

Sains Malaysiana 46(4)(2017): 623–628
<http://dx.doi.org/10.17576/jsm-2017-4604-15>

Preparation and Characterization of Fe_3O_4 /Regenerated Cellulose Membrane (Penyediaan dan Pencirian Membran Fe_3O_4 /Selulosa Terjana Semula)

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ABSTRACT

In this study, magnetic cellulose membranes (MCM) have been prepared by using cotton linter as cellulose source and NaOH/urea as cellulose solvent at different magnetite content. Cellulose was dissolved in pre-cooled NaOH/urea solvent at -13°C to form cellulose solution. The cellulose solution then was mix with magnetite (Fe_3O_4) nanoparticles synthesized via co-precipitation method of Fe^{2+} and Fe^{3+} in the presence of sodium hydroxide (NaOH) to form MCM. The MCMs formed at different percentage of Fe_3O_4 i.e., 10, 20 and 30%. Analysis from vibrating sample magnetometer (VSM) shows that the saturation magnetization of the MCM increase as the percentages of Fe_3O_4 nanoparticles increased. However, the addition of Fe_3O_4 nanoparticles in the regenerated cellulose membrane has decreased the crystallinity index of MCM. The surface morphology of the MCM showed that the Fe_3O_4 nanoparticles were dispersed in the pore of the membrane. Tensile test showed decreasing in the tensile strength of the cellulose membrane with the addition of Fe_3O_4 nanoparticle.

Keywords: Dissolution; Fe_3O_4 ; magnetic cellulose membrane; pre-cooled

ABSTRAK

Dalam kajian ini, membran selulosa bermagnet (MSM) telah disediakan dengan menggunakan linter kapas sebagai sumber selulosa dan NaOH/urea sebagai pelarut selulosa pada kandungan magnetit yang berbeza. Selulosa dilarutkan dalam pelarut NaOH/urea yang telah dilakukan pra-penyejukan pada suhu -13°C untuk menghasilkan larutan selulosa. Larutan selulosa tersebut kemudiannya dicampurkan dengan nanozarah magnetit (Fe_3O_4) yang disintesis melalui kaedah pemendakan Fe^{2+} dan Fe^{3+} dengan kehadiran natrium hidroksida (NaOH) untuk menghasilkan MSM. MSM yang terhasil mempunyai peratusan Fe_3O_4 yang berbeza iaitu, 10, 20 dan 30 %. Analisis daripada magnetometer getaran sampel (VSM) menunjukkan kemagnetan tepu bagi MSM bertambah apabila peratusan zarah magnetit meningkat. Walau bagaimanapun, penambahan zarah magnetit dalam membran selulosa terjana semula telah menjatuhkan indeks penghabluran MSM. Morfologi permukaan MSM menunjukkan bahawa zarah magnetit tersebar di atas liang membran tersebut. Ujian tegangan menunjukkan pengurangan dalam kekuatan tegangan membran selulosa dengan penambahan nanozarah magnetit.

Kata Kunci: Fe_3O_4 ; membran selulosa bermagnet; pelarutan; pra-penyejukan

INTRODUCTION

Cellulose ($\text{C}_6\text{H}_{10}\text{O}_5$) is a biopolymer material which is the most readily available resource on Earth (Zhang et al. 2010). It is a valuable source of energy, renewable, biodegradable and bio-compatible which produced from natural resources up to 1 trillion tonnes per year (Zhang et al. 2010). Cellulose is hydrophilic in nature and tends to absorb high proportion of water due to the large number of hydroxyl groups. The great amount of intermolecular hydrogen bonds in cellulose chains can be broken and modified for various purposes (Luo & Zhang 2010a; Ruan et al. 2004; Zhang et al. 2005). The hydroxyl groups in cellulose are the main focus in the modification of cellulose to be used in the formation of regenerated cellulose materials (Chen et al. 2006; Yang et al. 2001).

Cellulose dissolution process involves the breaking of inter and intra hydrogen bonds between the cellulose chains and destruction of crystals along the cellulose

chains to produce cellulose solution (Luo & Zhang 2010a; Ruan et al. 2004; Zhang et al. 2005). An eco-friendly solvent has been introduced which consists of alkaline (LiOH or NaOH) and urea which able to dissolve cellulose at low temperature and shorter time (Zhang et al. 2010). This solvent offers an inexpensive and less toxic system as compared to other solvents developed previously (Jin et al 2007; Liang et al. 2007). The dissolved cellulose can then be further applied for the preparation of regenerated cellulose products such as hydrogel (Chang et al. 2010; Kaco et al. 2014), films (Li et al. 2012), membranes (Kaco et al. 2015; Mohd Saidi et al. 2016) and beads (Gericke et al. 2013; Jin et al. 2007; Liang et al. 2007). The process of cellulose dissolution using different cellulose solvents provides a new method for the production of regenerated cellulose and can be used in various applications. The cellulose morphology changed after the dissolution and regeneration processes

which affected the properties of the regenerated cellulose produced (Mohd Saidi et al. 2016; Stepanik et al. 1998). Recently, hybrid composite materials have attracted more interest to many researchers in order to achieve excellent properties as compared to the parent material. This material possesses superior mechanical, magnetic, catalytic, optical and electrical properties (Yu et al. 2014). Magnetic-polymer carbohydrate such as cellulose has raised more attention due to the inherent properties of the substrate which is cellulose as well as the bonded particles exhibit magnetically responsive properties (Wu et al. 2011). Magnetic and hybrid magnetic composites materials are important materials which can be used for numerous applications including gas and magnetic sensors, electrodes, information storage, magnetic films and filtration and purification in water treatment process (Wu et al. 2011; Yu et al. 2014). Furthermore, technology involving cellulose with the addition of magnetite particles is environmentally friendly and can be used in the human body for drug delivery and biomedical products (Galland et al. 2013; Sivashankar et al. 2014).

Magnetite (Fe_3O_4) nanoparticles can be synthesized using aqueous or organic solutions method via chemical precipitation reactions (Wang et al. 2010), surface coating (Small & Johnston 2009) and *in-situ* synthesis (Munawar et al. 2010). On the other hand, the method of producing magnetic cellulose composites involves the method of lumen loading (Chia et al. 2008; Zakaria et al. 2005) and *in-situ* synthesis method (Wu et al. 2011). Lumen loading method on cellulose pulp involves the employment of coating powders and magnetic powders to be promoted into cellulose pulp resulting the external surface cleaned from any filler (Zakaria et al. 2005). The introduction of fillers (magnetite and other filler) in the lumen allows the inter-fiber bonding between the cellulosic fibers, hence, it does not interfere on the fiber surface (Chia et al. 2008; Munawar et al. 2010; Zakaria et al. 2005). On the contrary, both matrices and particles will co-deposits at the same time from the mixed precursor. The formed composites possess homogeneous and uniform properties (Wang et al. 2010). For *ex-situ* method, the hybrid composites produce by dispersing the inorganic nanoparticles in an organic solution. The biocompatible magnetic cellulose-chitosan hybrid gel microspheres have been fabricated from ionic liquids using *ex-situ* method (Liu et al. 2012; Yu et al. 2014).

In this study, cellulose was dissolved in NaOH/urea solvent to produce cellulose solution via pre-cooled method. The *ex-situ* synthesis of Fe_3O_4 nanoparticle were dispersed in cellulose solution and the effect of different loading of Fe_3O_4 in the system toward the physical and mechanical properties were studied. The samples were analyzed using X-ray diffraction (XRD), vibration sample magnetometer (VSM), scanning electron microscope (SEM) and tensile machine.

MATERIALS AND METHODS

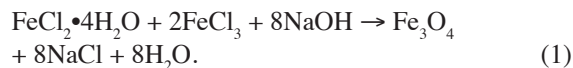
Cotton linter with viscosity average molecular weight (M_v) 9.0×10^4 was supplied by Hubei Chemical Fiber Co., Ltd. (Xiangfan, China) was used throughout the experiment. The pulp was ground using a blender and dried in an oven at 105°C for 24 h. The dried cellulose was kept in the desiccator until further used. Iron (II) chloride tetrahydrate ($\text{FeCl}_2 \cdot 4\text{H}_2\text{O}$), iron (III) chloride (FeCl_3), sodium hydroxide (NaOH), urea were supplied by System and sulphuric acid (H_2SO_4) was supplied by Sigma Aldrich. The chemicals were used as received.

PREPARATION OF CELLULOSE SOLUTION

Cellulose solution was prepared by dissolving 7 wt. % of NaOH and 12 wt. % of urea in 81 wt. % of distilled water and the solvent was pre-cooled at -13°C . The pre-cooled solvent was thawed and 4 wt. % of cellulose was dissolved in the solvent under vigorous stirring for 5 min using a mechanical stirrer until cellulose solution was formed. The cellulose solution was centrifuged at 8000 rpm at 5°C for 5 min to remove bubbles.

SYNTHESIS OF MAGNETITE NANOPARTICLES (Fe_3O_4)

Magnetite nanoparticles was synthesized via chemical co-precipitation method by using 9.94 g of $\text{FeCl}_2 \cdot 4\text{H}_2\text{O}$, 16.2 g of FeCl_3 and 16 g of NaOH with the molar ratio of $\text{Fe}^{2+}:\text{Fe}^{3+}=1:2$ and addition of 8 mol of NaOH and was dissolved in distilled water. The reaction was carried out under vigorous stirring at 60°C with the addition of nitrogen gas for degassing purposes and to prevent the oxidation of Fe^{2+} . The solution turned into black color after the addition of NaOH which indicates the formation of Fe_3O_4 . The solution was washed few times with distilled water to remove excess unreacted chemical. Then 1 mL of precipitated Fe_3O_4 was weighed and dried to obtain the dry weight of magnetite. The reaction mechanism of the process was represented in the following equation (Chia et al. 2006):



PREPARATION OF CELLULOSE AND MAGNETIC CELLULOSE MEMBRANE

The transparent cellulose solution was cast on a glass plate and coagulated in an acid bath containing 5% H_2SO_4 to form cellulose membrane (CM). The cellulose membrane was then washed with distilled water several times to remove excesses chemical.

In order to form magnetic cellulose membrane (MCM), the synthesized Fe_3O_4 was added into the cellulose solution and was stirred homogeneously. The cellulose solution containing Fe_3O_4 particles was cast on a glass plate and coagulated in 5% H_2SO_4 to form MCM. MCM

was produced at different Fe_3O_4 loading, i.e. 10, 20 and 30 wt. % and labeled as MCM10, MCM20 and MCM30, respectively. All samples were freeze-dried and air-dried for further characterization.

CHARACTERIZATION

X-ray Diffraction (XRD) (Bruker AXS D8 Advance) analysis was performed on all samples using radiation of $\text{Cu K}\alpha = 1.5458 \text{ \AA}$ in a diffraction angle (2θ) range of 5 to 60° with a step size of 0.0250° . The surface morphology observation of the film samples were studied using scanning electron microscope (SEM) model LEO 1450VP. The samples were sputtered with gold, observed and photographed. Vibration sample magnetometer (VSM) (LakeShore Model 7404) was used to analyze the magnetic properties of the MCM samples at room temperature. The MCM samples were cut into small pieces and placed in a uniform magnetic field. The samples then were tested with piezoelectric materials. The resulting changes in the electric field can be measured. The tensile properties of the CM and MCM films were analysed according to standard test method for Tensile Properties of Thin Plastic Sheeting (ASTM D882) using a Universal Testing Machine (Gotech (Taiwan) AI-3000) at a speed of 10 mm/min . The samples were cut to a size of $50 \times 10 \text{ mm}$ and five replicates were made (Kaco et al. 2015).

RESULTS AND DISCUSSION

X-RAY DIFFRACTION (XRD)

Figure 1 shows the X-ray diffraction pattern of the CM, MCM10, MCM20 and MCM30. The diffraction pattern of cellulose membrane showed three major diffraction peaks at $2\theta = 12.4^\circ$, 20.3° and 22.2° , which were assigned to the cellulose II crystal planes of $(1\bar{1}0)$, $(1\ 1\ 0)$ and $(2\ 0\ 0)$, respectively (Gan et al. 2015; Li et al. 2012). The XRD for the six main diffraction peaks at 30.19° , 35.58° , 42.91° , 53.46° , 57.24° and 62.63° which corresponded to

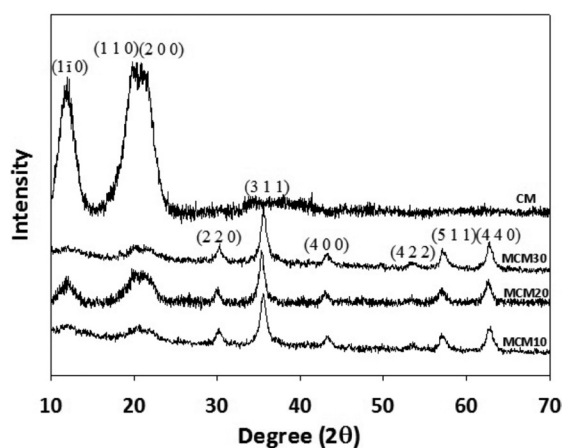


FIGURE 1. XRD patterns of CM, MCM10, MCM20 and MCM30

the crystal planes of Fe_3O_4 ($2\ 2\ 0$), $(3\ 1\ 1)$, $(4\ 0\ 0)$, $(4\ 2\ 2)$, $(5\ 1\ 1)$ and $(4\ 4\ 0)$ are referring to Fe_3O_4 nanoparticles (Luo et al. 2009). For the MCM10, MCM20 and MCM30 samples, they exhibited the crystallinity peaks of both pure cellulose and Fe_3O_4 . The presence of Fe_3O_4 peaks in MCM samples showed that Fe_3O_4 particles were encapsulated in cellulose matrix where the structure of Fe_3O_4 is barely changed. Therefore, it is proven that the Fe_3O_4 nanoparticles have been sheltered by a shell in the cellulose matrix (Luo & Zhang 2010b). The crystallinity index (CrI) for cellulose II (regenerated cellulose) decreased as increasing the Fe_3O_4 content in the samples as shown in Table 1. The existence of magnetite particles has disturbed the crystalline area of regenerated cellulose (Luo & Zhang 2010b).

TABLE 1. Crystallinity index of CM, MCM10, MCM20 and MCM30

Sample	Magnetite content (%)	Crystallinity Index (%)
CM	0	71.35
MCM10	10	58.10
MCM20	20	53.85
MCM30	30	44.74

SURFACE MORPHOLOGY

The morphologies of CM and MCM are presented in Figure 2. The surface of CM exhibited highly porous structure (Figure 2(a)). After the addition of Fe_3O_4 nanoparticles, many particles clustered presented and can be clearly seen on the surface of MCM (Figure 2(b), 2(c) and 2(d)). This indicated that the Fe_3O_4 nanoparticles have been deposited onto the CM. The increased amount of Fe_3O_4 nanoparticles in the MCM resulted in bigger clusters and clogged together. Moreover, the interaction between cellulose and Fe_3O_4 nanoparticles in the membrane has tightly entrapped within cellulose membrane via electrostatic interactions to support the Fe_3O_4 nanoparticles structure and nature (Galland et al.

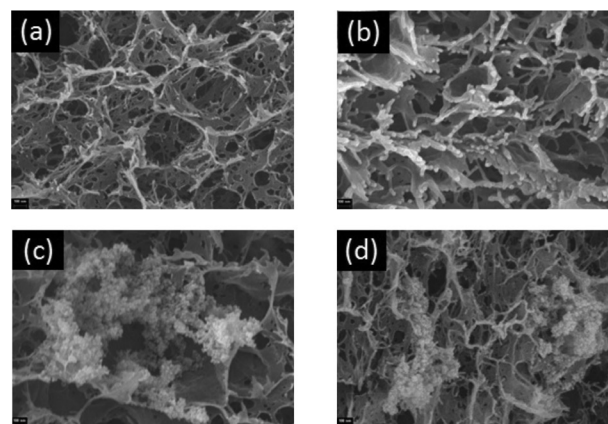


FIGURE 2. SEM images of (a) CM, (b) MCM10, (c) MCM20 and (d) MCM30

2013; Luo & Zhang 2010b; Yu et al. 2014). Besides, some pores were observed on the surface of MCM, indicated that the porous structure of CM was not totally blocked by the Fe_3O_4 (Galland et al. 2013; Yu et al. 2014). The MCM30 image showed many pores were still observed on the surface of the membrane indicating various interpenetrating pores inside the magnetic cellulose membrane.

MAGNETIC PROPERTIES

The formed CM and MCM produced can be seen in Figure 3(a) and Figure 3(b) where CM shows transparent property while MCM samples are in black color. Figure 4 illustrates the hysteresis loops of MCM samples at different magnetite content i.e. 10, 20 and 30% measured under VSM. All MCM samples showed the existence of a linear relationship between the magnetic properties and the magnetite content. The saturation magnetization (M_s) of the MCM30 has the highest value and the M_s reduced as the magnetite content decreased in the membrane where the immobilization of Fe_3O_4 nanoparticle in cellulose solution increases the M_s of MCM (Table 2). This was due to the low content of Fe_3O_4 nanoparticle in the MCM10 sample (Zhou et al. 2014) and the hysteresis loops also showed by the MCM samples were totally small. The small hysteresis loop indicated that the magnetic membrane has superparamagnetic property (Luo & Zhang 2010b). In the presence of an external magnetic field, the magnetic membrane with superparamagnetic property can be magnetized and attracted to that magnetic field. However, the magnetization of the magnetic membrane cannot be maintained when the external magnetic field was removed (Luo et al. 2009).

TENSILE PROPERTIES

The tensile strength of CM and all MCM samples were tested using tensile test machine at room temperature. Figure 5 shows the stress versus strain curves of the materials

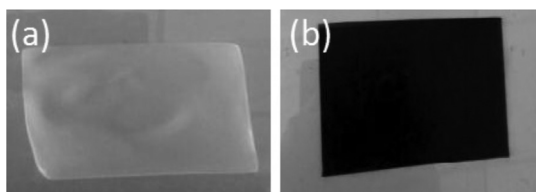


FIGURE 3. Photograph of (a) cellulose membrane and (b) magnetite cellulose membrane

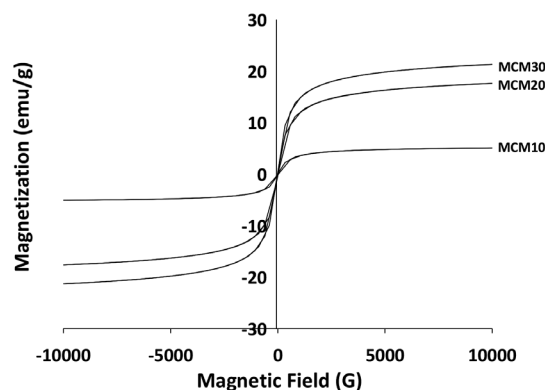


FIGURE 4. Hysteresis loops of magnetic cellulose membrane at different magnetite content (MCM10, MCM20 and MCM30)

tested. It shows that the CM has the highest tensile strength (74.4 MPa) and tensile strength reduced with the addition of Fe_3O_4 nanoparticles in the cellulose membrane. The tensile strength of MCM10, MCM20 and MCM30 were 33.1, 28.91 and 4.66 MPa, respectively. This was supported by the previous studies that showed the tensile strength decreased with increasing percentage of Fe_3O_4 nanoparticle on cellulose (Chia et al. 2006; Zakaria et al. 2005). The breakdown of interfiber bonding which is the hydrogen bond between the fiber may occur resulting to the reduction of tensile strength. The rigidity of the MCM structure due to the dispersion of nanoparticles via electrostatic interaction into the cellulose solution also may exist (Zakaria et al. 2005). Unlike cellulose membrane filled with graphene (Gan et al. 2015), the addition of magnetite particle in cellulose solution has not contributed any strength to the membrane.

CONCLUSION

Concisely, cellulose membrane and magnetic cellulose membrane were successfully produced from the mixing between cellulose solution and synthesized magnetite particles at different magnetite content via *ex-situ* process. The existing of Fe_3O_4 in XRD peaks together with cellulose II peaks indicate that Fe_3O_4 nanoparticle has been deposited onto the cellulose membrane. Moreover, the surface of the magnetic membrane shows deposition Fe_3O_4 nanoparticle. The magnetic cellulose membrane exhibited superparamagnetic properties with the increased of saturation magnetization by increasing the magnetite content. However, due to failure on hydrogen bonding

TABLE 2. Hysteresis loops of magnetic cellulose membrane at different magnetite content (MCM10, MCM20 and MCM30)

Sample	Saturation Magnetization, M_s (emu/g)	Retentivity, M_r (emu)	Coercivity (G)
MCM10	5.1109	0.1421	23.603
MCM20	17.362	0.3063	14.796
MCM30	21.521	0.3572	13.516

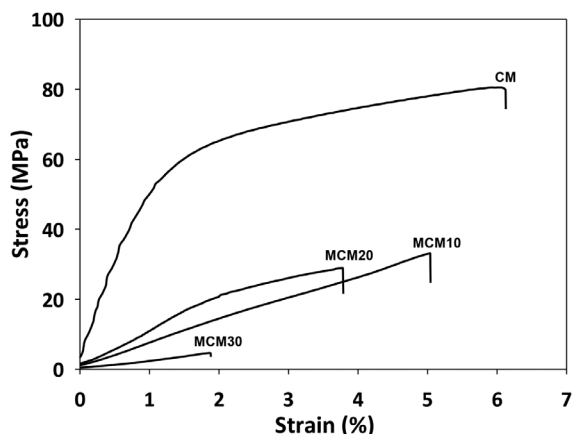


FIGURE 5. Stress-strain curves of cellulose membrane and magnetic cellulose membrane at different magnetite content (MCM10, MCM20 and MCM30)

during the mixing of Fe_3O_4 nanoparticle and cellulose, it has resulted on reduction of tensile strength.

ACKNOWLEDGEMENTS

The authors are grateful for the support of Universiti Kebangsaan Malaysia for the financial support (AP-2015-005,LRGS/TD/2012/USM-UKM/PT/04) and the Centre for Research and Instrumentation Management (CRIM) (UKM).

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Received: 13 April 2016

Accepted: 19 August 2016