A Fundamental Study on the Influence of Biomass Source and Operation Conditions on the Self-Assembly of Cellulose-Based Hydrogel Beads

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The research behind potential functionality and utilization of cellulose-derived products has expanded drastically over the years, gaining traction with the desire for more environmentally conservative products and procedures to replace those derived from petroleum as well as the characteristic versatility and modifiability of cellulose in application. The study of regenerated cellulose and its production with environmentally conscious methods paves the way for a sustainable alternative to petroleum-based products. Moreover, these products would benefit by the extensive ability of cellulose to be easily functionalized and chemically modified to fit a variety of applications and shapes. (Trygg et al., 2013).

Cellulose beads have been prepared using a variety of starting materials including various feedstocks and regeneration baths. One of the notable differences in determining which cellulose to use is the variation in solubility which determines characteristics that present after formation such as density and porosity (Gomez-Maldonado et al., 2021) (Akalin & Pulat, 2018) (Nie et al., 2021) (Gericke et al., 2013). Solubility is important in the determination of dissolution bath components. The variation in properties available due to the use of different materials has been understudied. Thus, this is an opportunity to better understand the potential when considering the use of various starting materials depending on the desired properties for the final beads.

The primary focus of this research was to understand the functional properties of cellulose in the formation and shaping of hydrogel beads that may be used in applications like drug delivery, water remediation, and molecule separation, as a substitute of plastic beads present in cosmetics, etc. (Gericke et al., 2013) (Niinivaara & Cranston, 2020). This was done by comparing the morphological characteristics between the resultant beads composed of the different cellulose feedstocks and acid regeneration baths.

The cellulose starting materials chosen for comparison in this work were cellulose nanofibrils (CNF) from soybean hulls, wood, and the more traditionally used dissolving pulp. The beads were regenerated in three different acid baths: sulfuric acid, nitric acid, and citric acid.

These hydrogel beads are formed through the dissolution and regeneration of cellulose. A solution composed of a cellulose starting material, sodium hydroxide, urea, and ultrapure water is mixed at low temperatures (-11°C) and then administered dropwise into an acid bath in which the solution coagulates to form small hydrogel beads (Gericke et al., 2013). The beads are then washed using ultrapure water until neutral pH to prevent further degradation during storage (Gomez-Maldonado et al., 2021).

Characterization using scanning electron microscopy (SEM) (Figure 1-8) showed an apparent difference in porosity between beads. This indicates a difference that may be studied further to better understand possible functionalization due to the given properties.

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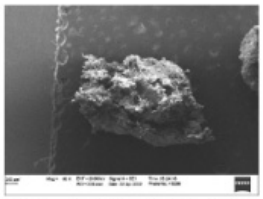

Fig. 1. SEM Dissolving Pulp Nitric Acid

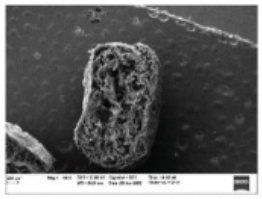

Fig. 2. SEM Dissolving in Pulp in Sulfuric Acid

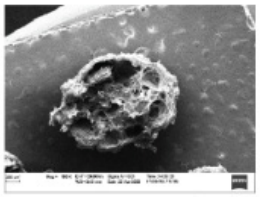

Fig. 3. SEM Dissolving Citric Acid

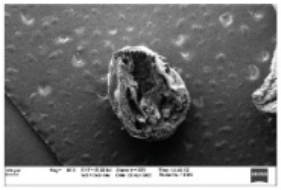

Fig. 4. SEM Wood CNF in Nitric Acid

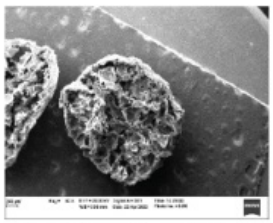

Fig. 5. SEM Wood CNF in Sulfuric Acid

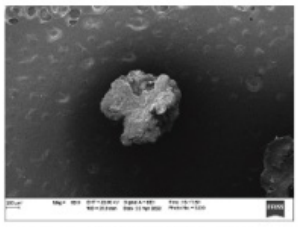

Fig. 6. SEM Wood CNF in Citric Acid

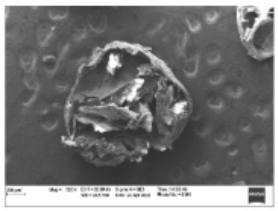

Fig. 7. SEM Soybean CNF in Nitric Acid

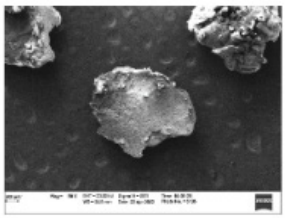

Fig. 8. SEM Soybean CNF in Citric Acid

Fig. 1-8. Depicts the solid content and diameter of each bead produced. Differences can be observed, especially in solid content. The beads produced in the sulfuric acid bath had a higher solid content than the ones in nitric or citric. Despite the differences in porosity and solid content, the diameter of the beads was relatively similar.

Further research needs to be conducted to determine what role these differences could play in the functionalization and utilization of cellulose-based hydrogels formed from various starting materials and regeneration baths.

In future work, we plan to continue to examine the findings presented and explore other properties of the beads such as thermal behavior and surface area. There will also be more in-depth investigations into the potential of these hydrogels in practical applications.

Table 1 Solid content and diameter measurements for each starting material regenerated in each acid bath.

	Nitric Acid	**Sulfuric Acid**	**Citric Acid**
Dissolving Pulp	Solid Content (%)- 8.48 Diameter (mm)- 2.88	Solid Content (%)- 15.62 Diameter (mm)- 3.22	Solid Content (%)- 9.45 Diameter (mm)- 3.07
Wood CNF	Solid Content (%)- 6.01 Diameter (mm)- 2.89	Solid Content (%)- 14.26 Diameter (mm)- 2.94	Solid Content (%)- 11.03 Diameter (mm)- 2.71
Soy-bean CNF	Solid Content (%)- 3.86 Diameter (mm)- 2.60	Solid Content (%)- 21.15 Diameter (mm)- 2.63	Solid Content (%)- 9.56 Diameter (mm)- 2.27

Statement of Research Advisor

Florrie's work is part of a larger effort in my group to understand the structure-properties relationship between sources of biomass and supramolecular structure of cellulose assemblies. In this work, Florrie has developed a strategic experimental design to advance our understanding of pore structure on regenerated cellulose beads, to be evaluated as adsorbents for water contaminants. Florrie has developed a great deal of skills during this period, including time management, data acquisition and analysis.

-Maria Soledad Peresin, College of Forestry, Wildlife and Environment.

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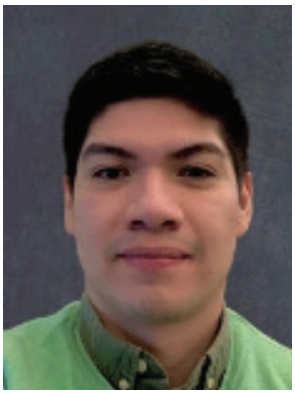
Authors Biography

Florence Adams is a junior-year student pursuing a B.S. degree in Sustainable Biomaterials and Packaging at Auburn University. She has been working as an undergraduate research fellow in the Sustainable Bio-based Materials Laboratory.

Sydney Brake is a Bachelor of Materials Engineering and Master of Forestry. Currently, she is a PhD student in the College of Forestry Wildlife and Environment focusing on fundamental interactions of materials and sustainable fiber processing methods for the development of functional, bio-based textiles with applications such as insecticidal nets and clothing.



Diego Gomez-Maldonado is a Post-Doctoral Research Fellow in the Sustainable Bio-based Materials Laboratory. He is very active in helping undergraduate student expand their knowledge.

Maria Soledad Peresin is an Associate Professor in the College of Forestry, Wildlife, and Environment at Auburn University. She is the principal investigator for the Sustainable Bio-based Materials Laboratory.