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Water Hyacinth for Superabsorbent Polymer Material

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Abstract: Superabsorbent polymers, as raw material for disposable material, will cause environmental problems. One of the organic materials that used for superabsorbent is carboxymethyl cellulose (CMC). Water hyacinth, which has a relatively high cellulose content, is potentially used as non-wood cellulose resources. Study on the synthesis of CMC from water hyacinth has been reported with a maximum degree of substitution (DS) of 0.72 with isopropyl alcohol as solvent. There are opportunities to find the best solvent in order to obtain CMC from water hyacinth with higher DS. Isobutyl alcohol, which has lower polarity than isopropyl alcohol, is a potential solvent. By mixing isobutyl alcohol with other solvent which has good ability to swell the cellulose it is expected to reduce the effects of steric hindrance on the isobutyl alcohol, the polarity of the solvent mixture is lower than isopropyl alcohol and water hyacinth-based CMC with a higher DS could be obtain. Synthesis of superabsorbent material from CMC with optimum DS will results in highest absorption capacity. With a broader range of DS value of CMC, the optimum DS is expected to be more easily determined. In addition, the water hyacinth-based CMC with a higher DS are expected to be more valuable.

Key words: Carboxymethyl cellulose • Water Hyacinth • Superabsorbent • Cellulose • Degree of Subtitution

INTRODUCTION

Superabsorbent polymers (SAP) are materials which, in the presence of water, exist in a gel state and can absorb large quantities of liquid. Polymer regarded as superabsorbent when it may absorb liquid 20 times or more than its own weight. The fibers with a unique structure swell on absorbing large quantities of water and become hydrogels and can retain most of the water even when they are pressurized [1], different from conventional absorbent which will release the liquid back when squeezed. Superabsorbent material widely used for products that require high hygienic, such as baby diapers and sanitary napkins. Superabsorbent was also used in the field of agriculture and plantation.

Based on the original sources, superabsorbent were often divided into two groups which are synthetic (petrochemical-based) and natural. Source of natural materials are the polysaccharide and polypeptide. The natural-based SAP are usually prepared by addition of some synthetic part to the natural substrates [2]. At this time, most of the superabsorbent material using a synthetic material such as acrylic compounds due to their superior price to performance balance [2]. However, the use of superabsorbent material in large quantities as a disposable material will cause environmental problems [3]. Therefore, the use of environmental-friendly materials need to be developed more. Polysaccharide that can be used as raw material superabsorbent material is the cheapest, most abundant, available and renewable organic material [2].

Among the various types of polysaccharides, cellulose is the most potential one, since it is the most abundant natural polymer with excellent biodegradability and biocompatibility. However, the poor solubility of cellulose in water and most organic solvents and the poor reactivity make it difficult to be directly made into other useful materials. The modified cellulose derivatives can overcome this drawbacks. Carboxymethyl cellulose (CMC) or sodium carboxymethyl cellulose (NaCMC) is a cellulose derivative with carboxymethyl groups (-CH₂-COONa) bonded to some of hydroxyl groups on cellulose

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Table 1. Chemical Composition of Several Types of Floets (%-w)[8]						
Fiber	Cellulose	Hemicellulose	Lignin	Pectin	Fat/Wax	
Cotton	90-99					
Flax	64-84	16-18	0,6-5,0	1,8-2,0	1,5	
Hemp	67-78	16-18	3,5-5,5	0,8	0,7	
Ramie	67-99	13-14	0,5-1,0	1,9-2,1	0,3	
Jute	51-78	12-13	10,0-15,0	0,2-4,4	0,5	
Kenaf	44-72	19	9-19	-	-	
Sisal	50-74	10-14	8-11	1	2	
Water Hyacinth [7]	60	8	17	-	-	
Water Hyacinth [9]	64,51		7,69			
Bamboo	40-55					
Bagasse	33-45					
Barley	48					
Oat	44-53					
Rice straw [10]	38,3	31,6	11,8			
Rye	50-54					
Wheat straw [11]	39	38,7	17	-	1,9	
Oil palm empty bunches [12]	44.4	30.9	14.2			
Sugarcane Bagasse [13]	43,6	33,5	18,1		0,8	
Bagasse [14]	50	25	25			

World Appl. Sci. J., 22 (5): 747-754, 2013

Table 1: Chemical Composition of Several Types of Fibers (%-w) [8]

backbone. The polar carboxyl groups provide the cellulose soluble, chemically reactive and strongly hydrophilic and so the application of CMC in superabsorbent fields becomes attractive and promising [4].

The use of non-wood cellulose sources has been developed since the wood price is relatively expensive and associated with forest conservation issues and one of which is water hyacinth. This article represent a literature review on the utilization of water hyacinth in a variety of applications, synthesis of CMC, including the water hyacinth-based CMC and the synthesis of superabsorbent materials based CMC.

Utilization of Water Hyacinth: The water hyacinth (*Eichornia crassipes*), a tropical species, is a free floating aquatic weed. These plants can grow very rapidly until it reaches a very high density, more than 60 kg/m² [5]. It leading to problems such as causing damage in the area around the farm because of competing on nutrients to crops, the rapid silting waters, decrease amount of light that goes into the water so that the solubility of oxygen in water decreases, making transport difficult around these waters and disrupt regional powerhouse. Various ways have been done to solve this problem, either physical removal, chemical and biological treatment [5].

Various utilization of water hyacinth has been done, such as in wastewater treatment, production of ethanol and biogas, compost, food for several types of animals and fish, paper-making, for a variety of crafts such as rope, baskets, chairs, as well as fiberboards to be used as partition walls, roofs, etc [5]. Utilization of water hyacinth as a source of cellulose has also been reported [6, 7].

Table 1 shows the chemical composition of the fibers from several types of non-timber plant. Some of cultivated plants such as cotton, flax, hemp, ramie, jute, kenaf and sisal has a high cellulose content up to 99%-w. These plants began widely used for textile materials. Bagasse, rice straw, wheat stalks, bamboo, oil palm empty bunches and water hyacinth are some examples of the non-cultivaton sources of cellulose. Among the plants that are not cultivated, water hyacinth contains a relatively high cellulose at 64.51%-w, so it has the potential to be used as a source of cellulose for various applications. In addition, with relatively low lignin content (*i.e.*, 7.69%-w) the water hyacinth will be more easily removed the lignin to obtain high-purity cellulose.

Isolation of cellulose from water hyacinth has been made through the chemical and mechanical treatment [6]. Lignin and hemicellulose was removed using sodium chlorite (NaClO₂) and NaOH, followed by ball milling, cryocrushing and sonication treatment to obtain uniform dispersion.

Abdel-Fattah, *et al.* used variation and combination of solvents to isolate cellulose from water hyacinth. Reagents used are NaOH, paracetic acid (acetic anhydride and H_2O_2), H_2O_2 under alkaline conditions (NaOH), NaClO₂, NaClO₂-NaOH, NaClO₂-H₂O₂

Table 2: The effect of treatment on the components in the water hyacinth [7]					
	Recovered component,%				
Pretreating agent	Cellulose	Lignin	Hemicellulose		
NaOH	86,1	14,03	12,5		
Paracetic acid	80,85	5,86	20,54		
H_2O_2	90,5	12,3	50,0		
NaClO ₂	100	16,3	100		
$NaClO_2 + NaOH$	89,55	3,96	60,44		
$NaClO_2 + H_2O_2$	92,24	1,98	46,82		
NaClO ₂ + paracetic acid	96,69	2,56	81,38		

World Appl. Sci. J., 22 (5): 747-754, 2013

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and NaClO₂-paracetic acid. The results is shown in Table 2. Paracetic acid was more effective than NaOH in delignifying water hyacinth. Lignin removal reached 94.14% as compared to using sodium hydroxide (*i.e.*, 86%). Comparatively the adverse effect of paracetic acid on hemicellulose is less than that of sodium hydroxide. Hemicellulose removal achieved 87.5% when using NaOH and 79.46% for paracetic acid. Using NaClO₂ could remove 83.7% lignin without dissolved participating of cellulose and hemicellulose [7].

Research on isolation of cellulose from other nonwood plants has been reported, such as the cellulose from bagasse [13, 14], wheat straw [11, 15], hemp [16], soybean [17], sugar beet [18], sisal [19], banana rachis [20], cassava bagasse [21], flax [22], cotton [23], kenaf [24], banana fibers [25], coconut husk [26], bamboo [27], rice straw [10, 28], rice husk [29] and grape skins [30]. Cellulose was isolated by various of chemicals, such as alkali, KOH or NaOH, acid (HCl, H₂SO₄, HNO₃ and acetic acid), H₂O₂ and NaClO₂ and sometimes combined with sonication treatment.

Sun, et al. isolated cellulose from sugarcane bagasse using three types of processes, alkaline-peroxide with and without ultrasonic, sodium chlorite in acid condition and a mixture of acetic acid and nitric acid. Treatment with sonication solubilized hemicellulose and lignin 0.8% and 1.5%, respectively, higher than without ultrasonic. Yield the cellulose within the three processes are not much different from 43.0 to 45.9% [13].

Bhattacharya, et al. isolated the cellulose microfibres from bagasse using the NaOH, NaClO₂ and acetic acid. Then hydrolyzed using the sulfuric acid to obtain microfibres [14].

Isolation of cellulose from wheat straw was carried by using mixture of acetic acid and water, mixture of formic acid, acetic acid and H₂O, methanol and H₂O, ethanol and H₂O and using HCl as catalyst [11]. H₂O₂ and cyanamide (CN_2H_2) is used for bleaching under alkaline conditions. The higher concentration of acetic acid, increasing intrinsic viscosity. The higher formic acid concentration, HCOOH, the higher the intrinsic viscosity. The use of methanol and ethanol are less satisfactory. The high temperatures and the use of strong chemicals will increase the delignification process, but will also led to degradation of polysaccharides, especially if there is a transition metal ion. Alemdar, et al. used NaOH, HCl and the cryocrushing to isolate the cellulose from wheat straw [15].

Another source of cellulose was soybean which is used as reinforcement in polymer composites. This isolation process used the NaOH, HCl and chlorine dioxide (ClO₂) [17]. The use of NaOH/KOH, H₂O₂, HCl and NaClO₂ for isolation of cellulose micro fibrils from banana rachis for biocomposite applications, was carried by Zuluaga, et al. [20]. KOH with higher concentration (*i.e.*,18%-w) yield cellulose with lowest content of xylose and mannose 11.92% and 7.83%, respectively. In order to isolate cellulose from kenaf was used NaOH-AQ (anthraquinone) and NaClO₂. Anthraquinone compounds was used to reduce the degradation of cellulose because of stabilizing the cellulose. The resulting product has a composition of the cellulose, hemicellulose and lignin 92%, 5.2% and 0.5%, respectively [24]. Elanthikkal, et al. isolated the cellulose from banana fibers using NaOH dissolve pectin and hemicellulose, NaOCl for to bleaching and sulfuric acid for the hydrolysis process [25]. Rosa, et al. isolated the nanowhisker cellulose from coconut husk fiber using sulfuric acid for delignification. The use of acid was not only expected to "dissolve" the amorphous phase but also to damage some crystalline areas resulting shorter nanofiber [26].

The other processes of cellulose isolation was delignification of bamboo using NaClO₂ while hemicellulose and starch removal using the NaOH [27], isolation the nanocrystal of cellulose from rice straw by using NaClO₂ and KOH [10] and rice husk delignification using the NaOH, ultrasonic and followed by H₂O₂ and tetra-acetylethylenediamine (TAED) treatment to take remaining hemicellulose and lignin [29]. Ping Lu and You-Lo Hsieh isolated the cellulose from grape skins by (1)

using the H_2SO_4 to hydrolyze polysaccharides and polyphenolics which soluble in acid and NaOH to dissolve hemicellulose and other polysaccharides which soluble in alkaline. Subsequently, H_2O_2 treatment to oxidize and dissolve the lignin and phenolic and NaClO₂ treatment to dissolve the remaining lignin, phenolic and other impuritis [30].

Synthesis of Carboxymethyl Cellulose (CMC): Carboxymethyl cellulose is produced by etherification of the hydroxyl groups with monochloroacetic acid (MCA) or sodium monochloroacetate (SMCA) in the presence of aqueous alkali. This reaction was conducted in two steps alkalization and etherification under heterogeneous conditions. The process was optimized with respect to DS by varying the reaction parameters such as concentration of NaOH, MCA, temperature and duration of reaction. A typical carboxymethylation method involving given below two competitive reactions was followed [8].

Cell-OH + ClCH₂COOH + NaOH
$$\rightarrow$$
 Cell-OCH₂COONa + NaCl + H₂O

Sodium carboxymethyl cellulose

$$ClCH_2COOH + NaOH \rightarrow HOCH_2COONa + NaCl$$
 (2)
Sodium glycolate

Production of CMC is simpler than that of most other cellulose ethers because all reactions are operated at atmospheric pressure using commercially available reagents. The etherifying reagent, SMCA, is easy to handle and very efficient [31].

CMC is a cellulose derivative which has the ability to suspend solids in aqueous media, stabilize emulsions and thicken solutions. It has many applications in industry such as food and beverages, pharmaceutics, paper, textiles, ceramics, lubricants, adhesives, detergents and cements [32]. The properties of CMC is strongly influenced by the degree of substitution (DS), i.e. the average number of hydroxyl groups on the cellulose substituted by carboxymethyl groups and the maximum is 3 [8]. CMC has a commercial value of DS from 0.4 to 1.4 [33]. The researchers developed a variety of methods to get higher DS to get a better commercial product [34].

One of the factors that affect the value of DS of CMC is the type of solvent used. The solvent is used as a medium for the substitution of functional groups of CMC. Effect of solvent in this process is its ability to dissolve etherification substances and to swell cellulose to

improve accessibility of the etherifying agent into cellulose structure [34]. Polarity and stereochemistry of organic solvents affect the carboxymethylation on cellulose. Lower polarity of the solvent, will increase the efficiency of carboxymethylation reaction [35]. Research on variations in the use of solvents has been widely used. Barai, et al. using isobutyl alcohol, isopropyl alcohol, ethanol and water as a solvent in the synthesis of CMC from water hyacinth and DS values were generated for each of these solvents are 0.28; 0.68; 0.62; and 0.14. Maximum DS 0.72 obtained using isopropyl alcohol. Isobutyl alcohol has a lower polarity, but it produced lower DS value. This was caused by the presence of three methyl groups surrounding the primary carbon atom which may provide steric hindrance to the reacting groups [35]. Pushpamalar, et al. also compare some of the solvent in the synthesis of CMC from sago waste. The solvents water. dimetilformamide (DMF), methanol. are dimethylsulfoxide (DMSO), isopropyl alcohol, ethanol and butanol. Maximum DS were 0.821, using isopropyl alcohol and NaOH 25% (w/v) [31].

Research of the solvent in the synthesis of CMC, shows that the use mixture of solution as a solvent in the process of alkalization and karboksimetilasi result in higher DS [34, 36-38]. Some research about variation of solvent mixture were ethanol and acetone mixture [36, 37], ethanol and isopropyl alcohol mixture [34, 37, 38] and mixture of dimethyl sulfoxide (DMSO) and tetrabutylammonium fluoride trihydrate (TBAF) [39]. The polarity of the solvent decreases as the number of carbon atoms in the solvent [34].

Some research on the synthesis of CMC using wood as raw material and acetone, ethanol, isopropanol and acetone-ethanol mixture and ethanol-isopropanol as solvent [37]. Maximum DS 0.73 achieved by using solvent mixture with a ratio of ethanol: isopropanol 1:1. DS values with ethanol, acetone and isopropanol were 0.39, 0.48 and 0.63, respectively. CMC synthesis from sisal cellulose and cellulose linter by using the solvent dimethyl sulfoxide (DMSO) and tetrabutylammonium fluoride (TBAF) produced DS up to 2.0 (using solid NaOH) and 1.63 using aqueous NaOH [39]. Ruzene, et al. synthesized CMC from bagasse by using ethanol/water at acidic and produce value DS 0.7 [40], while Dapia, et al. made the CMC from Eucalyptus globulus wood by using a isopropanol solvent [32]. CMC synthesis of cotton fiber produced commercially by isopropanol solvent DS value of 0.7 [41] and CMC from viscose rayon using ethanol yield DS 0.65 [42]. Ismail, et al. synthesized CMC by using a solvent mixture of ethanol-isopropanol produces maximum DS value of 0.7443 on the ratio of ethanol: isopropanol 1:1 [34]. CMC Synthesis by using solvent isopropanol, ethanol and isopropanol: ethanol 1:1 produces DS values 0.99, 0.98 and 1:02, respectively [38], whereas CMC from banana pseudo stem (*Musa cavendishii* LAMBERT) with isopropanol solvent yield DS 0.26 to 0.75 [43].

Synthesis of Superabsorbent Polymer Materials (SAPs):

Superabsorbent materials are high performance water absorbent and retention material with three-dimensional network structure. This material can absorb water and other liquids up to thousands times their own weight in a relatively short time. They can also maintain the condition of swollen although under pressure. Superabsorbent properties is due to the hydrophilic groups within it. Most of the superabsorbent material is made of a synthetic polymer-based acrylic acid (AA) or acrylamide (AM), which are very difficult to degrade and are not environmental-friendly. Decline in petroleum reserves and increasing environmental pollution caused by synthetic polymers into the shift to biodegradable materials [44]. Cellulose and its derivatives is one of the environmental-friendly material that can be used as raw material SAP.

Some of research on the synthesis of cellulose-based SAP material are using oil palm empty fruit bunches [45], wheat straw [46], flax yarn waste [44] and cellulose derivatives such as carboxymethylcellulose/CMC [47], hydroxyethylcellulose / HEC, CMC and combination of CMC-HEC [48]. Bao, et al. have compared the ability of water absorption from superabsorbent made of methyl cellulose (MC), hydroxypropyl methyl cellulose (HPMC), hydroxyethyl cellulose (HEC), carboxymethyl cellulose (CMC) and without cellulose [49]. The results showed that the superabsorbent with CMC material has the highest water absorption capacity. Some of the crosslinking agent was used for the manufacture of superabsorbent material. Marci, et al. synthesized superabsorbent through crosslinking of CMC and HEC with divinylsulfone (DVS) [50]. Another hydrogels that are made of cellulose and CMC using epichlorohydrin (ECH) as a crosslinking agent [51]. DVS and ECH are compounds that contained toxic. Demitri, et al. using citric acid (CA) as a crosslinker in the hydrogel manufacturing with raw materials CMC and HEC [52]. The results of this study showed that the hydrogel with environmentalfriendly crosslinker (i.e., CA) has the better ability to absorp water than the hydrogel with DVS. Adel, et al. synthesized hydrogel from CMC (cotton linter-based), with different DS values. This study reported that there

was an optimum DS value of CMC that result in highest water absorption capacity of superabsorbent materials [53].

Future Research of Water Hyacinth: There is an opportunity to produce CMC from water hyacinth with higher DS. Further studies on the synthesis of CMC from water hyacinth by using mixture of solvent to obtain a higher DS value are needed.

Isobutyl alcohol actually has the potential to be a better solvent because it has a lower polarity than isopropyl alcohol, but in fact it gave lower value of DS. If isobutyl alcohol mixed with other alcohol compounds that are easier to swell cellulose, such as isopropyl alcohol, it is expected to reduce the effects of steric hindrance on the isobutyl alcohol and get the polarity of the solvent mixture is lower than isopropanol, so it is expected generated higher DS.

Synthesis of superabsorbent material from water hyacinth-based CMC has not been developed. Citric acid is a crosslink agent which is cheap and evironmentally friendly. By using citric acid as crosslink agent, it is expected to obtain an environmental-friendly superabsorbent material from water hyacinth-based CMC.

With a broader range of DS value of water hyacinth-based CMC, the optimum DS value which could produce SAP material with higher absorption capacity is expected to be more easily determined. In addition, with a higher DS, the water hyacinth-based CMC are expected to be more valuable and wider applications.

CONCLUSION

Utilization of water hyacinth as raw material for superabsorbent material with CMC has not been developed. There are no data regarding the optimum DS value of CMC-water hyacinth that as a raw material of superabsorbent material.

Therefore, it is necessary to further study about synthesis of CMC from water hyacinth with appropriate solvent. Preparation of superabsorbent material from water hyacinth-based CMC can be conducted by using citric acid as crosslinking agent that is environmentalfriendly.

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