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EFFECT OF DIFFERENT TYPES OF WOOD PULP ON HYDROLYSIS REACTION TIME AND DEGREE OF POLYMERIZATION OF MICROCRYSTALLINE CELLULOSE POWDER

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ABSTARCT

Different types of wood pulps are used to manufacture microcrystalline cellulose. Percentage of alpha cellulose depends on type of wood pulp. Both wood pulp soft and hard wood pulp are usually used to manufacturing microcrystalline cellulose. Though the production process of wood pulp has a major bearing on it's quality, Soft wood pulp on most occasions has higher alpha cellulose content with excellent finished product quality in terms of yield, physical parameter i.e. bulk density, flowability and chemical parameters i.e. water soluble substances, ether soluble substances, degree of polymerization and residue on ignition. In this paper we examine different wood pulp effect on hydrolysis time and degree of polymerization of finished product microcrystalline cellulose. In this study we used four types of wood pulp and done reaction with same acid concentration, pressure, and temperature only hydrolysis time were different, after drying evaluate degree of polymerization of finished product MCC.

KEYWORDS: Wood pulp, Hydrolysis reaction, microcrystalline Cellulose, degree of polymerization.

INTRODUCTION

Pulp and paper are made from wooden cellulosic fibers. Wood is composed of cellulose, lignin, hemicelluloses and extractives (e.g. resins, fats and pectins). There are three types of cellulose present in the wood pulp, (i) Alpha cellulose (ii) Beta cellulose (iii) Gama cellulose. Generally two types of wood pulp are used to manufacturing MCC (a) Softwood pulp, (b) Hardwood pulp.[1,2] These woods differ considerably in chemical composition i.e. cellulose,

hemicelluloses and lignin and structure organization also differ i.e. regions which are more crystalline an amorphous.[3]

Microcrystalline cellulose (MCC) is native form of cellulose.[4] It is isolate from wood pulp by hydrolysis reaction.[5] Hydrolysis reaction is done in the presence of mineral acids and water at controlled temperature and pressure.[6] In wood pulp, cellulose chains are packed in layers held together by a cross-linkage polymer and strong hydrogen bond. Cellulose consists of liner chain of ß-14-D anhydroglucopyranosyl units. In hydrolysis reaction, high degree of polymers converts into low degree of polymers.[7] Degree of polymerization(DOP), the number of glucose units present in the cellulose chain) of finished product microcrystalline cellulose depends on source of wood pulp and different parameters of hydrolysis reaction i.e. reaction temperature, reaction pressure, acid concentration used in reaction and reaction time.[8]

Microcrystalline cellulose is used in various fields i.e. Pharmaceuticals industries, Cosmetic industries, Food industries, paint industries and detergent industries.[9] In pharmaceuticals MCC is a perfect excipient for making the formulations.[6] It is one of the most frequently used, to formulate solid dosage forms. It is non-reactive, free-flowing and versatile pharmaceutical excipient.[10] It has strong binding property to bind the pharmaceutical active ingredient, most extensively used filler and has inherent disintegrant properties.[11] In this study we used two types of wood pulp (soft and hard wood pulp) for examine the effect of different types of wood pulps on hydrolysis reaction and effect of hydrolysis time on Degree of polymerization of finished product microcrystalline cellulose.

MATERIAL AND METHODS

Material

Digital weighing balance (Mettler Toledo, Model no-ML802/A01) was used for weighting the sample. Class A graduated measuring cylinder (Electro lab instrument, Model No. ETD1020) was used for checking tapped density of HiCel^{TM}MCC. Hot air oven (Model no-PNX-14) was used for testing moisture content. In this study all chemicals were used AR grade.

Method

Wood pulp Degree of polymerization[12]

Weight accurately 0.125 g of moisture free wood pulp and place in 100 ml capacity glass beaker. Add 25 ml Cupriethylenediamine solution and stir the solution by using high speed stirrer (REMI) till the wood pulp is dissolved. Remove the stirrer and take a portion of solution the Cannon-Fenske viscosity meter and determine the efflux time at 27°C temperature and check density also at 25°C temperature. Using below formula calculate degree of polymerization.

$\mu = c.t.d$

Where,

μ= Viscosity of solution

c= Pipette constant

t= Time

$D = -2953+2523.8(\mu)^{0.25} - 0.021425(\mu)^2$

Where,

d= Density

D= Degree of polymerization

Alpha Cellulose content[12]

Weight accurately 3g of moisture free pulp and transfer into 250 ml beaker and kept in water bath at 20°C temperature. Take 75 ml of 17.5% sodium hydroxide solution in a graduated cylinder and kept at 20°C temperature in water bath. Wet the pulp sample with 15ml of sodium hydroxide solution and disintegrated it for 1 min with flat end of glass rod. Add 10ml more of sodium hydroxide solution to the pulp and mix it for 45 seconds. After that add 35ml of caustic soda solution and disintegrated it. The pulp should be free from lumps. Stirred and allow the mixture to stand on the water bath for 3 more minutes. Add into this another 10ml of sodium hydroxide solution and mix for 10minutes. While adding 30ml of caustic soda solution in three installment of 10ml each at different time intervals of 2.5, 5.00 and 7.5 minutes. Leave the beaker with the contents in the water bath for another 30min. Now add 100ml of distilled water at 20°C, mix with glass rod and leave the diluted mixture in water bath for 30min. Filter on tarred crucible by suction and collect the filtrate in a separate beaker. Rinse the beaker and residue with 25 ml of 8.3% sodium hydroxide solution at 20°C and transfer all the fibers to the crucible. Wash the residue with 50ml of distilled water at 20°C. Keep the filtrate aside for the determination of beta and gamma cellulose. Now place

the crucible on another suction flask. Wash the residue in crucible with 400ml of distilled water at 20°C and subsequently with 40ml of 10% acetic acid solution at 20°C. Allow soaking period of 5 min before applying the suction. Wash the residue again with distilled water till it is free from acetic acid as indicated by litmus paper. Dry the crucible in oven at 105°C and transfer the contents to a weighing bottle. Dry it to a constant weight.

Calculation- Calculate the percentage of alpha cellulose as follow;

$$x = \frac{100a}{m}$$

Where,

x= alpha cellulose percentage by weight

a= weight of precipitate in g

m= weight of pulp in g calculated on oven dry basis.

Hydrolysisof wood Pulp[13]

Wood pulp cut into the pieces and charged in reactor with mineral acid and water, hydrolyzed at specific temperature, pressure, acid concentration and time. Both wood pulps(Soft and hard wood pulp) hydrolyzed with same acid concentration, pressure, temperature only reaction time is different. After hydrolysis wood pulp breaks down into slurry. Thereafter it is washed and filtered with ammonia with the help of filter press for getting the conductivity below 100µs/cm, pH is neutral. After that slurry is dried by spray dried.

Degree of polymerization of microcrystalline cellulose[14]

Degree of polymerization of MCC, Weight accurately 1.3 mg microcrystalline cellulose in a 125 ml conical flask, Add 25 ml of water and 25 ml of 1 M Cupriethylendiamine hydroxide solution, dissolved properly with the help of glass rode. When dissolved properly, transfer an appropriate volume of the solution into a calibrate number 150 Cannon-Fenske. Allow the solution to equilibrate at 25±0.1 for not less than 5 minutes. Time the flow between the two marks on the viscometer, and record the flow time, t1 in seconds and Calculate the kinematic viscosity $(KV)_1$ of MCC taken by the formula: $t_1(K_1)$

In which k_1 is the viscosity meter constant (See viscosity 911 in USP). Obtain the flow time t_2 for a 0.5 M cupriethylene diamine hydroxide solution using a number 100 Cannon-Fenske or equivalent viscosity meter. Calculate the kinematic viscosity $(kv)_2$ of the solvent by the

formula, t_2 (k_2). In which k_2 is the viscosity, (η_{rel} of MCC specimen taken by the formula: $(KV)_1 / (KV)_2$

$$\eta = \frac{(KV)1}{(KV)2} \quad (1)$$

Determine the intrinsic viscosity, (η) c by interpolation, using the intrinsic viscosity table in reference table section. Calculate the DOP, it is denoted by "*P*".

$$P = \frac{(95)(\eta)C}{Ws\{\frac{(100-\%LOD)}{100}\}}$$

Where

WS= weight of sample (MCC)

%LOD= Loss on drying of sample (MCC)

Assay of Cellulose[15]

Weight 0.125 g MCC sample and transfer to a 250 ml capacity conical flask with the aid of 26 ml of water. Add 50ml of 0.083M postassium dichromate solution, mixed it. Add carefully 100 ml of Sulphuric acid and heat to boiling. Remove from heat and allow to cooling it at room temperature and transfer into 250 ml volumetric flask, dilute with water and make up volume. Titrate 50 ml of the resulting solution with 0.1M ferrous ammonium sulphate using 2 to 3 drops of ferroin sulphate solution indicator.

Bulk density[16]

Untapped Density

Untapped density is analyzed through graduated measuring cylinder. Take 20 gm of dry MCC powder pours into a graduated A grade 100 ml capacity cylinder slowly from the sidewall. Level the surface of sample in cylinder by slow movement and note down the occupied volume and calculate the untapped density of MCC by using following formula.

$$\text{Untapped density (BD)} = \frac{\text{Weight of powder in gram}}{\text{Occupied volume in ml}}$$

Tapped Density

Tapped density anlaysed by (Electro lab instrument, Model No. ETD1020), measuring cylinder placed in tapped density machine and fixed 100 tapped. After 100 tapped measured the volume of measuring cylinder and calculate the tapped density/porosity of MCC using following formula.

$$\text{Tapped density (TD)} = \frac{\text{Weight of powder in gram}}{\text{Occupied volume in ml}}$$

Hausner Ratioand Carr's Index[16]

The flow of powder measured by "Hausner ration". Hausner Ratio and Carr's index calculated by below formula-

Hausner Ratio (H.R) = $\frac{TD}{BD}$

Carr's Index (CI) = $100 \times \frac{TD-BD}{TD}$

Particle size (PSD)[16]

Average particle size was analysised by Sieve shaker, PSD software (Retch-Japanese instrument). PSD software operates through computer. Take cleaned mesh sieve with bottom pan and top cover. Check sieve shaker and set mesh sieve with sample being analyzed on sieve jet. Take weight of all required mesh sieve with bottom. Arrange the sieve mesh sequence from top mesh +60, mesh +200 and bottom. Weight accurately 10 gm of MCC powder with the help of weight balance (Mettler Toledo, Model no. ML802/A01) and put into top of sieve. Fill the initial weight of mesh sieve and bottom into PSD table and start. After 5 minutes take out the sieves and again take weight with retain sample. Fill into the PSD software table, in graph shows the particles size.

pH and Conductivity[1]

Take 5 gm of MCC powder add 40 ml of water mix 20 minutes with the help of glass rod and 20 minutes centrifuge (Remi elektrotechnik, Model no.-Remi-R-8CBL). Retain the supernatant for use analysis the pH by using pH meter (TOSHCON, Model no.-12/H/5563) and conductivity with conductivity meter (TOSHCON, Model no.-13J1354). Conductivity meter that has been standardized with a potassium chlorite conductivity calibration standard solution.

Moisture content[16]

Heat the shallow bottle in a hot air oven (Model no. PNX-14) at 105°C for 30 minutes after that cool it in desiccator at room temperature .Tare weight the Shallow bottle and take about 1 gm of MCC sample in shallow bottle, set oven at 105°C and kept for 3 hours. After 3 hours take out the shallow bottle allow to cool in desiccator at room temperature. When the shallow bottle is cool take weight again, Calculate moisture content by using the following formula.

$$\text{Moisture content} = \frac{\text{After drying weight of shallow bottle} - \text{empty weight of shallow bottle}}{\text{Sample weight in gram}} \text{ X } 100$$

RESULT AND DISCUSSION

Wood pulp evaluation

Both wood pulp are white color, with a brightness which is mentioned in table 1 and thick paper sheet. Degree of polymerization of both wood pulp and alpha cellulose content are summarized in table 1.

Table 1: Degree of polymerization and alpha cellulose of both soft and hard wood pulp.

Name of Test	Soft wood pulp	Hard wood pulp
Brightness	93.6	92.8
Degree of polymerization	1250	830
Alpha cellulose content	92.01	92.38

Hydrolysis of wood pulps

Hardwood pulp is hydrolysis earlier than soft wood pulp. Hardwood pulp in 120 minutes gives 230 degree of polymerization where soft wood pulp gives 270degree of polymerization. Effect of hydrolysis time on degree of polymerization is mentioned fig.1.

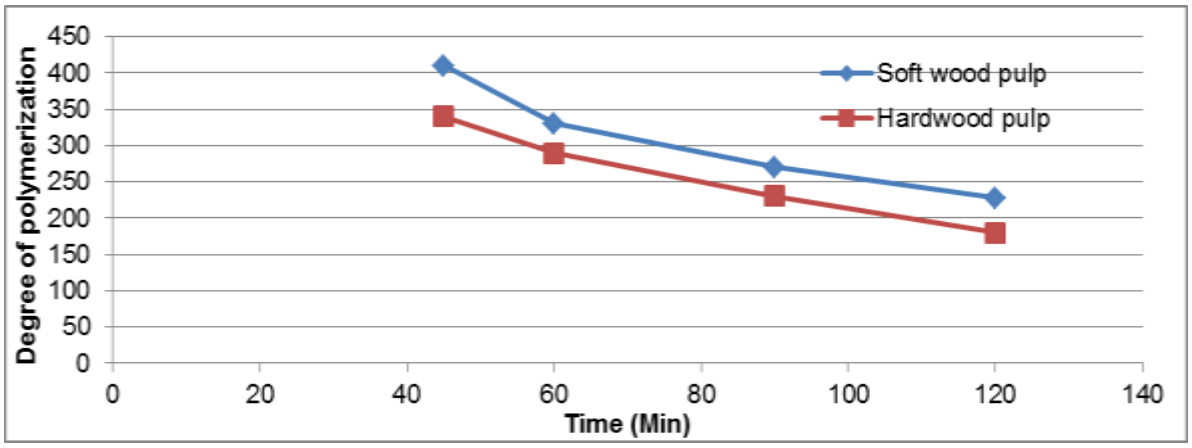

Fig. 1: Relation between hydrolysis time and degree of polymerization of soft wood and hard wood pulp with same temperature, presser, acid concentration.

Evaluation of microcrystalline cellulose

Microcrystalline cellulose is white free flowing, granular, crystalline powder. Physicochemical test results of microcrystalline cellulose are mentioned in table 2.

Table 2: Soft and hard wood pulp containing microcrystalline Cellulose result of physicochemical test.

Name of test		Soft wood pulp (MCC)	Hard wood pulp(MCC)
DOP		228	230
Assay (%)		99.99	99.98
Bulk density (g/CC)	Untapped	0.301	0.307
	Tapped	0.430	0.428
Hausner ratio		1.428	1.394
Carr's index (%)		30.00	28.27
Particle size(D50) (µm)		110	109
pH		6.25	6.23
Conductivity (µS/c)		44.6	44.5

Abbreviation

DOP: degree of polymerization, °C: centigrade, B.D: Bulk density, g: gram, MCC: Microcrystalline Cellulose, mg: milligram, PSD: Particle size distribution, L: liter, °: degree, %: percentage, H: Height, L: length, d: diameter, USP: United state pharmacopoeia, ml: milliliter, g/cc: gram per centimeter square, Hrs: Hours, mm: millimeter.

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Conflicts of interests

The authors state and confirm no conflict of interests. No direct funding was received for this study.

CONCLUSION

In this study we find out the correlation between wood pulp hydrolysis time and degree of polymerization, both wood pulps soft wood pulp and hardwood pulp are hydrolyzed at same acid concentration, pressure, temperature. Hard wood pulp is hydrolyzed in 90minutes where soft wood pulp takes time and hydrolyzed in 120 minutes.

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