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Research Letter

Improving formability of palm leaf materials for foodware manufacturing using sodium hydroxide treatment

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Abstract

Deformation response of *areca catechu* palm leaf-sheath, after hydration and aqueous sodium hydroxide treatments, is studied using Limiting Dome Height testing. The leaf material can undergo large shape changes in biaxial stretching, with forming strains as high as 0.56 reached using optimized sodium hydroxide (NaOH) treatment (5% for 2 h). These limit strains are higher than for many sheet metals. The treatment also lowers the forming force, another key attribute of formability. Using parametric studies, optimal treatment conditions are established for producing high-aspect ratio products. Implications for single-step forming of palm-leaf materials and sustainable manufacturing of eco-friendly foodware are discussed.

Introduction

The deleterious effects of plastics on the environment are well-documented, with single-use plastics being a prime culprit.[1,2] Single-use plastics dominate the very large foodware/packaging industrial sector.[3] Most of the foodware products, post-usage, enter landfills or are dumped into the ocean, adversely impacting health and contaminating groundwater resources.[4] Furthermore, single-use plastics have seen a resurgence in the recent pandemic environment, driven by enhanced demand for plastic bags, take-out food packaging, bottled water, and PPE. Nevertheless, the journey towards a greener earth, even if temporarily set back, is inevitable. This journey has spawned heightened interest in finding environment-friendly materials and sustainable manufacturing process alternatives for foodware and related packaging production.

Recently,[5,6] we expounded on the direct forming of *areca catechu* palm leaf (sheath) materials into foodware (e.g., bowls, plates and cups) in a single-step from the raw plant material (Fig. 1). *Areca catechu* (or betel palm) is a species of palm that has been cultivated in India, South-East Asia, East Africa, Hawaii and Pacific Islands. Its occurrence in India has a recorded history of ~ 2000 years,[7] with areca foodware products reported even 200 years ago.[8,9] Our work showed that areca palm-leaf sheath could be stretch-formed to moderate tensile strains (~ 0.3), typical of those needed to manufacture foodware products.[5] These limit strains are similar to those of ductile aluminum and copper alloy sheet metal,[10,11] and are realized when the leaf-sheath is first hydrated (water-exposure) prior to the forming. The direct forming of foodware from leaf sheath has many advantages including potentially low embodied energy in products (= materials + processing energy), and processing of the leaf sheath in a single step of deformation without use of any intermediate processing steps (e.g., pre-treatment, pulping and drying) that are energy- and water- intensive. The latter steps are typical of (pulp-based) production of paper and bagasse foodware.

The palm-leaf deformation processing (forming) is very much analogous to forming of sheet metal by stretching or punching using dies [Fig. 2(a)]. In contrast to the pulp-based foodware production, the stretch-forming approach completely avoids use of filler materials or additives; in the former case, the filler/chemical content is often as high as 30%.[12] Figure 1(d) shows commercial palm-leaf foodware (e.g., plate, bowl, spoon) produced by deformation processing of areca sheath. The sheath material, see [Fig. 1(b)], biodegrades (composts) in ~ 60 days, compared to hundreds of years for plastics. Since the palm is grown primarily for its nuts, the sheath—which is shed by the tree at periodic intervals, is essentially "waste material" that is utilized for producing highly eco-friendly foodware. This use of waste "raw" material, coupled with the very small energy of the forming process (see ensuing), highlights another key advantage of the palm-leaf manufacturing—products with low embodied energy. The energy advantages over paper and bagasse products are further amplified if the "recycling service energy" contribution provided by the Earth, often taken as free, is included.[13] Lastly, unlike bamboo-based foodware, which usually involves cutting down of the tree for obtaining the raw material,[14] the areca leaf-sheath is a renewable resource that is harvested from the same tree over many seasons.

Palm-leaf foodware is commonly manufactured in the emerging and developing economies of Asia in small sheds, with

Figure 1. Overview of *areca catechu* palm tree, sheath and products. (a) Tree with various elements labeled. (b) Raw sheath of different sizes detached from the tree. The sheath length is the longitudinal direction (LD), width is the transverse direction (TD), and out-of-plane normal to the sheath surface is the depth/thickness direction (DD). (c) Image of the sheath across its width direction showing the thickness dimension (middle image), note that the sheath is thinner at the edges; and adaxial (top image) and abaxial (bottom image) surfaces with striations. (d) Commercial foodware products produced from the leaf-sheath, with adaxial surface forming the inner side of foodware.

low-cost press equipment (<10,000 dollars) and a workforce comprised mainly of women. With limited access to research and technology resources, the manufacturers have relied mainly on empiricism and intuitive understanding of material behavior for carrying out the product design and manufacturing. To address this deficiency, we initiated a program to understand the formability of these materials as well as the variables controlling areca foodware (product) life.[5,6] The formability studies showed that a) the classical Limiting Dome Height (LDH) test Fig. 2(a), widely used to assess formability of sheet metals in plane-stress loading,[15–18] is a viable approach also for characterizing the deformation processing behavior (and forming limits) of the palm-leaf materials; and b) the palm leaf materials have high workability—uniform elongation of up to 35% in tension, thickness reduction of ~70% in compression/rolling, failure strain of ~35% in LDH test—especially under suitable hydration conditions.[6] These measured workability limits are comparable to those of ductile metals like Al and Cu.[10,11] The LDH test also closely mimics the (punch) stretch-forming process used to manufacture areca foodware such as plates and bowls.

In the present study, we build on the early workability testing results and explore how the formability of the sheath can be significantly enhanced by treating the leaf-sheath with an aqueous solution of sodium hydroxide (caustic soda) prior to the forming (Caustic soda is both US FDA and EU approved for general use in food industry). Any enhancement of the formability beyond that achieved by pure hydration treatments would be of value not only for improving the current stretch-forming process for palm-leaf foodware, but also for expanding the capability to produce high-aspect ratio, palm-leaf products. The use of the NaOH as a formability enhancer is suggested by the proven value of this treatment to (a) soften wood and achieve increased bending, and (b) increase densification of natural wood by compression, for improved strength properties.[19,20] Since the action of the NaOH solution in all of these cases is to weaken

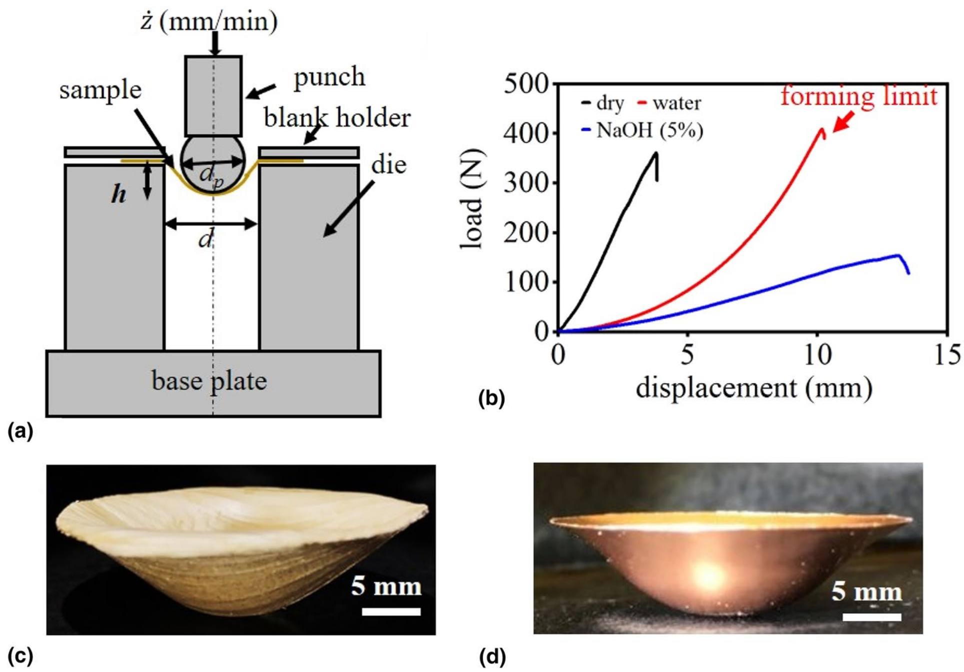

Figure 2. Limiting Dome Height (LDH) test. (a) Schematic of experimental setup with spherical punch for biaxial stretching. (b) Load–displacement curve for dry sheath, sheath hydrated with water for 2 h, and sheath treated with 5% NaOH aqueous solution for 2 h. Punch velocity ($\dot{z}$) is 5 mm/min. (c) Cup of depth ~ 10 mm formed from areca-sheath in LDH test with hydration treatment. (d) Cup of depth ~ 10 mm formed from copper sheet (thickness ~ 100 μm) in LDH test.

the bonding between cellulose fibers in natural wood, by partially removing hemi-cellulose and lignin, we hypothesized that there could also be an increase in the formability of the leaf-sheath due to this treatment.[21,22] Note that the leaf-sheath has a structure intermediate between that of wood and leaf.[23]

The results of the study, using LDH testing and structural characterization, show that the NaOH treatment does indeed produce a significant increase in the forming limit strain over and above that achieved by hydration treatments. Besides providing for improved understanding of the deformation behavior of leaf materials, the study results can be expected to be of value also for design of new palm-leaf foodware, and manufacturing (forming) process development and optimization.

Background: Areca palm and leaf-sheath

Areca catechu palm, popularly referred to as areca-nut palm or simply areca palm, is a medium-sized palm tree, that grows straight up to a height of ~ 20 m, and with a trunk that is 25–40 cm in diameter [Fig. 1(a)]. The palm is grown mainly for its nuts—areca or betel nuts. The leaves are 1–2 m long, pinnate, with numerous, crowded leaflets. The sheaths are sheet-like members, with physical attributes resembling a leaf-wood hybrid, that are connected to the central trunk of the tree [Fig. 1(b) and (c)]. They serve to protect inflorescence (flower arrangement) during the early stage of development, but are shed later. Each tree produces 10 to 15 leaf-sheaths every year. The sheaths used in foodware production [Fig. 1(b)] are usually mature, with lengths of 0.5 to 1 m, width < 0.5 m, and thickness < 4 mm. The density of the sheath material is 0.4 ± 0.1 g/cc. It is this sheath that is formed by a punching or stretch-forming process Fig. 2(a) into plates and bowls, very much analogous to forming of sheet metals.

The sheath has two distinct surfaces: adaxial surface that faces the stem, and abaxial surface that faces out (away from the stem), see [Fig. 1(c)]. The adaxial side (surface) forms the top (inner) side of a plate/bowl (foodware), while the abaxial side constitutes the bottom (outer) side of the plate/bowl [Fig. 1(d)]. The abaxial surface is much darker than the adaxial surface, besides being stiffer and stronger. Both surfaces have striations that run along the length of the sheath. These striations are the principal source of surface roughness, both in the sheath and in the formed product. The length direction of the striations is henceforth referred to as longitudinal direction (LD), while the direction perpendicular to the striations (width), in the plane of the sheath, is the transverse direction (TD). The out-of-plane normal to the sheath surface is parallel to the depth (thickness) direction (DD). In the raw sheath, the striations have peak-to-valley height of ~ 400 µm and lateral spacing of ~ 3 mm on the adaxial side; while on the abaxial side, the striations are much smaller in height (peak-to-valley ~ 50 µm) and more closely spaced (lateral spacing ~ 0.5 mm).[6] The sheath has a hierarchical microstructure that is intermediate between that of leaf and wood.[23]

Figure 1(d) shows some examples of commercially available food ware—plate, bowl and spoon. The product geometries highlight the large shape changes that the initially near-flat sheath material can undergo—illustrative of its high formability. In the commercial manufacturing process, the sheath is first hydrated for several hours (up to 12 h) before the forming, ostensibly to induce "ductile" behavior. Based on our discussions with manufacturers, the duration of this hydration treatment does not appear to be fixed or controlled. After the shape change is accomplished, heat is applied to the formed material for ~ 3 min by heating the forming die; this heating drives out the moisture and "locks" in the product shape. Commercial areca foodware products Fig. 1(d) typically have height (depth)-to-diameter (*h/d*) aspect ratios no greater than 0.2. There is broad interest from the manufacturers in increasing the sheath formability, for producing higher aspect-ratio products.

Experimental

A series of LDH experiments Fig. 2(a) was carried out with raw areca sheath under dry, hydrated and aqueous NaOH treatment conditions to assess sheath formability of relevance to foodware manufacturing. Raw areca sheath used in the experiments was obtained from a plantation in Tumkur, India, near Bangalore. The sheaths were collected during the summer of 2019. After collection, the sheaths were cleaned with dry compressed air and stored in plastic zipper bags to avoid contact with atmospheric moisture and prevent any fungus growth. The LDH test samples, of size 40 mm × 40 mm × 3 mm (thickness), were cut out from the central region of the sheath.

Limiting dome height (LDH) test

The Limiting Dome Height (LDH) test is one of the most widely used methods for characterizing formability of sheet metals.[16,17] The suitability of this test for studying the deformation processing characteristics of palm-leaf materials in stretch-forming has also been established recently.[5] For the testing, the 3 mm thick areca sheath sample, was clamped along its periphery in an LDH die and then axi-symetrically stretched (expanded) into cup (bowl) form, see [Fig. 2(a) and (c)]. The stretching was accomplished by pressing a hemispherical punch (diameter, $d_p = 25$ mm) against the unclamped central region (diameter, 30 mm) of the sample, as is typical of LDH testing. The sheath surface that forms the inner side of the cup is thus in contact with the punch, while the other side is free. A thin Teflon film (solid lubricant) was placed between the sheath and punch surfaces to reduce interface friction. An MTS tensile tester (maximum load—2 kN) was adapted for carrying out the test. The LDH test configuration in [Fig. 2(a)] closely resembles the sheath forming process for foodware, except that there is no bottom die in the LDH in contrast to the actual manufacturing process.[5] In sheet-metal forming, this type of loading is commonly referred to as biaxial stretching.

In a typical LDH test, the punch was moved downward at a speed of 5 mm/minute (displacement control mode) until the sample failed Fig. 2(b). The load was observed to increase with displacement over much of the test duration up and until onset of sample failure at a maximum penetration depth (*h*). This failure with the sheath material usually occurred at the pole of the sample, i.e., the deepest point in the cup. At the failure onset, a sharp load decrease was usually noted [Fig. 2(b)]. In contrast to the areca forming process, which uses heated dies to prevent sample spring-back and lock in the sample shape,[24] the punch in the LDH test was kept at room temperature. Spring-back of the sample was prevented by flowing hot air at 150°C for 3 min, after the maximum displacement was reached and just before releasing the load.

The sheath formability was assessed in terms of the maximum punch displacement and maximum (forming limit) strain developed at the sample pole (bottom-most point of cup), at failure; and forming force. Based on assumptions of zero friction at the interface between the punch and sheath, and isotropic material response, the maximum in-plane strain (ε) at the pole is estimated as $\varepsilon = \ln\left(1 + (2h/d)^2\right)$, using a solution for this loading case provided by Hill.[15,17] Here, *h* is the maximum punch penetration depth, and *d* is the diameter of the cup corresponding to its periphery (= die hole diameter) at this penetration depth. The solution applies for bulging of a diaphragm wherein the ratio of the diaphragm thickness to the cup diameter (*d*) is very small; bending and shear stresses can then be neglected (in our LDH test, this ratio is ~ 0.1). Both *d* and *h* were measured under load in the tests for each sample and used to estimate the pole strain. It is this pole strain (ε) that is reported as the forming limit strain for the sheath.

Parameter effects on formability

The principal parameter effect on formability that was studied was sheath-treatment type prior to the LDH (forming) test. The treatment parameters were dry (no treatment or as-received condition), hydration by sheath exposure to water for 2 h, and sheath exposure to aqueous NaOH solution (various concentrations and time durations). The NaOH concentrations used were 0% (pure hydration), 2%, 5%, 10%, and 15% by weight in water; and the time duration of exposure at each of these concentrations was 1, 2, 6, and 12 h. The forming limit strains were then obtained for each of these test parameter conditions, with five tests done at each treatment condition.

Characterization of sheath structure and treatments

The microstructure of the sheath before and after the treatments was characterized by optical microscopy, and high-resolution scanning electron microscopy (SEM, field emission Quanta 3D). Both the surface of the sheath and sheath cross-sections were examined to quantify microstructure changes. The microscopy was complemented by measurements of the mass of the samples, which provided estimates of any mass gain or loss arising from the treatment, and correlation with structural changes. Chemical analysis of sheath was done using Fourier Transform Infrared Spectroscopy (FTIR, Thermo-Nicolet Nexus 470 FTIR). This provided information about changes in the chemistry of the sheath due to the water and

NaOH treatments. FTIR analysis was performed on the dry and NaOH-treated sheath, using powder samples. The FTIR spectrum was obtained in absorbance mode in the 800–4500 cm^{-1} range (KBr beam splitter) by averaging over 36 repeated scans (resolution, 2 cm^{-1}), and analyzed using OMNIC software.

Results and discussion

Figure 2(c) shows a cup formed from the hydrated sheath (2 h. hydration) in the LDH test. The depth of the cup just prior to failure was ~ 10.2 mm giving an aspect ratio (h/d) of ~ 0.34. This cup is similar to smaller-scale bowls produced by the commercial sheath-forming process. However, its aspect ratio of 0.34 is much higher than that of the commercial products ($h/d < 0.2$). Dimension/shape measurements showed the LDH cup of [Fig. 2(c)] to closely resemble a hemispherical cap, except in the very vicinity of the clamped sample periphery. This resemblance occurred even though a lower die was not used to confine the deformation and constrain the shape change as in the commercial forming process.

Formability

The large beneficial effects of the hydration and 5% NaOH solution treatments on formability can be seen in the LDH load–displacement curves shown in [Fig. 2(b)]. Both treatments were applied for a duration of 2 h prior to the test; the selection of the 2-h duration was based on preliminary results with hydrated samples that showed a promising increase in formability[6] for this condition. The formability was assessed in terms of the capacity for shape change (limit strain, ε) and the forming force to produce a given shape change. The penetration depth at failure (h) for the dry, hydrated (2 h), and 5% NaOH treated (2 h) conditions, are seen to be 3.8 mm ($h/d = 0.13$), 10.2 mm ($h/d = 0.34$) and 13 mm ($h/d = 0.43$), respectively. The corresponding forming limit (pole) strains in the cup samples are 0.06, and 0.38 and 0.56. Based on both the penetration depth and forming limit strain, it is clear that the hydration and NaOH treatments result in a significant increase in the formability compared to the dry (non-hydrated) case. The aspect ratios achieved with the two treatments are 2.5X to 3.5X of that of the dry case, while the forming limit strains are 6X to 9X of that of the dry case. While both the hydration and NaOH treatments produce impressive increases in the forming limit strain, the NaOH forming limit of $\varepsilon = 0.56$ is approximately 50% greater than for the hydrated case ($\varepsilon = 0.38$). Similar large increase in the strain at failure due to hydration treatment has also been observed in uniaxial tensile testing.[6] Interestingly, the high limit strain of hydrated sheath in the LDH test is essentially the same as that of very ductile copper, compare for example [Fig. 2(c) and (d)].

Concurrent with the increased capacity for shape change, there is a corresponding large reduction in the forming force due to the hydration and NaOH treatments [Fig. 2(b)]. For example, the maximum load, which occurs almost at the failure limit (h ~ 3.8 mm), is ~ 350 N with the dry sample; whereas the corresponding load at the same penetration depth of 3.8 mm with the hydrated sample is < 50 N, an ~ 85% load decrease. This failure limit (maximum) load is even further drastically lowered with the NaOH treated samples relative to the hydrated samples—130 N vs 410 N—a nearly 70% reduction in the forming force with the NaOH treatment. The large forming force reduction is another manifestation of the significantly increased formability, due to the hydration and, secondly, even more so, due to the NaOH treatment.

Building on the promising formability results obtained with the 2-h treatments, a series of LDH tests was done with areca sheath samples subjected to NaOH treatments with varying concentrations and time duration exposures. The LDH formability results are summarized in Fig. 3 in terms of the forming limit strain. The main findings are as follows:

(a) The limit strain is seen to increase with NaOH concentration up to 5%, for the 2-h treatments. The highest formability (limit strain) across all the treatment combinations is seen to occur with the 5% NaOH treatment for 2 h. (Note that the highest formability with the hydration (water) treatment was also obtained with the 2-h exposure [Figs. 2(b) and 3 (0%)], though time-of-treatment effects are not significant here).

(b) For NaOH concentrations > 5%, there is a steep decrease in the formability. In fact, with the 10 and 15% NaOH (high) concentrations, the formability is low and similar to that of untreated (dry) sheath. A decrease in formability also occurs with the 5% NaOH when the time duration of the treatment is increased to 12 h. In contrast, with the pure hydration treatment (0% NaOH), the formability is not influenced by the duration of the time exposure.

(c) At the lower NaOH concentrations (≤2%), the time duration of the treatment was found to have negligible effect on the formability, analogous to the hydration treatment.

In summary, the 5% NaOH (2 h) treatment produces the largest increase in forming limit strain, being also much greater

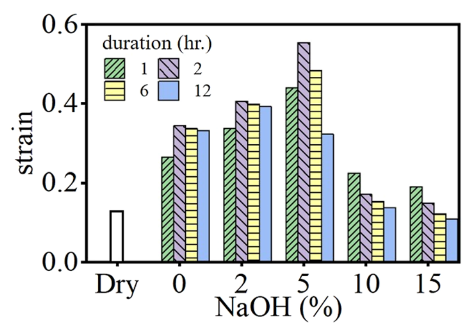

Figure 3. Effect of NaOH concentration and treatment time on forming strain. Dry refers to as-received sheath and 0% concentration to water hydration treatment (w/o NaOH). Five tests at each condition, error bar less than ± 10%.

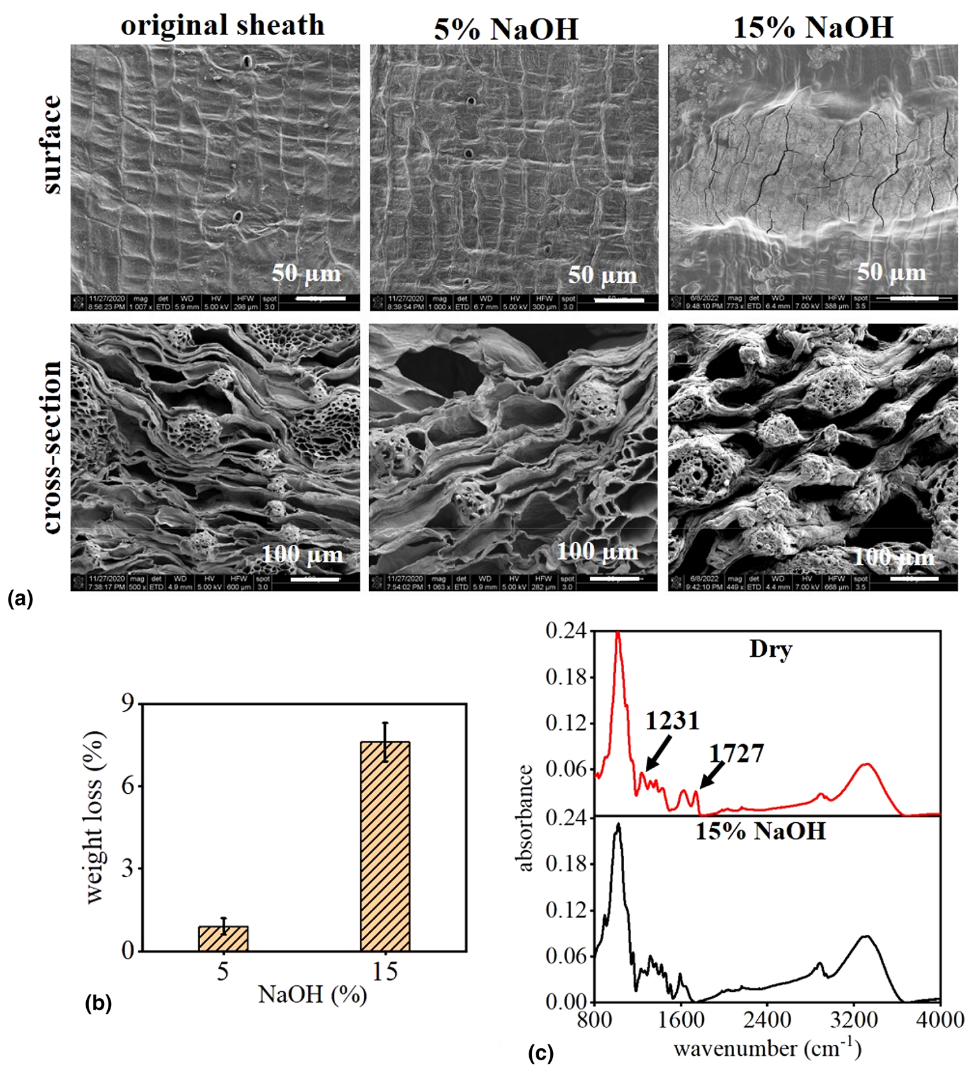

Figure 4. Effect of NaOH concentration on sheath structure. (a) SEM images showing sheath surface (top row) and transverse cross-section (bottom row) for three treatment conditions—original (dry); and 5% and 15% NaOH solution for 2 h. (b) Weight loss due to the NaOH treatments. (c) FTIR spectrum of sheath—untreated (dry) and after exposure to 15% NaOH for 2 h.

than that realized with a pure hydration treatment. While concentrations > 5% and/or treatment-time duration > 2 h. are found to be not beneficial for formability.

Structural changes and formability

SEM analysis of the sheath samples subjected to the 5% and 15% NaOH treatments (2-h exposure) have provided clues as to how these observed forming limit changes are a consequence of microstructure modifications resulting from the treatments. Figure 4(a) shows SEM images of surfaces (top row) and transverse (thickness × width) cross-sections (bottom row), respectively, of the original (initial dry) sheath, and sheath subjected to the 5% and 15% NaOH treatments. There is no discernible difference in the structure of the sheath surface or of the sheath cross-section between the dry and 5% NaOH treated sheaths. However, the surface of the 15% NaOH treated sheath shows significant swelling and cracking. Furthermore, the cell walls in this sheath appear to have disintegrated and dissolved, with significant structural damage to the material, see [Fig. 4(a) bottom row (right)].

Mass measurements and FTIR analysis on the untreated and treated samples confirmed and reinforced the SEM observations of the structural changes in the sheath samples. The mass measurements were made as follows, with 3 to 5 samples examined at each treatment (experiment) condition. In every experiment, the mass of the (initially) dry sheath sample was first measured. Each sample was then immersed in the 5% and 15% NaOH solution for 2 h, following which the sample was removed from the solution and washed several times with distilled water to remove all traces of the NaOH. The sample was then allowed to dry in the lab (ambient) environment for 48 h before being weighed again.

A loss of weight (mass) was seen to occur in the samples after the NaOH treatments. Figure 4(b) shows this weight loss plotted as a percentage change, with the initial mass of the sample serving as the datum. The weight loss is seen to be ~ 1% for the 5% NaOH treatment, and ~ 8% for the 15% NaOH treatment. The data, coupled with FTIR analysis of the samples (see discussion below) and observations reported in the literature with woods,[21,25,26] show that more mass in the form of hemicellulose and lignin is removed when the sheath is treated with the 15% NaOH solution. This larger-scale removal of hemicellulose and lignin with the 15% solution damages the material structurally, in addition to weakening the bonds between the cellulose fibers. In contrast, the 5% NaOH treatment appears to only weaken the bond between the cellulose fibers, while removing only smaller (negligible) amounts of hemicellulose and lignin.[21,27,28] This explains why the forming limit strain for the sheath samples increases with %NaOH at the lower concentrations (5% and below), wherein there is only intra-cellulose bond weakening with negligible structural damage [Fig. 4(a), middle column]. Whereas, the formability is lowered rapidly beyond the 5% threshold concentration, due to extensive structural damage to the material (e.g., [Fig. 4(a), right-most column images]). The optimum NaOH treatment is thus one that just causes a weakening of the bonds between the cellulose fibers to enhance material deformation capacity, but without inducing large-scale structural damage that can promote fracture.

The FTIR spectra of [Fig. 4(c)] confirm the large-scale removal of hemicellulose and lignin due to the 15% NaOH treatment. The spectrum peaks clustered around 1231 cm^{-1} and 1727 cm^{-1} from the dry untreated sheath [Fig. 4(c), top] are typical of plant material with cellulose, hemicellulose and lignin as the main constituents.[25,26] Both of these peaks are seen to disappear following the 15% NaOH (2-h) treatment [Fig. 4(c), bottom]. The 1231 cm^{-1} peak corresponds to –CO (bond) stretching in lignin, while the 1727 cm^{-1} peak is due to the stretching of –C=O in ester linkages of carboxyl group of lignin and hemicellulose.[25,26] Hence, the disappearance of these peaks indicates structural degradation with partial removal of the lignin and hemicellulose, and the observed larger weight-loss for this treatment.

Embodied energy

The LDH force–displacement curves can also be used to estimate the specific energy for the forming process, an important process performance as well as product sustainability index. The specific energy is the energy required to form unit mass (or volume) of the material. It can be estimated as the area under the load–displacement curve of [Fig. 2(b)] divided by the mass (or volume) of the sheath material deformed in the test. This calculation gives for the process specific energy, the following values 0.89×10^{-3} MJ/kg (dry), 2.5×10^{-3} MJ/kg (hydrated) and 1.18×10^{-3} MJ/kg (5% NaOH, 2 h) (Note: The specific energy for the dry sheath is smaller than for the hydrated and NaOH cases because this sheath fractures at a small strain).[6] These specific energy values are very small, ~2 orders smaller than for forming of metals.[18] Given the very low process specific energy, and coupled with the fact that the sheath is essentially waste material discarded by the tree, we estimate the embodied energy of area palm-leaf foodware (=material+process energy) as near-zero (at least 5 to 6 orders smaller than for equivalent plastic/paper products.[6,29] This very low embodied energy is another attractive feature of the palm-leaf products, and of their production by a direct single-step forming process from raw leaf-sheath.

Implications and challenges ahead

Our LDH testing has shown that the formability of areca palm-leaf material is significantly enhanced by use of aqueous NaOH treatments (≤5%) of suitable time duration. The highest increase in formability is obtained with a 5% NaOH treatment, with treatment duration of 2 h. This increase in formability with the 5% NaOH is characterized by an (a) increase in the forming limit strain by as much as 9X for over that of the dry sheath, and 50% over that of the hydrated sheath; and (b) approximately 70% reduction in the forming force compared to the hydrated sheath. The 2 h, 5% NaOH treatment can thus be taken as optimum for the sheath forming process. The formability in terms of the limit strain is found to decrease sharply for NaOH concentration > 5%. The optimized (maximum) forming limit strain is equal to or greater than that of copper in the LDH test. The action of the NaOH in improving the formability likely occurs by its action in weakening the inter-cellulose bonds. The optimum concentration and time duration of the treatment are determined by a competition between this bond weakening which enhances deformation capacity, and structural damage to the material due to leaching out of the hemi-cellulose and lignin that increases the propensity for material fracture.[21] The proposed sodium hydroxide (caustic soda) treatments can be used in foodware manufacturing practice, as caustic soda is classified as food-grade and designated as safe for general use in food by the FDA. It has also been widely used in food processing and as a pH adjusting agent in drinking water.

While we did not explore, herein, with the NaOH treatment, the formability differences arising from punch loading on the adaxial surface (present study) versus the abaxial surface, we do expect a difference in the forming limit strain based on prior studies with water-treated (hydrated) sheath.[5] The earlier work showed that the formability (limit strain) is about 25% higher with the abaxial surface loading. We expect a similar difference also with the NaOH treatment. But for reasons most likely pertaining to aesthetic features, structural integrity, and surface diffusion characteristics, areca foodware manufacturers appear to have settled for the less-formable, adaxial loading configuration in practice.

Our findings are also consistent with known facts about how NaOH treatments can soften wood, so as to increase its "bend formability" as well as its densification by compression.[19,20] In fact, it was these wood-softening effects of the NaOH that led to the hypothesis underlying the present study—that NaOH treatments can improve formability of the leaf-sheath in stretch-forming processes. Future work will further explore and verify this hypothesis, that is rooted in the findings reported herein. The observations and proposed hypothesis should also aid in identifying other types of formability-enhancing treatments for palm-leaf materials and woods, as well as for plant species beyond palms.

From a manufacturing standpoint, for palm-leaf foodware products to become competitive and potentially displace plastic and other paper/bagasse based foodware, it is critically necessary to scale-up the forming process in multi-dimensional ways—increased production rate via process optimization and automation; capability to produce a range of products, especially higher-aspect ratio foodware; and achieve at least 10× reduction in manufacturing cost at scale, to name just the main ones. It is also important that the raw material be abundantly available. Our findings pertaining to formability enhancement of leaf materials by suitable aqueous treatments are an important step in this scale-up journey. Firstly, they suggest opportunities for improving the range of foodware products that can be produced from leaf materials, especially high aspect ratio products like cups, bowls, tumblers and shell-type packaging. Secondly, they point to possibilities for increasing foodware production rates and reducing manufacturing cost by manufacturing process optimization. Thirdly, by establishing the LDH test, originally developed for sheet-metal formability (forming limit diagrams), as a viable technique also for characterizing formability of plant materials, and forming parameters and pre-treatments, a path forward for rapidly screening plant materials (beyond areca palm) for foodware applications is potentially available. This screening could also aid in addressing the raw material supply challenge. Lastly, the near-zero embodied energy of palm-leaf products produced by the single-step, direct forming approach, and the very short duration (~60 days) in which they bio-degrade, make them attractive candidates for sustainable manufacturing of new classes of eco-friendly foodware.

Concluding remarks

The deformation response of *areca catechu* palm leaf-sheath has been characterized using the Limiting Dome Height (LDH) test, for hydration and aqueous NaOH treatments. The leaf material is found to have high formability, with limit strains as high as 0.56 being achieved with NaOH treatment. These strains are comparable to or exceed those of ductile sheet metals. The NaOH and hydration treatments also greatly reduce the forming force, another key attribute of material formability. The largest formability benefits are realized with an optimized treatment, 5% NaOH (by weight) concentration in water and a 2-h exposure. The beneficial effects on formability of the optimized NaOH treatment arise by just-sufficient weakening of the inter-cellulose bonds in the leaf. However, aqueous treatments with NaOH concentrations > 5% and/or time exposures > 2 h adversely affect formability, via structural damage to the material resulting from removal of hemi-cellulose and lignin. The LDH formability data have also enabled estimation of the forming specific energy. Using this estimate, we have shown that the embodied energy of the palm-leaf products is very small, nearly zero.

The results suggest approaches for increasing foodware production rates, and diversifying the range of product shapes (e.g., high-aspect ratio structures) that can be formed from the palm-leaf materials. They also highlight the exciting potential and opportunity for producing eco-friendly foodware from plant leaf materials, using single-step deformation processing routes.

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Data availability

The data that support the findings of this study are available from the corresponding author upon reasonable request.

Declarations

Conflict of interest

The authors declare that they have no conflict of interest.

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