Mass Transfer During the Pre-Usage Dehydration of Polyvinyl Alcohol Hydrogel Wound Dressings

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Abstract

Nowadays, hydrogels are widely used as wound dressings in biomedical applications. Similar to other types of the moist (wet) wound dressings, it is necessary to have sufficient information about their dehydration kinetics during the pre-usage period (e.g. storage duration) and also the wound healing process. In this work, hydrogel wound dressings based on polyvinyl alcohol were prepared by cyclic freezing-thawing method and their dehydration process investigated by experimental and mathematical methods. The dehydration tests were performed at atmospheric pressure at 37 °C, using hydrogels containing initial water content of 70, 80 and 90 wt\% and thickness of 1, 3 and 4 mm. The dehydration kinetics of each hydrogel was determined by plotting the relative water loss (ratio of water loss to initial water content) versus time. The obtained results showed that the dehydration rate has inverse dependency to the thickness of wound dressing. On the other hand, the initial water content of the wound dressing has not significant effect on its dehydration rate. To compare the obtained results with theoretical data, the mathematical modeling on the basis of diffusion mechanism was developed to predict the dehydration process of the wound dressings. The results obtained from the mathematical modeling were in agreement with the experimental results showing that the main phenomenon governing the dehydration of the wound dressings is the diffusion.

Keywords: Dehydration; Hydrogel; Mass transfer; Polyvinyl alcohol; Wound dressing.

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1. Introduction

Rapid and proper healing is important in the treatment of wounds. In cases of severe and large amounts of skin loss, immediate coverage of the wound surface with a dressing is needed \cite{1}. Wound dressings are usually used to encourage various stages of wound healing and create better healing conditions.
They often cover the wound surface to accelerate its healing [2]. Based on the types of wounds and modes of healings, numerous materials are developed for use as wound dressing [3, 4]. Among the wound dressings, special attention has been paid to hydrogels because of their unique interesting properties which can meet the essential requirements of ideal wound dressings including: immediate pain control, easy replacement, transparency to allow healing follow up, absorb and prevent loss of body fluids, barrier against bacteria, oxygen permeability, good handling, and control of drug dosage, etc. [5].

Polyvinyl alcohol (PVA) hydrogel is one of the well-known polymer gels that due to its good biocompatibility has been used in numerous biomedical applications, for example as wound dressings in wounds management [2, 6-9]. There is a number of methods for crosslinking PVA chains in order to produce PVA hydrogels, including electron beam irradiation [10], bulk mixing with crosslinking agents such as glutaraldehyde and also freezing-thawing cyclic process [11-13]. In an ideal condition, a desirable wound dressing should create and keep a moist environment on the wound surface. Therefore, having enough information about the dehydration kinetics of the wound dressing during healing process and pre-usage duration is necessary.

In this work, the hydrogel wound dressings based on PVA were prepared by the freezing-thawing process. The dehydration kinetics of the prepared hydrogels during the pre-usage period was investigated either by the experimental method and mathematical modeling.

2. Materials and methods
2.1. Chemicals
All experiments utilized commercial grade PVA having a degree of polymerisation of 1700 and a saponification value of greater than 98%, was purchased from the Nippon Synthetic Chemical Industry Co., Ltd. Japan. Double distilled water (DDW) was used to prepare all aqueous solutions.

2.2. Preparation of PVA hydrogel wound dressings
PVA hydrogels were prepared by repeatedly cyclic freezing and thawing process. For this purpose, aqueous solutions containing 15% PVA were used. These solutions were mixed slowly and heated up to 90 °C for a period about 4h to achieve complete dissolution. Then the aqueous solutions were poured into plastic moulds and placed at -20 °C for 24h to induce crystallisation. After the freezing process, they were subsequently allowed to thaw for 24h at 23 °C. This cyclic process was repeated three times for each solution. The hydrogel films had various thicknesses of 1, 3, and 4 mm.

Dehydration tests were performed in an oven with constant temperature of 37 °C and humidity about zero (dry air) by weighting the hydrogels at some predetermined time intervals.

3. Mathematical modeling
The modeling of the dehydration process of the hydrogel wound dressings was performed according to the diffusion model. For this purpose, we consider a sheet of hydrogel with thickness of 2b (Figure 1) having the initial water concentration of $C_0$ which is exposed to the environment (air) with water concentration of $C_a$. If the concentration of the water inside the hydrogel is greater than its value in air ($C_0>C_a$), then the water will diffuse from the hydrogel into the environment due to its concentration gradient.

The dehydration kinetics (water release) of the hydrogel wound dressings can be determined by using the second Fick's law. The system (hydrogel) is symmetric and
therefore we can consider a half of system (0 ≤ z ≤ b). Therefore, we must solve the below partial differential equation (Equation 1) with the governing initial and boundary conditions (Equations 2-4):

\[
\frac{\partial C(z,t)}{\partial t} = D \frac{\partial^2 C}{\partial z^2} \quad 0 \leq z \leq b \quad \text{Equation (1)}
\]

\[
C(z,0) = C_0 \quad \text{Equation (2)}
\]

\[
\frac{\partial C(0,t)}{\partial z} = 0 \quad \text{Equation (3)}
\]

\[-D \frac{\partial C(b,t)}{\partial z} = K[C(b,t) - C_a] \quad \text{Equation (4)}
\]

where, D and K are the diffusion and mass transfer coefficients of the water in the hydrogel and air, respectively. To solve the above initial and boundary value problem, a new function was defined as follows:

\[
\Theta(z,t) = C(z,t) - C_a \quad \text{Equation (5)}
\]

Inserting Equation 5 into Equations 1-4 yields:

\[
\frac{\partial \Theta(z,t)}{\partial t} = D \frac{\partial^2 \Theta}{\partial z^2} \quad 0 \leq z \leq b \quad \text{Equation (6)}
\]

\[
\Theta(z,0) = C_0 - C_a \quad \text{Equation (7)}
\]

\[
\frac{\partial \Theta(0,t)}{\partial z} = 0 \quad \text{Equation (8)}
\]

\[-D \frac{\partial \Theta(b,t)}{\partial z} = K[\Theta(b,t)] \quad \text{Equation (9)}
\]

The solution of the Equation 6 can be determined using the method of separation of variables (product method) as follows:

\[
\Theta(z,t) = (C_0 - C_a) \sum_{i=1}^{\infty} A \cos\left(\frac{\beta_i z}{b}\right) \exp\left(-\frac{\beta_i^2 D t}{b^2}\right) \quad \text{Equation (10)}
\]

where, A is defined according to the Equation 11 and \(\beta_i\) are the roots of the Equation 12:

\[
A = \frac{2 \sin \beta_1}{\beta_1 + (\sin \beta_1)(\cos \beta_1)} \quad \text{Equation (11)}
\]

\[
\beta_1 \tan \beta_1 = \frac{bK}{D} \quad \text{Equation (12)}
\]

Combining Equations 5, 10 and 11 gives the concentration of the water inside the system as a function of time (t) and position variable (z), as follows:

\[
\frac{C(z,t) - C_a}{C_0 - C_a} = \sum_{i=1}^{\infty} \left(\frac{2 \sin \beta_i}{\beta_i + (\sin \beta_i)(\cos \beta_i)}\right) \cos\left(\frac{\beta_i z}{b}\right) \exp\left(-\frac{\beta_i^2 D t}{b^2}\right) \quad \text{Equation (13)}
\]

The rate of the water released from the system into the environment can be determined using the first Fick's law:

\[
\frac{dM_t}{dt} = -S \frac{dC}{dz} \bigg|_{z=b} \quad \text{Equation (14)}
\]

where, S is the surface area of the system. Inserting Equation 13 in 14 and integrating from 0 to t yields the amount of water released from the system up to time t (M_t) as follows:

\[
M_t = kS \left(\frac{2}{\beta_1 + (\sin \beta_1)(\cos \beta_1)}\right) \left(1 - \exp\left(-\frac{\beta_1^2 D t}{b^2}\right)\right) \quad \text{Equation (15)}
\]

Finally, the fraction of the released water from the system at the given time of t can be determined using the below Equation:

\[
\frac{M_t}{M_a} = \sum_{i=1}^{\infty} \left(\frac{C_0 - C_a}{C_0 - C_a}\right) \left(\frac{2 \sin \beta_i}{\beta_i + (\sin \beta_i)(\cos \beta_i)}\right) \left(1 - \exp\left(-\frac{\beta_i^2 D t}{b^2}\right)\right) \quad \text{Equation (16)}
\]
where, \( M_\infty \) is the amount of the initial water inside the half of the system, which can be determined according to the below Equation:

\[
M_\infty = \text{BC}_0
\]  
Equation (17)

4. Results and discussion

The effects of the thickness and the initial water content of the PVA hydrogel wound dressings on their dehydration kinetics were investigated either by the experimental and mathematical methods. The mathematical modeling results were obtained using the values of \( 10^{-10} \) (m²/s) and \( 10^{-7} \) (m/s) for \( D \) and \( K \), respectively.

Figure 2 shows the dehydration kinetics of the PVA hydrogel wound dressings for various amounts of the hydrogel thicknesses, either by the experimental and mathematical methods. In the Figure 2, the \( M_t/M_\infty \) curves were plotted against the time of dehydration for different samples having different thicknesses (based on Equation 16) and compared with the obtained experimental results. Figure 2 demonstrates a good agreement between the experimental and mathematical results and reveals that the main phenomenon governing the dehydration of the wound dressings is the diffusion. In Figure 2 we can also see by both experimental and mathematical results that the dehydration rate has inverse dependency to the thickness of wound dressing, in a manner which increasing the thickness of the wound dressing causes to decrease the fraction of water released from it at a specific time.

Figure 3 shows the \( M_t/M_\infty \) values versus time for the PVA hydrogel wound dressings which were obtained by the experimental and mathematical methods. Figure 3 reveals an excellent agreement between the experimental and mathematical results and confirms this idea that the main phenomenon governing the dehydration of the wound dressings is the diffusion. Figure 3 also shows that the initial water content of the wound dressing has not significant effect on its dehydration rate.

5. Conclusion

In this work, the dehydration kinetics of the PVA hydrogel wound dressings were studied by both experimental and mathematical methods and the effects of the thickness of the wound dressing and its initial water content amount on the dehydration process investigated. The results showed that the dehydration rate of the PVA hydrogel wound dressing has inverse dependency to its thickness. On the other hand, the initial water content of the wound dressing has not significant effect on its dehydration rate. The results obtained from the mathematical modeling (based on diffusion model) were in agreement with the measured experimental results showing that the main phenomenon governing the dehydration of the PVA hydrogel wound dressings is the diffusion.
References


