

# Absorption of heat cured acrylic resin reinforced with rice husk nanoceluloce (*Oryza Sativa L.*)



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## Abstract

**Objective:** This study aims to determine chitosan as a coupling agent and the PMMA denture base's water sorption with nano cellulose rice husk as reinforcement material.

**Material and Methods:** This type of research is an experimental laboratory. The research sample consisted of four groups, each group consisting of 8 samples selected by simple random. The T1, T2, T3, and Control groups were PMMA with 1%, 2%, three chitosan, and without chitosan. This study uses two kinds of tests, water absorption test and Scanning Electron Microscope (SEM).

**Results:** The heat cured acrylic resin group with 3% chitosan had the lowest water sorption  $15.54 \pm 3.48 \mu\text{m}/\text{mm}^3$ . The heat cured acrylic resin group without chitosan had the lowest water absorption  $32.85 \pm 4.82 \mu\text{m}/\text{mm}^3$ . There was a significant difference in the water absorption test ( $p < 0.05$ ). The SEM showed the homogenous image but still had porosity and agglomeration.

**Conclusion:** This study concludes that addition of chitosan in base denture acrylic has decreased water absorption.

**Keywords:** Chitosan, Coupling agent, Denture base acrylic resin, Nanocellulose rice husk, Water absorption

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## Introduction

Tooth loss can cause problems in masticatory function, speech, facial structure, and facial aesthetics.<sup>1</sup> Denture is needed to correct these deficiencies. The denture base is the part of the denture that is attached to the soft tissue. Denture base materials can be made of metal and non-metal. The material often used in fabrication of denture base material is acrylic resin.<sup>2</sup>

Acrylic resin is a non-metallic denture base material that has been used since 1935. Heat cured acrylic resin is an acrylic heat polymerization resin consisting of powder as polymethyl methacrylate (PMMA) and liquid methyl methacrylate (MMA).<sup>3</sup>

Heat cured acrylic has the advantages of good dimensional stability, good aesthetics, and high thermal conductivity. The disadvantages of heat cured acrylic include low flexibility, high porosity, residual monomers and water absorption.<sup>1</sup> Water absorption in acrylic resin occurs in a humid environment. This occurs due to the polar nature of acrylic resin. Water absorption occurs by separating the weak polymer chains, then a diffusion process occurs with water molecules.<sup>4</sup>

The water absorption of acrylic resin is influenced by polymer properties, degree of conversion, cross-link strength, and hydrophilic properties of the polymer. The water molecules absorbed in the acrylic polymer act as a plasticizer which makes the

polymer soft and results in physical changes of the polymer. In addition, water absorption will affect the mechanical strength, discoloration, and dimensional stability which ultimately leads to fractures and failure of the denture base.<sup>5</sup>

One alternative that can be used in the modification to enhance acrylic properties is the addition of natural fibers.<sup>4</sup> The natural fiber content that can be used as a reinforcement for acrylic resin is nanocellulose.

Nanocellulose has characteristics of nano-sized particles that experience increased crystallinity, aspect ratio, surface area, and increased dispersion and biodegradability.<sup>6</sup> Natural fiber with cellulose content that is easily found is rice husk.<sup>7</sup>

Acrylic is an inorganic material while nanocellulose is an organic material, the combination of these materials requires a mediator called a coupling agent. Coupling agent is a material that is added to enhance the bond between the matrix and the filler.<sup>8</sup> One of the coupling agents that is often used is chitosan. Chitosan comes from chitin derivatives that come from shells of crustacean such as shrimp, lobster and crab.<sup>8</sup>

Chitosan is composed of two main groups, group amina ( $-\text{NH}_2$ ) and the hydroxyl group ( $-\text{OH}$ ). Group  $-\text{NH}_2$  serves as a coatings of other materials that alter the nonpolar groups hydrophilic surface to become hydrophobic, whereas

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the -OH group forming hydrogen bonds that allow an increase in the bond energy between filler and matrix.<sup>9</sup>

Based on the background and limited literature, researcher is interested in conducting research on the effect of adding chitosan as a coupling agent with a concentration of 1%, 2% and 3% on the water absorption capacity of the denture base heat polymerization acrylic resin with reinforcement of nanocellulose rice husk and surface morphological characteristics of the heat cured acrylic.

## Material and Methods

This research is a true experimental research with randomized pre-posttest only control group design. This research was carried out at Nanoscience and Nanotechnology Research Institute (LPNN), Advanced Materials Processing Laboratory, Institut Teknologi Bandung and Laboratory of Inorganic Chemistry Jenderal Soedirman University.

This study used heat cured acrylic mixed with 1% nanocellulose from rice husks. Thirty-two denture base samples with a diameter of  $50 \pm 0.01$  mm and a thickness of  $1 \pm 0.1$  mm were used as research samples which were divided into four groups. The control group (C) is a group of acrylic resin heat cured addition of 1% nanocellulose rice husk without the addition of chitosan. Treatment group 1 (T1) with the addition of 1% nanocellulose rice husk and 1% chitosan. Treatment group 2 (T2) with the addition of 1% nanocellulose rice husk and 2% chitosan. Treatment group 3 (T3) was a group with the addition of 1% nanocellulose rice husks and 3% chitosan.

The work stage begins with the manufacture of nanocellulose from rice husks. The synthesis of rice husk nanocellulose in this study was carried out by the acid hydrolysis method. The rice husks were weighed 60 grams then alkaline hydrolysis with sodium hydroxide (NaOH) 4M. Then rinsed with distilled water until it reached Ph 7, then dried in an oven at 60°C for 24 hours. Next process is bleaching using 2.5% sodium hypochlorite (NaOCl), and then put in a NaOCl solution get cellulose.

The next process is the acid hydrolysis process using  $H_2SO_4$  45% stirrer for 90 minutes. Then rinsed and neutralized with 0.5 M NaOH solution. Then the neutralization results is sonicated and centrifuged and a nanocellulose gel is formed. Nanocellulose gel obtained was centrifuged then dried. The final result in the form of nanocellulose powder is then mashed and weighed.

The work stage was then continued with the preparation of a chitosan solution. The 1% chitosan solution was made by making acetic acid solution with composition of 2 ml of acetic acid and 98 ml of distilled water, then added 1 gram of chitosan and

stirred until homogeneous using a magnetic stirrer. The 2% chitosan solution was made by making an acetic acid solution then added 2 grams of chitosan. The 3% chitosan solution was made by making acetic acid solution then added with 3 grams of chitosan until there was no precipitate.

The next step was the heat cured acrylic and the nanocellulose stirred using an ultrasonic homogenizer, then added with chitosan solution according to the ratio of the factory rules. The polymer was stirred slowly 60 times using a stainless-steel spatula, then the mixing jar was closed and waited until it reached the dough stage. The acrylic resin was then put into a mold that had been smeared with CMS, then cured at 100°C for 30 minutes using a water bath. After that the mold was let to cool down to room temperature.

Heat cured acrylic samples are done finishing and polishing. Then all samples were desiccated using a desiccator for 24 hours. Then the sample that has been put into the desiccator is taken and weighed to get the weight before soaking (Pre).

One sample was taken using simple random sampling for SEM test before immersion. All samples were then immersed in artificial saliva for 7 days at 37°C, and carried out post-immersion SEM tests. Measurement of water absorption was carried out using the formula:<sup>5,10</sup>

$$\text{Water absorption capacity} = \frac{m_2 - m_1}{v}$$

Description:

$m_2$  = mass after immersion ( $\mu\text{g}$ )

$m_1$  = mass after the first drying ( $\mu\text{g}$ ).

## Results

Based on the results of the calculation of the water absorption test in all samples in [table 1](#).

The results of the water absorption value were then tested statistically the normality was performed using the Shapiro-Wilk test. The significance value (p) of all groups is more than 0.05 ( $p > 0.05$ ). The data homogeneity test was done using the Levene's test. Based on the homogeneity test, the data showed that the p value was  $p > 0.05$ . Data that were normally distributed and homogeneous were analyzed statistically parametric using One-Way ANOVA. The One-Way ANOVA test results obtained the water absorption value in [table 2](#).

The One-Way ANOVA test results show a p value of 0.000 ( $p < 0.05$ ), which means that there is a significant difference between the water absorption value of the sample group. Test Post Least Significant Differences (LSD) was carried out to identify groups with significant differences. LSD test results can be seen in [table 3](#).

**Table 1 Mean and standard deviation of water absorption values of denture base heat cured acrylic polymerization**

No	Sample Group	Water sorption (µm / mm <sup>3</sup> ) ± SD
1	Group T1 1% Chitosan and 1% Nanocellulose	28.66 ± 3.74
2	Group T2 2% Chitosan and 1% Nanocellulose	22.27 ± 2.61
3	Group T3 3% Chitosan and 1% Nanocellulose	15.54 ± 3.48
4	Control Group 1% Nanocellulose and without chitosan	32.85 ± 4.82

Source: Data processed primer, 2020.

**Table 2 One-Way ANOVA test results**

No	Group	Number of samples	Value p
1	T1	8	0.000*
2	T2	8	
3	T3	8	
4	Control	8	

Source: Processed primary data, 2020.

Note: \* -there is a significant difference (p≤0.05)

**Table 3 Post Hoc LSD test results water sorption**

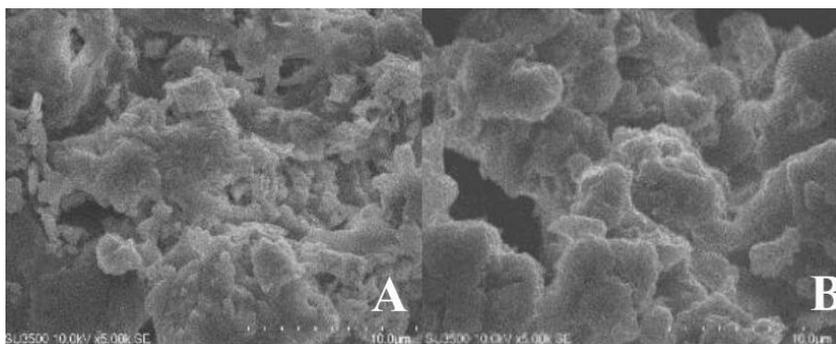
Group	T1	T2	T3	C
T1		0.009**	0.000**	0.122
T2			0.000**	0.000**
T3				0.002**
C				

Source: Processed primary data, 2020.

Note:

a. \* -there is a significant difference (p≤0.05) between groups

b. \*\* - there is a very significant difference (p≤0.01) between groups



**Figure 1** A. SEM Results PreandPost,B.Immersion group T1 with magnification of 5000times. Source: Processed Primary Data 2020

The LSD test results in Table 3 show that the T2 T3 treatment group showed a very significant difference (p≤0.01) compared to the control group.

The results of the Scanning Electron Microscope (SEM) pre and postgroupwith the addition of

1%, 2%, 3% chitosan and without the addition of chitosan can be seen in the figure below.

The morphological characteristics of the T1 group in figure 1A show the porous diameter in the sample Pre was 0.846-1.709 µm while the diameter of the sample Post was 1.286-1.661 µm. In the sample Post figure 1B the agglomeration is less than that of the sample Pre.

The morphological characteristics of the T2 group in figure 2A show the porous diameter in the sample Pre was 0.370-0.840 µm while the diameter of the samplePost was 0.956-1.649 µm figure 2B.

The morphological characteristics of the T3 group in figure 3A show that there is a homogeneous picture compared to the T1, T2, and control groups. The porous diameter in the Pre sample was 0.458-0.677 µm while the Post sample was 0.420-0.851 µm figure 3B. The T3 group looks more homogeneous and has less agglomeration than the T1 T2 and Control groups.

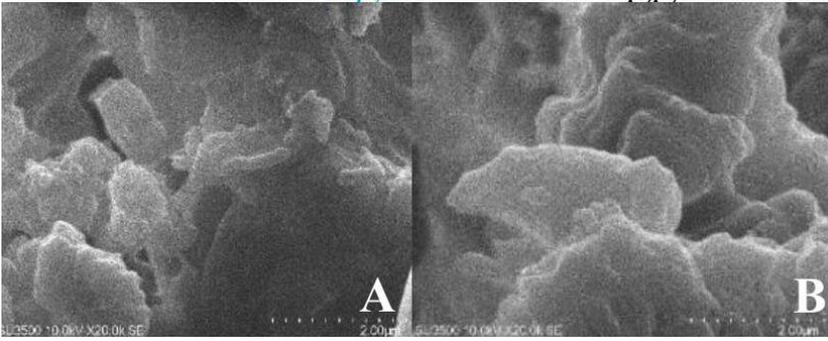
The results of the morphological characteristics of the control group in figure 4A show that porous diameter of the Pre samples was 0.472-1.365 µm while the Post samples were 0.474-0.923µm. The Post figure 4B sample shows greater porosity and more agglomeration.

## Discussion

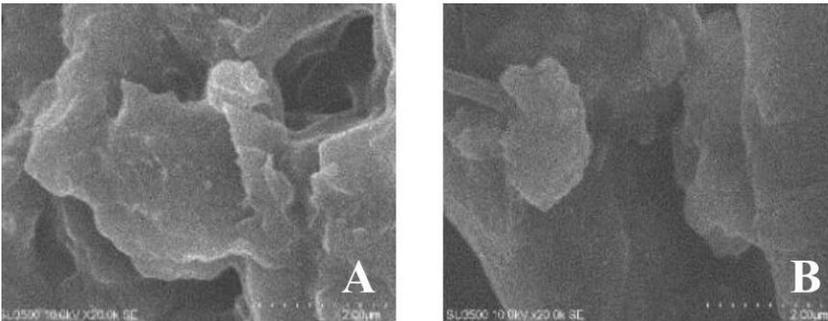
In the results of this study, the group with the lowest water absorption was in the group with the addition of 3% chitosan and 1% nanocellulose and the highest in the group with the addition of 1% nanocellulose and without the addition of chitosan. As has been presented by the ISO specification no 1567 of 1999, the water absorption in heat polymerized acrylic resin should not exceed 32 µg/mm<sup>3</sup>.<sup>10</sup>

The highest water absorption value in this study was found in the group that added 1% rice husk nanocellulose without the addition of chitosan. The high rate of water absorption is due to the hydrophilic properties of acrylic and nanocellulose resins and the tendency of agglomerated rice husk nanocelluloses. Acrylic resin consists of polymethyl methacrylate which has an ester group (R-COOR'), that has hydrophilic properties. Nanocellulose has a hydroxyl group (-OH) which is hydrophilic in its molecule so that the oxidation process occurs.<sup>9</sup>

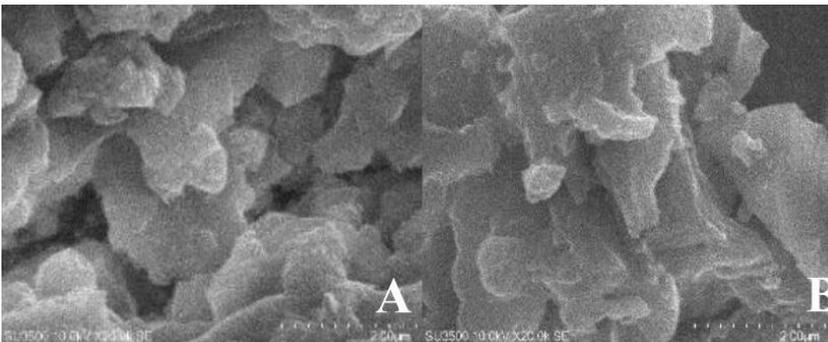
This ion exchange reaction will cause a small porous (microporosity). Microporosity causes water molecules to enter between the chains of the molecule's chains and causes the polymer chains to be filled with water. The chain that is filled with water causes a change in mass which indicates water absorption.<sup>11</sup>



**Figure 2** A. SEM Results Pre and Post, B. Soaking group T2 with a magnification of 20,000. Source: 2020 Primary Data Processed



**Figure 3** A. SEM Results Pre and Post, B. Immersion in the T3 group with a magnification of 20,000 times. Source: 2020 Primary Data



**Figure 4** A. SEM Pre and Post, B. Immersion in the Control group at a magnification of 20,000 times. Source: 2020 Processed Primary Data

Another cause of water absorption is the presence of agglomeration of nanocellulose which reduces the ability of nanocellulose to fill internal porosity. Nanocellulose has several characteristics, including having a high surface area. The nanometer size of the nanocellulose particles and the active nature of the nanometer will fill the gaps and help the matrix and bond filler.<sup>12</sup>

The lowest water absorption value in this study was in the group that was added with 3% chitosan and 1% nanocellulose (T3). In this group, it appears that chitosan in this study acts as a coupling agent. The function of the coupling agent includes creating chemical bonds between the matrix and the filler.<sup>8</sup> The decrease in water absorption is due to

the higher chitosan concentration. The higher the chitosan concentration used, the more particles that interact with each other, the lower the ability to absorb water.<sup>11</sup>

The concentration of chitosan is directly proportional to its viscosity. The high viscosity causes the friction forces between the particles in the chitosan solution to increase and the more attractive to one another which is influenced by the strong cohesion between particles. The content of bioactive substances (-NH<sub>2</sub>) can increase the chemical bond. The strong surface energy makes the polymer highly hydrophobic. The higher the hydrophobic nature of a polymer, make the polymer difficult to absorb water in wet or humid conditions.<sup>13</sup>

This is in line with the research conducted by Tekaleet al.<sup>5</sup> which concluded that the addition of a coupling agent as a nanoparticle strengthening effect on water absorption in acrylic resin.<sup>5</sup> In the SEM of the T3 group, it was also seen that more homogeneous. This is due to the fact that many bioactive compounds are formed and are stable. In this group there was little agglomeration, which indicates that the concentration of chitosan affects the distribution of nanocellulose more evenly.<sup>9</sup>

## Conclusion

Based on the research that has been done, it was found that the addition of chitosan as a coupling agent in the denture base of heat cured acrylic can reduce water absorption. The lowest average value of water absorption was in the group of addition of 3% chitosan (T3) of  $15.54 \pm 3.48 \mu\text{m} / \text{mm}^3$  and the highest in the group without the addition of chitosan (C) was  $32.85 \pm 4.82 \mu\text{m} / \text{mm}^3$ .

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## Conflict of Interest

The authors report no conflict of interest.

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