples without sol-gel treatment release curcumin faster than treated samples. Overall, sol-gel coating retards the release rate by about 30%. Radin *et al.* have demonstrated that silica sol-gel microspheres provide drug (bupivaciane and vancomycin) release following both the Higuchi, and the first-order model. The applicability of more than one model to a certain release system is not surprising. It is worth noting that these two models share key assumptions: the release process is purely-diffusion controlled with no matrix swelling or erosion and perfect sink conditions are maintained.[28] These two expressions also imply decreased drug release with time, which can be explained based on the increase in diffusion path length as the degree of medium penetration, and drug extraction, proceed. Matrix swelling during immersion can be ruled out as the inorganic silica xerogel is typically rigid.

To verify that there is no dissolution of silica matrix in our experimental regime, the amount of silicon (in the form of silicic acid, $Si(OH)_4$) leached out of the sol-gel treated samples after 24 h immersion was determined using the heteropoly blue method.[29] The release medium was digested with $NaHCO_3$ to convert unreactive silicon compounds, if present, into the molybdate-reactive form suitable for determination. Results show that no silica matrix dissolution occurred during 24 h, confirming that the curcumin release kinetics of the sol-gel coated fabrics are mainly diffusion-controlled as predicted by the first-order and Higuchi models. It is known that silica can undergo hydrolysis to form silicic acid and the hydrolysis rate would be greatly increased under

alkaline conditions. For nonporous amorphous silica, the equilibrium concentration of silicic acid is only 70 ppm at 25°C, and in this case silica solubility is therefore negligible.[30]

The comparable rate constants for most sol-gel coated samples can be explained by their identical chemical compositions and the homogeniety of the curcumin-embedded sol-gel coating. The coating thickness variation, due to different amounts of precursor, did not appreciably change either the release mechanism or the release kinetics. Although release is expected to be faster with greater curcumin loading, these effects were not noticeable under our experimental conditions (which was restricted due to the solubility limit of curcumin).

Specific surface area and porosity

The N_2 gas adsorption isotherms of untreated cotton and T30-30 are displayed in Figure 5(a). Based on the IUPAC classification,[31] the adsorption isotherm of cotton is of type II, being characterized by an intermediate plateau followed by a continuous rise at a relative pressure close to unity (unrestricted monolayer-multilayer adsorption). This type of isotherm is generally observed in non-porous, or macroporous materials. Cotton textiles contain numerous pores including intra-fiber, inter-fiber, intra-yarn and inter-yarn pores. Their pore sizes depend on fiber size, type of weave, the number of yarns per inch and the presence of yarn twists. The inter-fiber and inter-yarn pore sizes lie in a range consistent with those of macropores, which are considered too large (> 50 nm) to be reliably measured using the nitrogen gas adsorption technique. It has been reported that the inter-fiber pore sizes in woven cotton are between 2.0-4.5 μm[32], while those of

inter-yarns are certainly larger. In this study, the BJH method employed is suitable for the calculation of sizes (2-50 nm) spanning the mesopore to small macropore range only. Since the aim is to observe the change in porosity caused by the silica xerogel coating, the measurement of larger macropores (which is possible by mercury intrusion porosimetry) associated with the textile structure was not carried out. A lack of hysteresis in the desorption branch indicated the absence of mesopores, and the calculation of BJH pore size distribution of the untreated cotton was therefore unachievable. The T30-30 isotherm appears similar to that of cotton but with a small hysteresis loop characteristic of a type IV isotherm. This hysteresis loop suggests the presence of mesopores which clearly originate from the xerogel structure. The total pore volume was estimated from the amount of nitrogen adsorbed at a relative pressure of 0.99; the values obtained are very low, i.e. 0.003 $cm^3 g^{-1}$ for the untreated sample and 0.004 cm^3 g^{-1} for T30-30, respectively. A slightly higher pore volume in T30-30 was attributed to the open mesopores present in the xerogel structure. Based on the BJH calculation from the desorption isotherm, the mean diameter of these mesopores is 14.3 nm. The BJH pore size distribution of T30-30 is shown in Figure 5(b). An assessment of the micropore volume (pore size < 2 nm) using the t-plot method indicated that no micropores existed in either sample.

[insert Figure 5.]

Regarding the determination of sample specific surface area, the BET method was applied to the adsorption isotherm at the relative pressure between 0.05-0.30. The BET specific surface area of the untreated cotton was 1.95 m^2 g^{-1}. For the sol-gel coated cotton, the specific area increased slightly to 2.23 m^2 g^{-1}. Both results were

obtained with excellent correlation coefficients greater than 0.99. Similar values have been reported in another sol-gel coated cotton system.[17]

The higher pore volume and surface area in the treated samples are a consequence of the porous sol-gel structure. However, the difference between treated and native cotton was only small, suggesting that the extent of fabric sol-gel coating was not high. This is preferable when one wants to preserve the overall characteristics of the original textile in the fabricated material.

Because of the greater porosity creating a more tortuous pathway for any fluid to diffuse in or out of the xerogel structure, after immersing in alcoholic solution coated samples retain a greater number of curcumin molecules than the control (Figure 6). This is highlighted by the yellow color being stronger in fabrics having higher TEOS, and curcumin concentrations. This can be expected since curcumin is adsorbed only weakly onto cotton fibers. A large proportion of the curcumin molecules leach out of the uncoated cotton fabric soon after immersion, leading to poor color fastness to washing. This process can be slowed down by entrapping them within the sol-gel network. When the coated fabrics are used in wound dressing applications, the effective residence period of the biomedically active compound can be extended. The amount of the active substance as well as the release duration can be tailored easily by adjusting the sol-gel processing parameters (the concentration of the sol-gel precursor, for instance).

[insert Figure 6.]

Sample morphology

SEM micrographs were taken from samples containing 0.07 g curcumin for a comparison of their morphological properties. As displayed in Figure 7, the C-70 sample shows fibers having a smooth surface, with twists and convolutions characteristic of cotton. Homogenous curcumin adsorption on the cotton fiber was obtained, with no indication of curcumin agglomeration. In our study, the sol-gel coatings do not form bridges between adjacent fibers, which is desirable as air and vapor permability is required for their potential application as wound dressings. This indicates that the coatings are thin enough such that the open structure of the fabrics is not closed up on fabrication of the material.

[insert Figure 7.]

Mechanical properties

Light weight and high flexibility are not the only important features for functioning textile materials; having mechanical properties suitable for the targeted application is also required. In this research, the tensile strength, percentage elongation at break, and tearing force for the treated fabrics were investigated. As seen in Figure 8(a), tensile strength in the warp direction of the fabrics was not altered by sol-gel coating. In the weft direction, coating with curcumin and sol-gel increases the tensile strength from a low to moderate degree (increased by 7% for C-30, and 52% for T30-50). Therefore, the sol-gel network can help dissipate the tensile force exerted upon the fabrics, promoting resistance to tensile deformation. According to Figure 8(b), a decrease of percentage elongation at break was observed with increasing TEOS concentration in both warp and

weft directions, with the effect more significant in the former as a result of increased stiffness imparted by the inorganic silica coating. The slight increase in tensile strength, and a smaller elongation at break were also reported for other sol-gel coated cotton fabrics.[33]

As expected, the sol-gel coating of fabrics resulted in lower tear resistance (Figure 8(c)). This can be explained partly by a reduction in yarn slippage. The silica network present on the fiber surface increases the fiber surface friction and, as a consequence, prevents yarn slippage. Since the tearing energy cannot be dissipated efficiently with poor yarn slippage, the tear propagation is then more pronounced. This results in the lowered tearing strength typically observed in sol-gel coated fabrics.[33,34] Our results reveal that the reduction in tear resistance is only slight; the sample with the lowest tearing strength (T30-30) shows about 15% reduction in the warp direction. From the SEM images, there is no interlocking between the cotton fibers arising from the silica network, therefore the fibers can still be displaced to a reasonable degree.

[insert Figure 8.]

Antibacterial activity

Curcumin is an effective colorant for cotton and can also provide potential antibacterial activity to fabrics. As shown in Table 4, the sol-gel coating results in higher antibacterial activity in the treated fabrics over the control. The fabrics without sol-gel treatment revealed very low antibacterial activities against *E.Coli* and no activity against *S.aureus*, indicative of a very low concentration of curcumin being present. However, on sol-gel treatment, the fabrics showed improved antibacterial effects against both strains. The

percentage reduction in the bacteria counts for *S.aureus* increased rapidly with increasing curcumin concentration, from 0 to 99.41% with the addition of 30-70 mg of curcumin in samples treated with 30 mmol TEOS. On the other hand, values against *E.Coli* increased only slightly from 28.44 to 37.03% for the same sample set. Similar observations were found with the curcumin-dyed cotton: curcumin was more effective in inhibiting *S. aureus* than *E. coli.*[35]

Conclusions

This work investigated the coating of cotton fabrics with an acid-catalyzed sol-gel and curcumin for potential application in controlled release wound dressings. Prolonged and more efficiently controlled curcumin release, as well as antibacterial properties can be realized via sol-gel coating of fabrics. A study of the release kinetics revealed that the release process is diffusion-controlled, with gas sorption analysis confirming the ability of the sol-gel coating to retain curcumin molecules. The coated fabrics exhibit reduced ductility but their strength is mostly preserved. Overall, the application of sol-gel coating with the release of nontoxic herbal substances such as curcumin may result in the eventual application of these textiles as next generation, controlled-release wound dressings.

Funding

This work was supported by the Thailand Research Fund (TRF); The Office of the Higher Education Commission (OHEC); and Thammasat University [grant number MRG5580207].

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Table 1. Composition of starting solutions used in preparing the fabric samples

Sample label	TEOS (mmol)	Curcumin (mg)	0.01 M HCl (ml)	EtOH (ml)
C-30	-	30	7.50	17.50
T10-30	10	30	7.50	15.25
T20-30	20	30	7.50	12.50
T30-30	30	30	7.50	10.75
C-50	-	50	7.50	17.50
T10-50	10	50	7.50	15.25
T20-50	20	50	7.50	12.50
T30-50	30	50	7.50	10.75
C-70	-	70	7.50	17.50
T10-70	10	70	7.50	15.25
T20-70	20	70	7.50	12.50
T30-70	30	70	7.50	10.75

Table 2. Sample percentage wet-pickups and percentage add-ons

Sample label	% wet pickup	% add-on
C-30	73.4±4.0	0.2±0.1
T10-30	82.4±1.1	5.1±0.6
T20-30	85.2±2.2	9.1±0.4
T30-30	84.9±3.3	12.8±0.2
C-50	74.8±4.0	0.7±0.2
T10-50	82.9±4.0	5.0±1.4
T20-50	83.1±3.2	9.4±0.2
T30-50	86.9±1.3	13.4±0.7
C-70	78.6±2.7	1.0±0.2
T10-70	83.0±5.9	5.0±0.4
T20-70	88.0±3.1	10.1±0.1
T30-70	85.6±3.4	13.0±0.2

Table 3. Release rate coefficients, and their corresponding R^2 values derived from four different kinetic models

Sample	Zero-order		First-order		Higuchi		Hixson-Crowell	
	K_0 (% h^{-1})	R^2	K_1 (h^{-1})	R^2	K_2 (% $h^{-0.5}$)	R^2	K_3 (% h^{-1})	R^2
C-30	4.530	0.96	0.090	0.98	19.376	0.98	0.110	0.98
T10-30	2.948	0.96	0.044	0.98	12.689	1.00	0.060	0.97
T20-30	2.777	0.97	0.043	0.98	12.226	0.95	0.057	0.90
T30-30	2.947	0.95	0.042	0.95	12.497	0.95	0.058	0.95
C-50	4.826	0.99	0.082	0.98	21.764	0.96	0.106	0.99
T10-50	3.671	0.99	0.064	1.00	16.860	1.00	0.082	1.00
T20-50	4.254	1.00	0.067	0.99	17.758	0.97	0.089	0.99
T30-50	3.36	0.97	0.047	0.98	14.271	0.98	0.065	0.98
C-70	4.819	0.93	0.082	0.96	18.917	0.97	0.105	0.94
T10-70	3.215	0.95	0.058	0.97	13.683	0.96	0.073	0.96
T20-70	3.033	0.97	0.045	0.97	12.723	0.95	0.061	0.97
T30-70	2.889	0.93	0.042	0.97	12.440	0.96	0.057	0.93

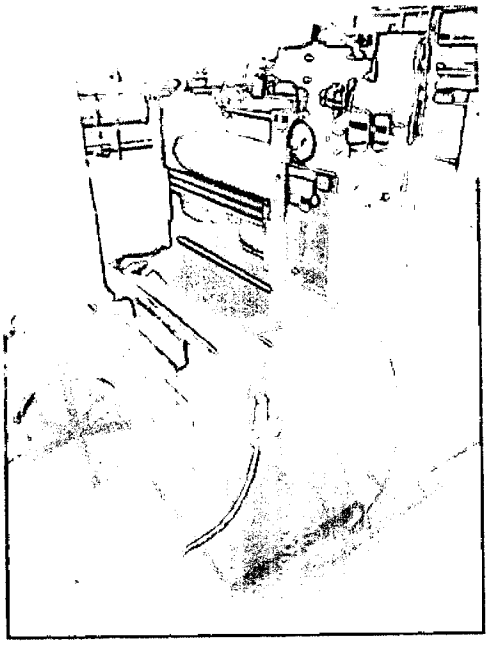
Table 4. Percentage reduction in bacterial levels as determined according to AATCC 100-1999

Sample	Bacteria reduction percentage (%)	
	Staphylococcus aureus	*Escherichia coli*
C-30	0	5.83
C-50	0	16.88
C-70	0	0.83
T30-30	0	28.44
T30-50	38.23	30.60
T30-70	99.41	37.02

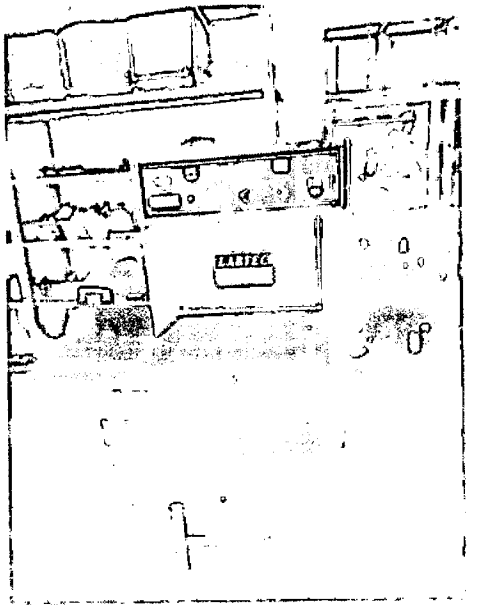
ภาคผนวก ค

เครื่องมือวิทยาศาสตร์ภายในห้องปฏิบัติการ

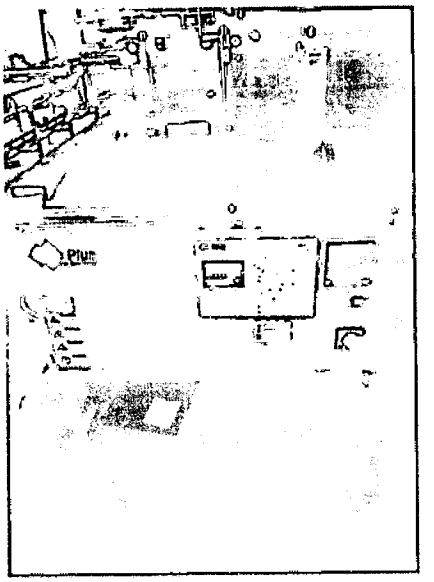
1. เครื่องบีบอัดผ้า (Padding mangle)



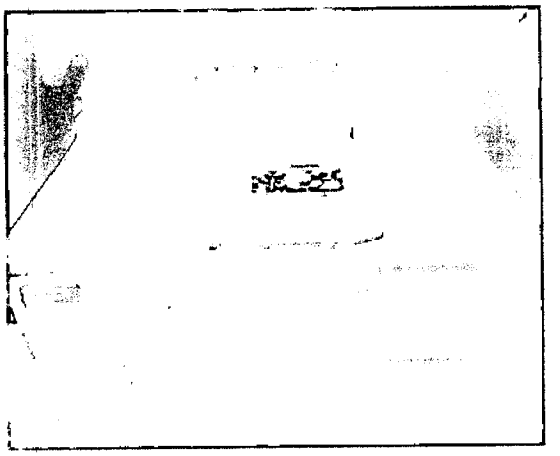
2. เครื่องอบแห้ง (Laboratory mini-dryer)



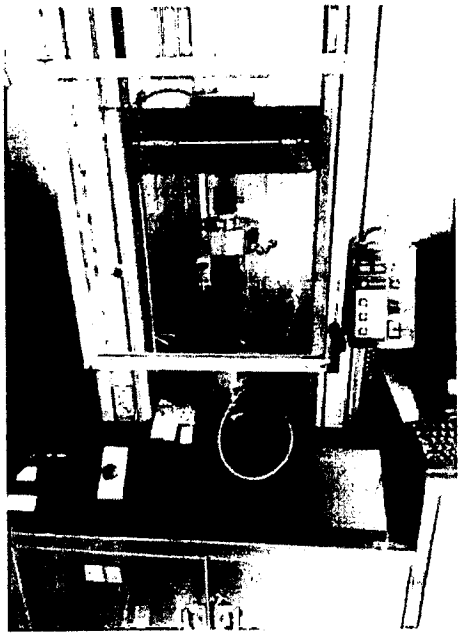
3. อ่างน้ำควบคุมอุณหภูมิ (Water Bath shaker)



4. เครื่องวัดการดูดกลืนคลื่นแสง (UV-Visible Spectrophotometer)



5. เครื่องวัดความต้านทานการดึงยืด



6. เครื่องวัดความต้านทานการฉีกขาด