CELLULOSE CHEMISTRY AND TECHNOLOGY

EFFECT OF LIGNOCELLULOSIC FILLER ON MECHANICAL AND RHEOLOGICAL PROPERITES OF ALGINATE-BASED DENTAL IMPRESSION MATERIALS

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Dental alginate impression materials have some disadvantages, such as poor dimensional stability due to syneresis and imbibition, fast setting time in a hot environment, low tensile resistance and poor rheological properties. In this study, lignocellulosic material was incorporated as a filler into commercially available alginate impression material. The powder was mixed in different ratios (2%, 4%, and 6%) to prepare different experimental materials (E1, E2, and E3, respectively). The samples were tested to evaluate their dimensional stability, setting time, tensile strength and rheological properties. The dimensional stability significantly increased by the addition of lignocellulosic powder (2%, 4%) to alginate, as compared to control groups. However, the addition of 6% lignocellulosic powder decreased the dimensional stability, as compared to the control. Also, the mean values of setting time, tensile strength and viscoelasticity were the highest in E3 series, followed by E2 and the least in the control groups. The mean value of the flow and drip properties was reduced statistically by adding increasing concentrations of lignocellulosic powder to alginate. Thus, the addition of lignocellulosic powder to alginate impression material improved the dimensional stability, tensile strength, setting time, viscoelasticity, but reduced the flow properties.

Keywords: lignocellulose, alginate impression material, dimensional stability, setting time, mechanical properties, rheological properties

INTRODUCTION

In dentistry, prime importance should be given to the accuracy of the impression for the fabrication of a successful and well-fitted prosthesis. The impression material should be ideally capable of reproducing fine surface details and be stable in dimensions over time to allow the operator to pour the impression at convenience. Irreversible hydrocolloids are most commonly used as impression materials for the fabrication of prosthesis, as well as for treatment planning in dentistry. In dental practices, alginate is one of the most commonly used impression materials due to its numerous advantages, including its low density, good aesthetics, low cost, ease of manipulation, hydrophilic nature, and good patient acceptance.[1] However, alginate impression materials also have several limitations, such as poor dimensional stability, limited elasticity, rapid setting time, and low tensile strength.[2] Syneresis (water exudation) and imbibition (water absorption) that occur over time may be responsible for causing dimensional instability. Therefore, these impressions are not suitable for storage and should be poured immediately or within a few minutes.[3]

The properties of impression materials, such as dimensional stability, setting time, flow properties, viscoelasticity and tensile strength, are essential factors in their longevity. The requirements for good quality impression materials also include excellent tissue surface details and disinfectability. Several studies have investigated the impact of water quality and quantity on the mechanical properties of alginate impression materials, demonstrating that both excessive and insufficient water levels can alter their performance. Raszewski *et al.*[4] found that water rich in calcium ions improves the mechanical properties of alginate. Choudhury *et al.*[5] reported that the addition of various disinfectants, including sodium fluoride, povidone-iodine powder and some other materials, led to improved antimicrobial property of the material, but did not show any statistically significant difference in the dimensional stability. The effect of storage conditions was studied and the results revealed that environmental moisture can cause unfavorable changes in dimensions. When examining the effect of storage time, it was found that the dimensional stability calculated after minimum 1 hour to maximum 120 hours showed statistically significant difference.[6] Shafiq *et al.*[7] studied the dimensional accuracy of casts obtained from alginate impression material after 1 hour and 2 hours delayed pouring. Another study carried out by Walker *et al.*,[8] who evaluated the casts after 24 hours and 48 hours delayed pouring. Results demonstrated a marked difference in dimensional stability values of casts obtained at various time intervals and the conventional alginate (JeltratePlus) changed dimensionally by -0.33% between 30 minutes and 48 hours. Garrofe *et al.*[9] evaluated the dimensional stability of different irreversible hydrocolloids by taking photographs at different time intervals up to 96 hours. Statistical evaluation showed that the dimensional stability of irreversible hydrocolloid impression materials is significantly affected by time.

Cellulose, hemicelluloses and lignin, the polymeric constituents of lignocellulose, are among the most abundant biopolymers in the plant world, and can be obtained as a by-product from the pulp and paper or wood processing industries. Due to the abundance of lignocellulosic biomass, its use has been of interest for various applications, such as in the production of biofuels[10] and composites.[9] It has been remarked that, in composites, lignocellulosic fibers act as reinforcement, adding mechanical strength to the matrix material, while lignin can serve as a binder. Gouveia *et al.*[9] reported that adding lignin to medium density fiber boards led to improved mechanical properties and dimensional stability of the composite materials. The binding ability of lignin remarkably improved the dimensional stability of fiber boards. Water sorption and thickness swelling properties were also significantly decreased.

In this study, lignocellulosic biomass particles were added in various concentrations to an alginate impression material to improve its dimensional stability, setting time, tensile strength, viscoelasticity and flow properties, which, to the best of the authors' knowledge, has not been reported so far. The lignocellulosic powder was mixed in different ratios (2%, 4%, and 6%) in commercially available alginate impression material. Then, the prepared experimental materials were investigated in terms of dimensional stability, setting time, tensile strength and rheological properties in accordance with ISO standards.

EXPERIMENTAL

Materials

Alginate impression material was purchased from Hygedent, South Korea (batch No. 21563). Distilled water (99.5% purity) and NaOH solution (molecular weight 40 g/mol) were obtained locally.

The lignocellulosic material (LM) was obtained as a sawdust residue from a local *Dalbergia sissoo* wood processing plant. The sawdust particles were passed through a sieve (Endecotts, UK) to obtain uniform size of 200 µm.

Pretreatment of lignocellulosic material and sample preparation

The obtained LM (200 µm particle size) was then treated with 1% NaOH solution at room temperature for 2 hours, in order to increase its porosity to ensure its effective interaction with commercial alginate.[11]

Commercial alginate powder was mixed mechanically with distilled water at room temperature (25 °C) in a rubber bowl for 1.5 min according to the manufacturer's instructions – and was considered as control. For experimental groups, the lignocellulosic powder was added to the alginate powder in a ratio of 2, 4 and 6% to make formulations E1, E2 and E3, respectively. For experimental samples, 5 g of alginate powder and 20 mL of water were mixed with a spatula,[12] in a 100 mL bowl for 1.5 minutes. The samples and their denotations are tabulated in Table 1.

Characterization of lignocellulosic material (LM)

The lignocellulosic sawdust particles were characterized using SEM and FTIR spectroscopy. For morphological analysis, a JSM-IT-100 Scanning Electron microscope was used. The LM samples were placed on aluminum stubs under an argon atmosphere and sputter coated for 90 seconds and 10 mA current. After coating, micrographs were recorded at magnifications of 95x and 1000x at an accelerated voltage of 15 KV.

Fourier Transform Infrared (FTIR) spectroscopy was performed using a Thermo Scientific Nicolet iS10 (US), equipped with Attenuated Total Reflectance (ATR) for identifying the characteristic functional groups of lignocelluloses. Spectra were recorded in the range of 600-4000 cm^{-1}.

Table 1
Composition of control and experiments groups

Code	Groups	LM loading
C	Control (commercial alginate impression material)	0% LM
E1	Experimental group 1	2% LM
E2	Experimental group 2	4% LM
E3	Experimental group 3	6% LM

Mechanical properties of samples

Dimensional stability

The dimensional stability was assessed using the protocol described by Walker *et al.*[8] Following the manufacturer's instructions, alginate was mixed with distilled water and then placed into a polyvinyl carbonate (PC) ring mold, with dimensions of 6 mm in height and 26 mm in diameter, allowing it to set for 1 minute. After application of separating medium to the glass plate, the PC ring mold was placed over it. A clean metal test die sprayed with separating agent (wax) was pressed in PC ring mold center. In order to simulate oral environment, the whole assembly was placed in a water bath of distilled water having 35 °C temperature and 1 kgf/cm^2 pressure for 3 minutes. Then, the assembly was removed from the bath and the metal die was separated from the impression and rinsed thoroughly. The impression was stored in a plastic bag at ambient laboratory temperature. Dimensional stability was then evaluated by measuring the horizontal line of the impression 3 times to the nearest 0.01 mm with a caliper.[13] The mean of these 3 horizontal measurements of the impression was compared with the middle line of the die. The measurement was repeated three times. The following formula was used to calculate percentage dimensional change:

Percentage dimensional change = [(A–D)/D] x 100 (1)

where A is the mean of alginate impression measurement, D is the die measurement.[8]

Tensile strength

Tensile strength was assessed using the method described in ISO 3349:2009. The alginate powder and liquid were mixed and the mixture was placed into a PC ring, as described above. After three minutes of setting, a five-centimeter sample was cut at a longitudinal notch and put in a UTM (St. Louis, MO, USA). Tensile strength was measured and machine tension was applied at a rate of 0.5 mm per minute,[14] as shown in Figure 2. Equation (2) was used to determine the tensile strength:

T (tensile stress) = F (force)/A (area) (2)

where T = stress (N/m^2), F is the maximum force recorded just before failure, A is cross-sectional area.

Physico-chemical properties

Setting time

ISO 1563:1990 was used to calculate the setting time of alginate and the experimental materials. Specimen molds (pipe) having 30 mm diameter and 16 mm height, made from plastic, were placed on a glass plate. The alginate powder (control), lignocellulosic materials (experimental groups) and liquid were mixed at room temperature (26 °C) and poured into these molds. After 2 minutes of setting, flash was removed with a wax knife to level with mold height. An acrylic stick with 10 cm in length and 6 mm in diameter was use to touch the alginate surface in the molds. Setting measurement was assessed at 10 second interval with the acrylic stick. Alginate setting time was counted starting with alginate mixing until no stickiness was found. The experiments were repeated three times.[15]

Consistency evaluation

The flow test was performed using the method described in ISO 4823-2021. To assess the flow behavior of the alginate impression material and the experimental materials, a 1 mL syringe was used to dispense the sample onto a hydrophilic silica glass plate. The material was allowed to set for 3 min, the sample diameter was measured with a digital caliper (NSK, Tokyo, Japan). The difference between T_0 (immediately after mixing) and after 3 minutes was the flow value. 10 samples of the control and of each formulation were tested.

Drip test

ISO 4823-2021 was used to perform the drip test for the control and experimental samples. A 1 mL syringe was used to pour 0.1 mL of the impression material on a hydrophobic glass plate. After pouring in a specified spot, the glass plate was placed in perpendicular position to allow the sample to drip for 5 minutes. The lower end of the dripped material was marked with a line, and the distance from the center of the upper end to the lower end line was measured. 10 samples of the control and of each formulation were tested.

Rheological evaluation

The viscoelastic characteristics of the control and the modified impression materials were assessed using a rheometer (Anton Paar MCR 301). A dynamic oscillation-time sweep test was conducted using two parallel aluminum plates, set 400 mm apart with a diameter of 10 mm. After placing 0.15 mL of the impression material onto the plate heated to 30 °C, the changes in storage modulus (G'), loss modulus (G") and loss tangent (tan δ), at a maximum torque of 2000 Nm, were measured. The time from the beginning of mixing and the rheometer measurement was under a minute. For rheological evaluation as a function of time (during 5 min), a 1 Hz oscillating frequency was used to record the storage modulus (G'), loss modulus (G") and loss tangent (tan δ).

RESULTS AND DISCUSSION

Characterization of LM

Fourier-transform infrared spectroscopy (FTIR)

The characteristic bands of lignocellulose can be noted in the spectrum of the *Dalbergia sissoo* wood sawdust particles shown in Figure 1. The stretching vibration band of OH at 3346 cm^{-1}, the stretching of CH_2 at 2930 cm^{-1}, the stretching of the C=C benzene ring at 1600 and 1506 cm^{-1}, the deformation peak of the C–H aromatic ring at 1416 cm^{-1},[16] stretching of C=O in hemicelluloses at 1728 cm^{-1}, stretching for C-O-C at 1320 cm^{-1} and 1196 cm^{-1}, as well as C–O stretching in cellulose and hemicelluloses at 1020 and 819 cm^{-1},[17] can be remarked in Figure 1.

Scanning electron microscopy (SEM)

Scanning electron microscopy (SEM) is characterization technique used to analyze the surface properties and micro-/nanostructure of solid samples.[18] Figure 2 shows the SEM micrographs of lignocellulosic particles (of 200 µm particle size) at magnifications of 95X and 1000X. The images reveal particles of irregular morphology and shape, with a tendency to form agglomerates. The particles have sizes in the range of 100-150 µm. The materials have a rough surface, with irregular morphology, similarly to the findings reported by Orelma *et al.*[19]

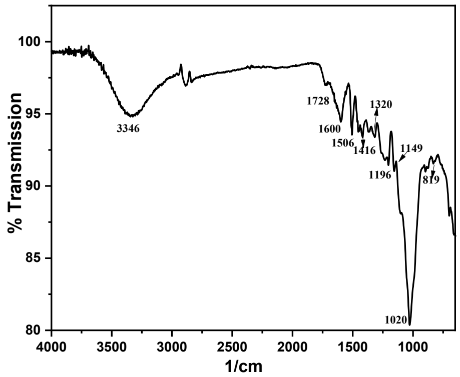


Figure 1: FTIR spectrum of lignocellulosic material

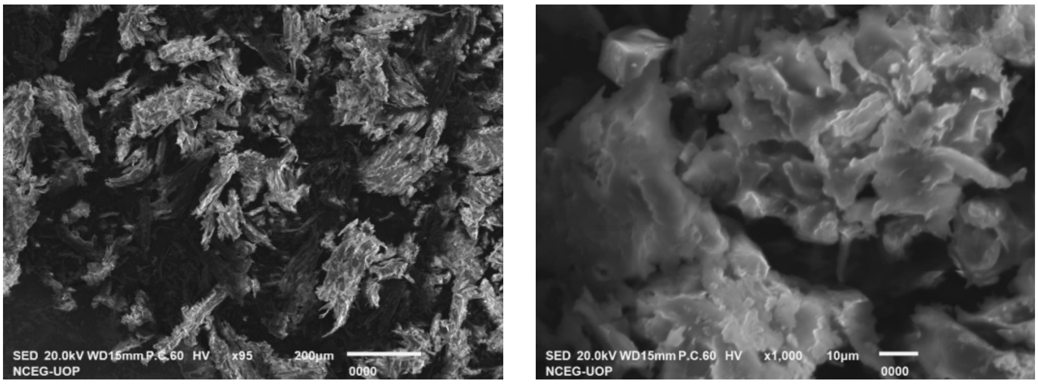


Figure 2: Micrographs of lignocellulosic material at different magnifications

Mechanical properties of experimental materials

Dimensional stability

Figure 3 shows the dimensional stability (%), with respect to shrinkage/expansion, of the control and experimental groups after 1, 2, 4 and 6 hours. The percentage dimensional stability of the control group decreased after 1, 2, 4 and 6 hours by -4.5±0.3, -4.3±0.3, -9.3±0.4 and -12.6±0.4 %, respectively. As regards the behavior of experimental materials over time, their behavior differed depending on the lignocellulosic powder loading. Thus, the percentage dimensional stability of E1 series decreased as follows: -2.3±0.3, -2.7±0.3, -4.4.3±0.4 and -4.4±0.4 % after 1, 2, 4 and 6 hours, respectively. For E2, the decrease was less significant, with values of -0.6±0.3, -0.8±0.3, -1.4±0.4 and -1.2±0.4 % measured after 1, 2, 4 and 6 hours, respectively. However, in the case of higher loading – E3 series –, an increase in percentage dimensional stability was recorded, as follows: 2.8±0.3, 2.1±0.3, 4.7±0.4 and 5.2±0.4 %,. after 1, 2, 4 and 6 hours, respectively. Thus, at every time point, the highest shrinkage was measured for the control group, followed by E1 and E2, while E3 showed expansion of the material (Fig. 3).

The dimensional stability significantly increased upon addition of increasing loadings of lignocellulosic powder to alginate, as compared to the control groups at baseline ($p<0.001$), one hour ($p<0.001$), two hours ($p=0.022$), 4 hours ($p<0.001$) and 6 hours ($p<0.001$). However, the addition of LM (6%) decreased the dimensional stability with p-value<0.001, compared to the control.

Manufacturers recommend that alginate impression should be converted to cast within 15 minutes and it should be kept in 50% humidity medium in a wet towel. It should not be immersed in disinfectant liquid, rather it should be sprayed with disinfectants. Other recommendations for optimizing dimensional stability of alginate include careful handling to avoid distortion due to heavy weight of dental stone, if not supported by the border of trays.[20,21] Tray customization for border extension will be required. Alginate impression is unable to reproduce the anatomy of the maxillary buccal vestibule in the distal area and the mylohyoid fossa in the lower arch.[22,23] Still, it has been noted that alginates with a lower ratio of calcium to sodium lose less water, compared to alginates with a higher ratio of calcium to sodium, and they exhibit greater dimensional stability.[24] Also, alginates containing higher ratios of filler to alginic polymer showed improved dimensional stability.[25]

Setting time

Figure 4 shows the mean setting time of the control and the experimental groups. The highest setting time was observed for group E3 (4.81±0.19 minutes), while the lowest value was observed for the control group (3.36±0.23 minutes).

Alginate impression materials can be classified into two types based on their setting time: type I – fast setting, having 2 minutes setting time, and type II – normal, having 4.5 minutes setting time.[26] Previous literature showed that both manipulation and altering the composition of the alginate mixture can prolong its setting time. The ratio of alginate powder and water is very important for optimal setting time.[27] The most reliable way is to control additive concentrations for optimizing the setting time of alginate. Thalib *et al.*[27] conducted a study to determine the effect of the amount of accelerator (calcium sulfate) in alginate impression materials on setting time. Their results showed that when using 17 g of accelerator, the setting time was 3.1 minutes, while with 15 g of accelerator the setting time increased to 6.1 minutes. Another study also confirmed that reduced accelerator addition can prolong the setting time as compared to the control group.[26]

Other aspects should also be considered when fabricating alginate materials.[28] Cooled water can decrease the collision among the molecules during chemical reaction and prolong the setting time.[29] Previous literature reported that the water temperature in the range of 13-28 °C can have a significant effect on the setting time.[30]

The quality of water, specifically, its mineral contents, can also affect the setting of alginate. Bradna *et al.*[26] showed that tap water leads to a lower setting time, compared to distilled water, and increased concentration of cations (Na^+, Ca^{2+}, and Al^{3+}) can accelerate the setting of alginate impression materials.[31]

Tensile strength

The highest tensile strength was found for E3 samples with maximum added lignocellulosic powder (0.44 ±0.05 N/m^2) and the least was recorded for the control group (0.26±0.08 N/m^2). Figure 5 visually illustrates that the tensile strength of alginate increases as the concentration of lignocellulose loading increases. The results indicate that the lignocellulose filler acts as reinforcement, improving the tensile strength of alginate impression materials.

One of most common drawbacks of alginate impression materials is their low tensile resistance. In undercut areas and unsupported trays, the alginate yields to tensile stress easily, and because of this, it is not recommended as impression material for fixed prostheses and obturator fabrication, like for cleft palate *etc*. In the literature, different approaches are mentioned to improve the tensile resistance of alginate impressions.

A study was conducted on compare the tensile strength of four irreversible hydrocolloid (alginate) impression materials (Tare-Free Alg, Jeltrate, Identic, and Kromopan). Their results showed the difference between brands was statistically significant.[32]

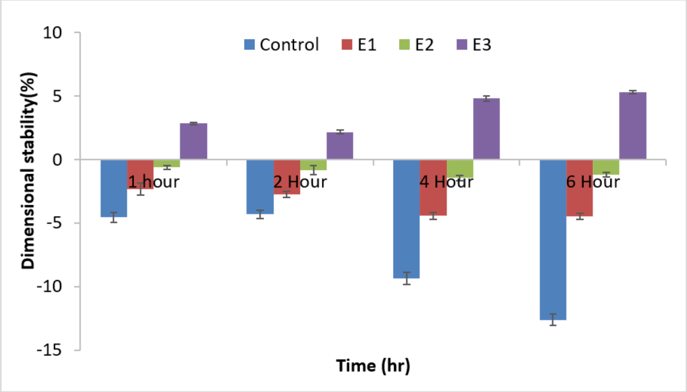


Figure 3: Dimensional stability (%) of control and experimental groups (E1, E2 and E3) at 1, 2, 4, and 6 h (n=5)

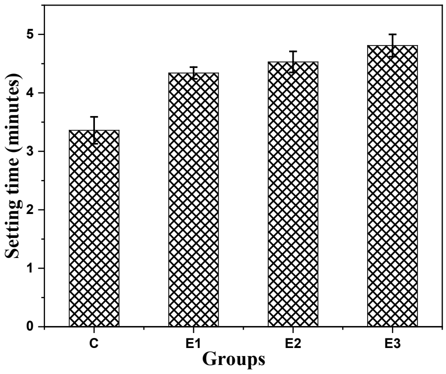


Figure 4: Setting time of control and experimental groups

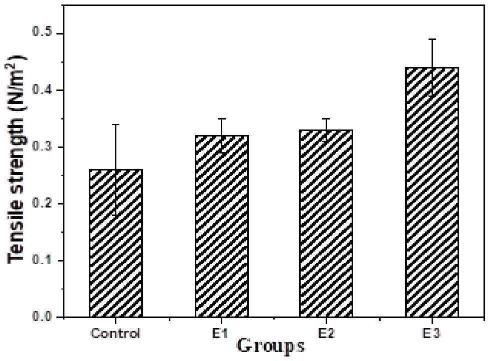


Figure 5: Tensile strength of control and experimental groups

Table 2
Mean and ±SD values of Young modulus, deformation, and tensile strength

Sample groups	Tensile strength (N/m^2)	Young modulus (MPa)	Plastic strain (%) (deformation)
Control group	0.26±0.08	0.0134±0.012	70.648±6.002
E1	0.32± 0.03	0.0216±0.014	55.143±6.206
E2	0.33± 0.02	0.0231±0.005	42.588±2.807
E3	0.44 ±0.05	0.0278±0.017	37.953±2.896

Another study showed that increasing the powder/liquid ratio in alginate impression material can improve the tensile strength of alginate statistically, but not clinically.[33]

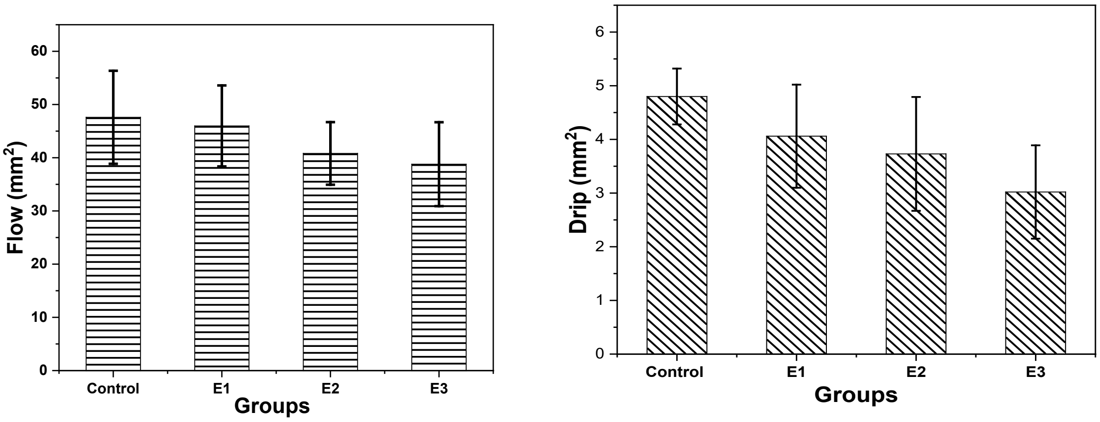
Flow and drip testing

Figure 6 (A and B) shows the flow and drip properties, respectively, of the control and experimental groups. High drip and flow values were observed for the control group (C), followed by E1, E2 and E3, respectively.

Good flow and drip properties of alginate impression materials are important as they allow obtaining a more accurate impression, with elaborate tissue details. In the present study, the control group shows the highest flow and drip values. By adding lignocellulosic powder to the alginate, the flow and dripping of alginate decreased. This is because lignocellulosic particles are light weight and have irregular shape, showing resistance to flow and dripping, thus these properties decreased, while viscosity increased.

In a study by Anwar *et al.*, the authors incorporated various kaolin clay concentrations in alginate, and it was found that the samples with the highest (8%) loading of kaolin showed the highest mean value of flow, followed by the other samples in decreasing order of the loading (4%, 2%), and the control groups with the lowest mean value of flow. The same behaviour was observed for the drip, as the drip rate and the mean value of drip decreased with decreasing loading of clay. Thus, by adding kaolin clay to an alginate impression material, higher flow and drip properties were obtained. This can be explained by the fact that kaolin is inorganic, with spherical particle shape and high weight, so increasing its percent addition improved the flow and drip properties of alginate.[34]

Halim *et al.* reported the highest flow of an alginate impression material as 69.70 mm,[26] whereas in the present study, the highest flow was 46.60 mm. Sastrodihardjo *et al.* studied the flow characteristics of various commercial alginate materials, and found that these were affected by the addition of the Betadine gargle solution.[35]



A

B

Figure 6: Flow (A) and drip (B) properties of control and experimental groups (E1, E2 and E3)

Viscoelasticity

The storage modulus of the control and modified alginate impression materials as a function of setting time is shown in Figure 7. The storage modulus G′ increased over time and recorded the highest value for E3 (6%) group. Meanwhile, the control group had the lowest value. It should be remarked that the pattern of the storage modulus of the control and was different from that of E1, *i.e.*, it showed a slow increase over time.

The loss modulus of the control and the experimental materials during the setting phase was illustrated in Figure 8.

Over time, the loss modulus G″ exhibited an increase, with the highest values observed in E3 (6%). In contrast, the control group registered the lowest values. Furthermore, the behavior of the loss modulus for E1 and E2 displayed differences, characterized by a gradual increase over time.

The loss tangent, represented as tan δ, showed a rapid decrease over time. Notably, in the control and E1 groups, this decline occurred quickly up to the 140-second mark, while in the E2 and E3 groups, it followed a more gradual descent up to 190 seconds, as depicted in Figure 9.

The addition of lignocellulosic material powder, which served as filler particles and enhanced the physical properties, increased the viscoelasticity of the alginate impression material. The loss modulus and storage modulus increased, while the loss tangent decreased as the filler addition was raised. Similar findings have been reported by H. Malektaj *et al.*, when incorporating varying amounts of $CaCl_2$ and montmorillonite clay as cross-linkers to produce a variety of alginate–MMT nanocomposite hydrogels. As montmorillonite clay and alginate chains form physical bonds, the elastic moduli increased linearly with the concentration of montmorillonite clay at all pH values investigated.[36]

L. Singer *et al.* assessed the elastic recovery of experimentally produced self-disinfecting dental alginates. To create antimicrobial-modified dental alginate groups, alginate powder was mixed with 0.2% silver nitrate ($AgNO_3$ group) or 0.2% chlorohexidine solution (CHX group), as opposed to pure water. Additionally, a third modified group was prepared using *Boswellia sacra* (BS) extract oleoresin. The extract was used to reduce silver nitrate to produce silver nanoparticles (AgNPs). According to the results, every tested group exhibited elastic recovery values for elastic impression materials that were within an acceptable range and in compliance with both

ADA and ISO standards. By adding CHX, $AgNO_3$, and BS + AgNPs as filler particles, elastic recovery was enhanced.[37] Other studies also demonstrated similar effects of the fillers in alginate, considering the mixing technique, filler content, or powder-liquid ratio.[38,39,40,41]

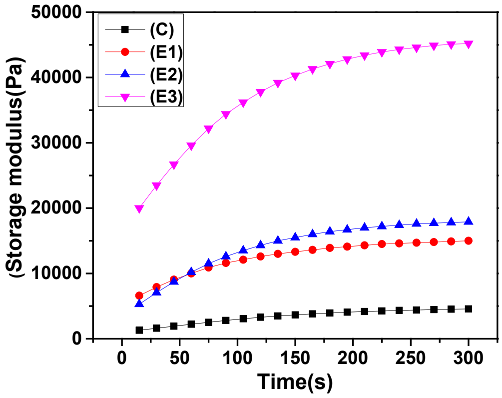


Figure 7: Storage modulus (G′) of control and experimental groups (E1, E2 and E3)

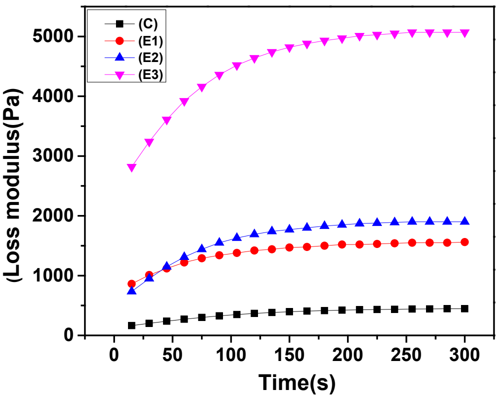


Figure 8: Loss modulus (G″) of control and experimental groups (E1, E2 and E3)

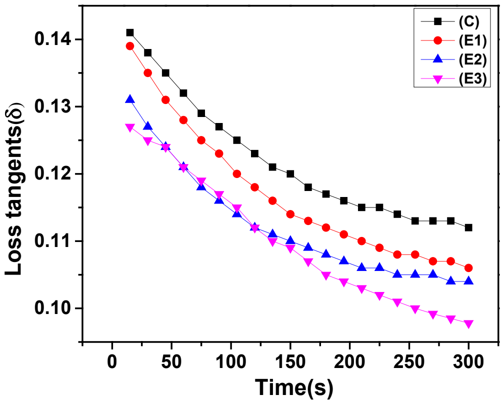


Figure 9: Loss tangent of control and experimental groups (E1, E2 and E3)

Statistical analysis

Data were analyzed using R package version 4.1.3; analysis was done by using “tidyverse” in R, while graphs were created using “ggplot2”. Means and SD were calculated for continuous data like dimensional stability. To assess the mean difference of dimension stability, setting time, tensile strength and flow properties between groups for each stage (immediate, 1-, 2-, 4-, and 6-hours storage), separately one-way ANOVA was performed. To find the significance of the results, Tukey’s test was performed; a p-value ≤ 0.05 was considered significant for each test.

The limitations of this study include the fact that *in vivo* tests should be performed in order to find out the clinical efficacy of the materials; also, the effects of saliva would need to be considered in future work.

CONCLUSION

The lignocellulosic material (LM) was uniformly mixed with alginate, to prepare experimental impression materials. The characteristic peaks of lignocellulose were observed by FTIR analysis. The lignocellulosic filler was added in loadings of 2%, 4% and 6%, and its effects on dimensional stability, tensile strength, setting time, young modulus, deformation and viscoelasticity of experimental samples were studied. The test results showed that the filler addition to alginate prolonged the setting time (E3 > E2 > E1 > C), while the flow and drip properties decreased (E3 < E2 < E1 < C). The addition of the lignocellulosic filler improved the dimensional stability, except in group E3 – with the highest filler loading. Among all samples, the least dimensional change was observed for E2. Also, the tensile strength and viscoelasticity properties increased with the addition of LM (E3>E2>E1>C).

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REFERENCES

[1] G. Cervino, L. Fiorillo, A. S. Herford, L. Laino, G. Troiano *et al.*, *Mar. Drugs*, **17**, 1 (2018), https://doi.org/10.3390/md17010018

[2] S. Mousavi, M. Rahbar, F. Rostamzadeh, K. Jafaria and S. Hekmatfar, *Pesqui. Bras. Odontoped. Clin. Integr.*, **19**, 137 (2019), https://doi.org/10.4034/PBOCI.2019.191.60

[3] M. E. Sayed and P. Gangadharappa, *Indian J. Dent. Res.*, **29**, 477 (2018), https://doi.org/10.4103/ijdr.IJDR_426_17

[4] Z. Raszewski, A. Nowakowska-Toporowska, J. Weżgowiec and D. Nowakowska, *Dent. Med. Probl.*, **55**, 43 (2018), https://doi.org/10.17219/dmp/82179

[5] G. K. Choudhury, R. Chitumalla, L. Manual, S. K. Rajalbandi, M. S. Chauhan *et al.*, *J. Contemp. Dent. Pract.*, **19**, 113 (2018), https://doi.org/10.5005/jp-journals-10024-2222

[6] S. Aalaei, R. Ganj-Khanloo and F. Gholami, *J. Dent.* (Tehran), **14**, 31 (2017), https://doi.org/10.5455/ijhrs.000000048

[7] U. Shafiq, S. Rahim, A. Saleem and M. Anwari, *Pak. Oral Dent. J.*, **36**, 495 (2016), http://doi.org/10.1016/S0022-3913(11)60108-X

[8] M. P. Walker, J. Burckhard, D. A. Mitts and K. B. Williams, *Angle Orthod.*, **80**, 1110 (2010), http://doi.org/10.2319/031510-150.1

[9] S. Gouveia, L. A. Otero, C. Fernández-Costas, D. Filgueira, Á. Sanromán *et al.*, *Polymers*, **10**, 642 (2018), https://doi.org/10.3390/polym10060642

[10] F. H. Isikgor and C. R. Becer, *Polym. Chem.*, **6**, 4497 (2015), http://doi.org/10.1039/C5PY00263J

[11] D. N. Barmana, M. A. Haquea, T. H. Kang, G. H. Kim, T. Y. Kim *et al.*, *Environ. Technol.*, **35**, 232 (2014), http://doi.org/10.1080/09593330.2013.824009

[12] M. P. Walker, J. Burckhard, D. A. Mitts and K. B. Williams, *Angle Orthod.*, **80**, 1110 (2010), http://doi.org/10.2319/031510-150.1

[13] M. P. Walker, J. Burckhard, D. A. Mitts and K. B. Williams, *Angle Orthod.*, **80**, 1110 (2010), http://doi.org/10.2319/031510-150.1

[14] A. Fayaz and A. Noori, *J. Dent. Sch.*, **34**, 28 (2016), https://doi.org/10.22037/jds.v34i1.24686

[15] J. Barr and R. J. Bowen, *J. Dent. Mater.*, **34**, 28 (2016), http://doi.org/10.14219/jada.archive.1968.0369

[16] Z. J. Zhang, H.-M. Li, S.-Q. Sun, L.-Q. Huang and I. Noda, *J. Anal. Lett.*, **46**, 2597 (2013), https://doi.org/10.1080/00032719.2013.805414

[17] Y. Liu, J. Li, G. Fan, S. Sun, Y. Zhang *et al.*, *Spectrochim. Acta A Mol. Biomol. Spectrosc.*, **1124**, 180 (2016), https://doi.org/10.1016/j.molstruc.2016.02.035

[18] K. Vernon-Parry, *Instrum. Viewpoint*, **13**, 40 (2000), http://doi.org/10.1016/S0961-1290(00)80006-X

[19] H. Orelma, A. Tanaka, M. Vuoriluoto, A. Khakalo and A. Korpela, *Wood Sci. Technol.*, **55**, 331 (2021), http://doi.org/10.1007/s00226-021-01265-x

[20] V. V. Nandini, K. V. Venkatesh and K. C. Nair, *J. Conserv. Dent.*, **11**, 37 (2008), http://doi.org/10.4103/0972-0707.43416

[21] H. S. Kim, C. G. Lee and E. Y. Lee, *Biotechnol. Bioprocess. Eng.*, **16**, 843 (2011), http://doi.org/10.1007/s12257-011-0352-8

[22] H. S. Kim, C. G. Lee and E. Y. Lee, *Eng. Biol.*, **16**, 843 (2011), http://doi.org/10.4103/1119-3077.197012

[23] A. R. B. de Olival, N. L. da Penha Júnior, J. V. F. Câmara, A. C. C. D. Simões, J. R. E. V. dos Santos *et al.*, *Sci. J. Dent.*, **6**, 37 (2018), http://doi.org/10.3390/dj6030037

[24] N. Aoyama, I. Hayakawa, N. Akiba and S. Minakuchi, *Prosthodont. Res. Pract.*, **6**, 239 (2007), http://doi.org/10.2186/prp.6.239

[25] M. M. Kulkarni and R. U. Thombare, *J. Clin. Diagn. Res.*, **9**, 98 (2015), http://doi.org/10.7860/JCDR/2015/13627.6407

[26] C. Halim, A. Cahyanto, S. Sriwidodo and Z. Harsatiningsih, in *Procs. AIP Conf. Proc.*, AIP Publishing, 2018, http://doi.org/10.1063/1.5021221

[27] B. Thalib, N. Hamrun, M. D. Utama, E. Machmud, S. Arpa *et al.*, *Mater. Today Proc.*, **3**, 100 (2020), http://doi.org/10.1016/j.mcpsp.2020.100105

[28] A. J. W. Phillips, "Phillips' Science of Dental Materials", Saunders, 2003, pp. 41–43, https://medarc.in/wp-content/uploads/2024/04/phillips-science-of-dental-materials.pdf

[29] J. E. Christensen, PhD Thesis, University of California, Los Angeles, 2020, http://doi.org/10.48550/arXiv.1907.13331

[30] D. J. Indrani and N. J. Matram, *J. Dent. Jakarta*, **46**, 5 (2013), http://doi.org/10.20473/j.djmkg.v46.i1.p5-8

[31] P. Bradna and D. Cerna, *J. Prosthet. Dent.*, **96**, 443 (2006), http://doi.org/10.1016/j.prosdent.2006.10.006

[32.] B. I. Cohen, M. K. Pagnillo, B. L. Musikant and A. S. Deutsch, *J. Prosthodont.*, **7**, 111 (1998), https://doi.org/10.1111/J.1532-849X.1998.TB00189.X

[33] R. M. Abdelraouf, R. E. Bayoumi and T. M. Hamdy, *Polymers*, **13**, 2923 (2021), https://doi.org/10.3390/polym13172923

[34] S. Anwar, S. Liaqat, R. Ullah, Z. Iqbal, F. Rahman *et al.*, *J. Appl. Polym. Sci.*, **29**, 602 (2024), http://doi.org/10.1080/1023666X.2024.2398152

[35] S. Sastrodihardjo and K. I. Harahap, *J. Mater. Kedokt.*, **7**, 33 (2018), http://doi.org/10.32793/jmkg.v7i2.371

[36] H. Malektaj, A. D. Drozdov, E. Fini and J. Christiansen, *Constr. Mater.*, **29**, 244 (2024), http://doi.org/10.3390/molecules29010244

[37] L. Singer and C. Bourauel, *Gels*, **9**, 429 (2023), http://doi.org/10.3390/gels9050429

[38] Z. Raszewski, A. Nowakowska-Toporowska, J. Weżgowiec and D. Nowakowska, *Dent. Med. Probl.*, **55**, 43 (2018), http://doi.org/10.17219/dmp/82179

[39] G. A. Zarb, R. Jacob and S. Eckert, in "Prosthodontic Treatment for Edentulous Patients", edited by Boucher C. O., Elsevier India, 2012

[40] K. Ginjupalli, R. Alla, T. Shaw, C. Tellapragada, L. K. Gupta *et al.*, *Mater. Today Proc.*, **5**, 16258 (2018), http://doi.org/10.1016/j.matpr.2018.05.117

[41] K. Ginjupalli, T. Shaw, C. Tellapragada, R. Alla, L. Gupta *et al.*, *Dent. Mater.*, **34**, 158 (2018), http://doi.org/10.1016/j.dental.2018.03.016