Meta-Analysis on the Dissolution of Bamboo (*Bambusa*) Cellulose using NaOH/urea Aqueous Solution

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Abstract: Bamboo cellulose is a non-dangerous, biodegradable polymer with high elastic, compressive quality thereby beneficial in commercial and pharmaceutical industries. However, preparing it involves a complex procedure of solvent dissolution. NaOH/urea solution is a common solvent for cellulose dissolution, but its efficiency varies with temperature and concentration. Thus, this study aims to synthesize evidence on the efficiency of NaOH/urea solution in bamboo cellulose dissolution; identify the most suitable concentration and temperature of NaOH/urea; and determine the relationships between its concentration, temperature and bamboo cellulose' dissolution rate. Extracted data indicated the bamboo source sample, NaOH/urea concentration, temperature, and dissolution results from five databases and utilized Quality of Reporting of Meta-analyses (QUOROM) and Assessment of Multiple Systematic Reviews (AMSTAR) instruments for the studies' quality assessment. Among the studies, 93% utilized the concentration ratio of 7:12:81; therefore, concentration's minimal changes did not profoundly affect the dissolution, given the same temperatures. Out of fifteen studies, eight used -12°C affirming that minimal changes in temperature affect the dissolution results. The Chi-square test revealed that only temperature and concentration indicate a significant relationship (x2=5.793, P<0.10). The heterogeneity test displayed a small amount of heterogeneity (I2= 33.42%, P<0.10; I2= 1.8%, P<0.10) on the gathered data that may be clinically unimportant, making the data considerably homogeneous. Hence, this provides significant evidence validating the efficiency of 7:12:81 NaOH/urea aqueous solution at -12°C in the dissolution of bamboo cellulose.

Key Words: bamboo cellulose; bamboo cellulose dissolution; NaOH/urea; meta-analysis; heterogeneity test

1. INTRODUCTION

Bamboos are distinguished by woody and hollow culms, intricate rhizomes and branching processes, narrow leaf blades, and visible sheathing organs. One main component of bamboo is cellulose ($C6H12O6$), one of the most universal natural polymers on earth. It is a non-dangerous, biodegradable polymer with high elastic and compressive quality, but it has across the board use in different fields, like the pharmaceutical industry and the construction industry that incorporates cellulose insulation. However, the cellulose must be disintegrated first through solvation to melt it (Gupta et al., 2019).

Preparing bamboo cellulose is difficult and usually takes numerous steps, as there are many solvents to experiment with. Local cellulose must be dissolved in a solvent in order to melt it. Most of the dissolvable frameworks known have a restricted limit of disintegration that is poisonous and costly, restricting their mechanical capabilities. Another approach in cellulose dissolution shows that cellulose is soluble in aqueous NaOH underneath 268 K inside a particular focus scope of NaOH. This framework is modest, possibly non-contaminating, utilizes extremely regular synthetics, and is moderately simple to deal with (Alves, 2015). Therefore, the focal point of this study is to collect and analyze related studies about the dissolution of bamboo cellulose using NaOH/urea. Specifically, this work determines the most suitable concentration and temperature that can maximize the solubility of bamboo cellulose using NaOH/urea and the relationships between the concentration and temperature of NaOH/urea and the solubility of bamboo cellulose.

2. METHODOLOGY

In identifying the articles that were reviewed in this study, the researchers followed the created criterion while specifying certain "identifiers" from the paper and made use of a checklist set adapted from Porritt et al. (2014) in Table 1. The following set of keywords was used for searching: "Bamboo Cellulose," "Cellulose Dissolution," "Bamboo Cellulose Solvation," "Dissolution of Bamboo Cellulose," and "NaOH/urea." The bamboo sample sources were sorted into three categories, namely Bamboo Pulp, Bamboo Pulp Boards, and Pretreated Bamboo.

Table 1. *Criteria and identifiers for selection of studies (Porritt et al., 2014)*

Criteria	Details	Mark
Date	Any date of the study	✓
Language	Articles in the English, Filipino, or Chinese Language	✓
Peer-review	Only peer-reviewed studies	✓
Setting	Bamboo cellulose solvation done under given temperatures	✓
Methodology	Mixed-method research	✓
Publication	Only articles found in the following databases: Scopus, ScienceDirect, JSTOR, Directory of Open Access Journals (DOAJ), and ResearchGate	✓

This also utilized Quality of Reporting of Meta-analyses (QUOROM) (Russo, 2007) and Assessment of Multiple Systematic Reviews (AMSTAR) (Pizarro et al., 2021) instruments in assessing the quality of the studies from the database search. After the article screening process using Preferred Reporting Items for Systematic Reviews and Meta-Analyses (PRISMA) guidelines (Moher et al., 2009), as displayed in Figure 1, data will be extracted from the studies eligible for inclusion. These extracted data were presented in a tabular format that identified the NaOH/urea concentration, temperature, and dissolution rate from each study. Moreover, statistical analysis increased the reliability of findings through the heterogeneity chi-square test and Higgins' I^2 heterogeneity statistic, as these tests are also required in a meta-analysis. After conducting all statistical treatments, these results were used in further analysis in determining the most suitable concentration and temperature for bamboo cellulose's maximized solubility in NaOH/urea aqueous solution.

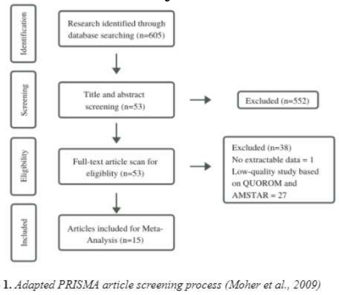

Figure 1. *Adapted PRISMA article screening process (Moher et al., 2009)*

3. RESULTS AND DISCUSSION

In gathering studies, a total of 605 studies were available from the databases that were searched using the keywords. From these studies, 53 studies chosen based on the title were screened and underwent the identification of studies wherein screening of the abstract and full text was executed. Of these, only 15 studies have met the criteria of quality assessment and were used for data extraction, as shown in the screening process in Figure 1.

Furthermore, the study of Yang et al. (2018) used bamboo pulp boards obtained from Guizhou, China which were dissolved with 7% NaOH/12% urea/81% distilled water at -12 °C. It was found that as soon as cellulose fibers are placed into NaOH/urea aqueous solution, they swell, and part of the intra- and intermolecular hydrogen bonding are destroyed. The work of Chen et al. (2015) used bamboo pulps which were obtained after delignification with sodium chlorite and alkaline treatment with 25% potassium hydroxide from bamboo. It was dissolved in a 7/12 wt % NaOH/urea solution at -12.6 °C. In this study, bamboo pulps just swelled in the NaOH/urea aqueous solution since it had been demonstrated that only cellulose with a viscosity-average molecular weight below 10.0×10^4 Da could be completely dissolved in NaOH/urea aqueous solution. Shi et al. (2017) used commercial bamboo dissolving pulp boards provided by Sichuan Lee & Man., while commercial bamboo dissolving pulp boards provided by Sichuan Liwen Paper Co. Ltd. were also used by Shi et al. (2017). Both were dissolved with 7% NaOH and 12% urea aqueous solution at -13 °C, with a dissolution rate of 43.4.

Additionally, Lin et al. (2017) used bamboo pulp obtained from Sichuan, China, and dissolved it in 7% NaOH, 12% urea, and 81% H2Osolution at -13 °C. After stirring for 10 min, they observed a homogenous cellulose solution. Moreover, bamboo (Phyllostachys heterocyla) pulp from Guizhou Chitanhua Paper Industry Co., Ltd. was used by Zhu et al. (2015), which was dissolved at 7% NaOH and 12% urea, precooled to 4 °C and maintained at -12 °C. It had a dissolution rate of 100% with a transparent and viscous bamboo pulp cellulose solution. Similar results were observed by Nguyen et al. (2019) where they used micron-size White Bamboo (Dendrocalamus membranaceus Munro) fibrils, a pretreated bamboo from Hoa Binh Province, Vietnam, which was dissolved in 7% NaOH, 12% urea, and 81% distilled water at 5°C. Their results show almost 24% dissolution with a semi-transparent to transparent cellulose. In the study of Tang et al. (2017), cellulose was obtained from a bamboo dissolving pulp board from Sichuan, China, where it was dissolved in NaOH/urea/water solution (7:12:81 by weight) at -12°C. In the cellulose I crystals, the hydrogen bonds were destroyed when the cellulose I crystals were dissolved in NaOH/urea solution.

Moreover, Li et al. (2011) also used pretreated bamboo, but with partially delignified bamboo (Neosinocalamus affinis) culms which were 100% dissolved with 7% sodium hydroxide/12% urea solution at -12 °C. Figure 2 summarizes the dissolution rate of the aforementioned individual studies while Table 2 summarizes the data extracted from these studies.

Out of the 15 studies, 14 studies (93%) utilized the 7:12:81 ratio for the concentration of NaOH/urea aqueous solution (see Figure 3). Almost all of these studies showed the said concentration to be 100% effective for bamboo cellulose dissolution. Varying dissolution rate may have resulted from other factors like vigorous stirring (Nguyen et al., 2019), ultrasound/ethanol pretreatment (Li et al., 2021), and vacuum oven drying (Shi et al., 2017 & Shi et al., 2017); yet, they are still proven to be effective. However, the study of Ma et al. (2017) used the concentration ratio of 7.5:11:81.5. The minimal change in the concentration did not greatly affect the dissolution of bamboo cellulose, given that the temperature is the same. For these reasons, the optimum NaOH-urea-distilled water ratio for dissolving bamboo cellulose is 7:12:81.

Table 2. *Summary of the data extracted*

Bamboo Source Sample	NaOH-urea-distilled water Concentration	Temperature	Dissolution Rate	Author/s	Year
Bamboo Pulp	7:12:81	-12.5 °C	100%	Li et al.	2015
Bamboo Pulp	7.5:11:81.5	-7 °C	100%	Zhai et al.	2018
Pretreated Bamboo	7:12:81	-12 °C	61%	Kong et al.	2021
Bamboo Pulp	7:12:81	-12 °C	75.1 -77.7%	Li et al.	2010
Bamboo Pulp	7:12:81	-12 °C	83.6% - 86.6%	Li et al.	2021
Bamboo Pulp Boards	7:12:81	-12 °C	61%	Yang et al.	2018
Bamboo Pulp	7:12:81	-12.6 °C	0%	Chen et al.	2015
Bamboo Pulp Boards	7:12:81	-13 °C	43.4%	Shi et al.	2017
Bamboo Pulp Boards	7:12:81	-13 °C	43.4%	Shi et al.	2017
Bamboo Pulp	7:12:81	-13 °C	100%	Lin et al.	2017
Bamboo pulp	7:12:81	-12 °C	100%	Zhu et al.	2015
Pretreated Bamboo	7:12:81	5 °C	24%	Nguyen et al.	2019
Pretreated Bamboo	7:12:81	-12 °C	100%	Lou et al.	2015
Bamboo Pulp Board	7:12:81	-12 °C	100%	Tang et al.	2017
Pretreated Bamboo	7:12:81	−12°C	100%	Li et al.	2011
Bamboo pulp	7:12:81	-12 °C	100%	Zhu et al.	2015
Pretreated Bamboo	7:12:81	5 °C	24%	Nguyen et al.	2019
Pretreated Bamboo	7:12:81	-12 °C	100%	Lou et al.	2015
Bamboo Pulp Board	7:12:81	-12 °C	100%	Tang et al.	2017
Pretreated Bamboo	7:12:81	−12°C	100%	Li et al.	2011

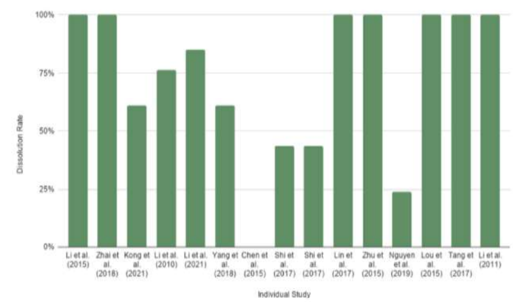

Figure 2. *Pooled dissolution percentages of individual studies*

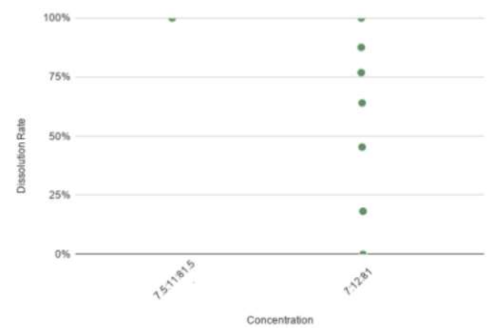

Figure 3. *Effect of concentration of the solution on dissolution rate of bamboo cellulose*

Meanwhile, 8 out of 15 studies dissolved bamboo cellulose at a relatively lower temperature, specifically at -12°C. Nearly all of the studies show that bamboo cellulose can be 100% dissolved using NaOH/urea at -12°C, as shown in Figure 4. Results obtained by Li. et al. (2011) and Zhu et al. (2015) present that the most effective and efficient way to completely dissolve pretreated bamboo and bamboo pulp was at -12°C. On the other hand, Nguyen et al. (2019) and Chen et al. (2015) dissolved cellulose at 5°C and -12.6°C. Minimal change in the temperature affected the dissolution results since at -12.6°C, the bamboo pulp just swelled in the NaOH/urea aqueous solution, and 0% of the pulp was dissolved. Moreover, Zhai et al. (2018) argued against the application of temperature above 0°C in the solution as cellulose would not be dissolved in that case as the swelling, softening, and dissolving exothermic processes can only protect the pulp fibers at low temperatures below that degree. Under these conditions, the most ideal temperature for the solvation of bamboo cellulose is at -12°C.

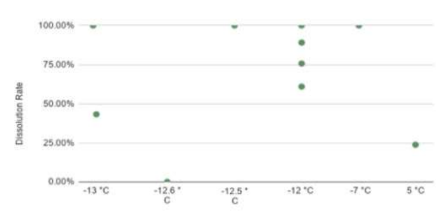

Figure 4. *Effect of solution's temperature on dissolution rate of bamboo cellulose*

To solidify the findings, statistical analyses were done. Chi-square test was used to confirm if there were significant relationships between variables. Upon analysis, bamboo sample sources and temperature of the aqueous solution ($x2=3.945$, $P<0.10$) were independent of each other. Concentration of aqueous solution and bamboo sample sources ($x2=1.587$, $P<0.10$) also have no significant relationship. Conversely, temperature and concentration ($x2=5.793$, $P<0.10$) of the aqueous solution indicates relation. Furthermore, determining statistical heterogeneity is important for a meta-analysis to detect variability among factors influencing the intervention. In this analysis, a p-value of 0.10 was used to determine statistical significance as nonsignificant results were not considered evidence for heterogeneity. The Higgins' I2 test revealed a small amount of heterogeneity on the gathered data that may be clinically unimportant, thereby making the data considerably homogeneous: ($I2= 33.42\%$, $P<0.10$ for source and temperature; $I2= 1.8\%$, $P<0.10$ for source and concentration). Visual assessment of the plotted rates in Figure 4 showed that four studies (Lin et al., 2017; Shi et al., 2017; Chen et al., 2015; Nguyen et al., 2019) accounted for the heterogeneity with extreme temperatures as the possible reason ($I2= 69.52\%$, $P<0.10$). Hence, the substantial evidence corroborates the guaranteed efficiency of NaOH/urea aqueous solution at low temperature in the dissolution of bamboo cellulose.

4. CONCLUSIONS

Accounting for 93% of all studies, the 7:12:81 ratio for the concentration of NaOH/urea aqueous solution dominated the data and mostly exhibited a 100% efficiency for the dissolution of bamboo cellulose. Therefore, the optimum NaOH-urea-distilled water ratio for dissolving bamboo cellulose is 7:12:81. Meanwhile, 8 out of 15 studies dissolved bamboo cellulose at a relatively lower temperature, specifically at -12 °C. Minimal change in the temperature affected the dissolution results with temperatures exceeding 0 °C labeled as protection-inefficient and receding -12 °C resulted in swelling; thus, the most ideal temperature for the solvation of bamboo cellulose is at -12 °C. The Chi-square test showed that both the relationship of bamboo sample sources and temperature of the aqueous solution and the relationship of concentration of aqueous solution and bamboo sample sources have no significant relationship. Meanwhile, temperature and concentration indicate relation. The test for heterogeneity displayed a small amount of heterogeneity on the gathered data that may be clinically unimportant thereby making the data considerably homogeneous. The heterogeneity of the data ($I2= 69.52\%$, $P<0.10$) was mainly affected by the extreme temperatures used by the individual studies. Overall, the significant evidence validates the efficiency of NaOH/urea aqueous solution at low temperature in the dissolution of bamboo cellulose.

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