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Tensile Behavior and Diffusion of Moisture through Flax Fibers by Desorption Method

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Abstract: There has been a substantial increase in the usage of natural fibers and biodegradable polymers in composite materials due to the recent focus on sustainability of materials. Flax fibers have exhibited higher mechanical properties compared to most other natural fibers available. However, one of the major challenges faced in the use of flax fiber is its hydrophilicity. In this study, the tensile behavior of flax fiber tows removed from commercially available woven fabrics were investigated at different moisture levels. The breaking tenacity of fiber tows was shown to increase with an increase in moisture content of up to 25%. After this point, additional absorption of moisture resulted in a decrease of fiber tenacity. In addition, the diffusion process through flax fiber mat with different areal densities was investigated and the diffusion coefficients were determined using the desorption curves. Diffusion rates were not found to significantly change with varying areal densities of 200 to 400 gsm, but were significantly different when exposed to temperatures of 55 °C versus 80 °C.

Keywords: natural fibers; flax fiber tows; tenacity; diffusion; moisture desorption; diffusion coefficient

1. Introduction

Natural fibers are obtained from plants, animals and some by geological processes (alteration and metamorphism of basic igneous rocks rich in magnesium silicates [1,2]. Natural fibers are increasing in utilization compared to engineered materials because of their biodegradability, sustainability, and lower costs [3,4]. In addition, other benefits include reduction in CO_2 emission, less dependency on foreign oil resources, reduction in energy consumption and the most important one, recyclability [5]. There have been numerous studies on natural fiber technology as well as their use as reinforcement in polymer composites and improved sustainability over traditional composite reinforcements [6–16].

Flax is as a type of crop fiber which is grown both for fiber (linen) and for seed oil (linseed) depending on the variety. Due to properties such as lower thickness, better thermal insulation and diminished skin irritation, since ancient times, it has been used as a choice for linen [17]. Nowadays, flax fiber is finding more ways to be utilized in variety of industries including structural (aerospace) [18], sports [19,20], automotive [21] and medical applications [22].

There are more than 180 species of flax [23] growing in regions such as India, Bangladesh, United States of America, Canada, China, Ethiopia, Russia, Ukraine, and France [24]. Properties of characteristic filaments depend on the variety of the plant, territory in which it is developed, the age of the plant, harvesting methods, and the extraction strategy utilized [25,26].

The mechanical behavior of flax, as a type of multicellular fiber, is defined by the physical, mechanical, and chemical properties of the morphological constituents such as cellulose, hemicellulose, lignin, and pectin [27,28]. The flax strands can be up to 1 m long and made of filament bundles with lengths ranging between 2 to 5 cm and widths fluctuating between 10 and 25 µm. They are adhered together by a gelatin interface which is a polyhedron that promotes better packing efficiency [25]. Figure 1 is a micrograph of a bundle of flax fibers that was captured using a JEOL JSM-6490LV Scanning

Electron Microscopy (SEM) (JOEL Solutions for Innovation, Peabody, MA, USA) at North Dakota State University, where the non-uniform polygonal cross sections of the fibers are shown. A characteristic fiber might be additionally characterized as an agglomeration of cells in which the width is immaterial in examination compared to its length. In certain applications, regular filaments are supplanting fiberglass strands in fortified polymers, where the rigidity of the fiber is not as imperative as the solidness [11].

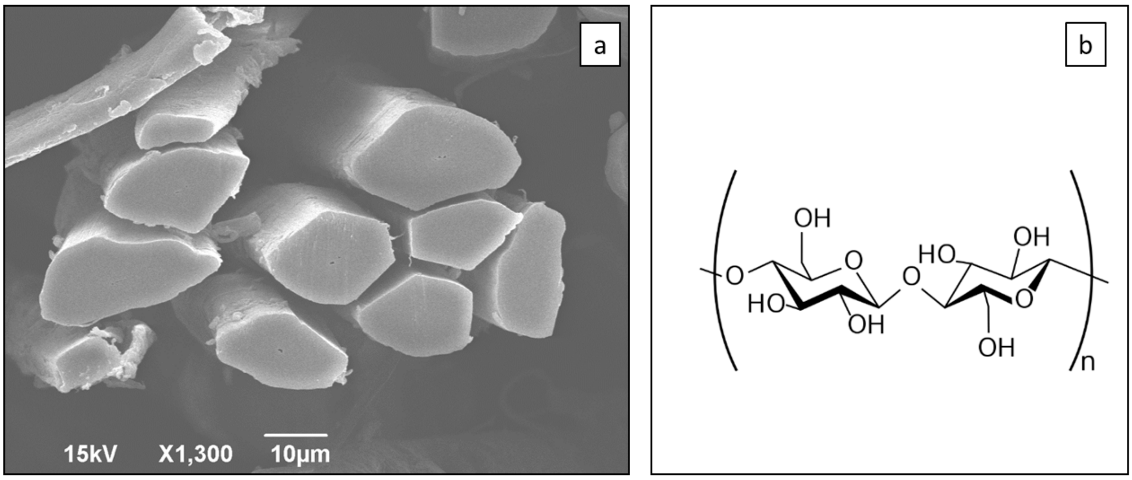

Figure 1. (**a**) Scanning Electron Microscopic image of cross section of bundle of flax fiber, (**b**) repeating unit of cellulose.

In order to understand the effect of moisture on the mechanical performance of flax fiber or polymer matrix reinforced with flax fiber, it is important to understand the overall structure of flax fibers. Flax fibers consist of 60%–71% cellulose contained in two main layers of fiber [29,30]. The first layer dispositioned during plant growth, the primary wall, contains both cellulose and hemicellulose [31–34]. The secondary wall contains mainly cellulose and includes three sub-layers consisting of helically wound highly crystalline cellulose chains called micro-fibrils [35]. The secondary wall contributes to up to 70% of the fiber's young's modulus. Therefore, higher cellulose content will result in higher tensile modulus [36]. This molecular structure of cellulose dictates its chemical and physical properties. The cellulose content and its orientation within the flax fiber will define the properties of fiber and resultant composites [37]. Also, the presence of three hydroxyl groups in each repeating unit, will make cellulose a hydrophilic molecule [36,38]. A repeating unit of a cellulose molecule is depicted in Figure 1.

The hydrophilic nature of the flax fibers is the most observed drawback in the usage of these fibers in structural applications. Natural fibers in general, and flax fiber in particular, are moisture reliant as their mechanical properties are progressively influenced by smaller changes in water content which reacts as a plasticizer. Furthermore, the hydrophilic nature of flax fiber will lead to absorption of moisture and the result would be presence of voids at the interface of fiber and matrix and this would thus affect the quality of ensuing composites. Therefore, the study of the diffusion of moisture through flax fibers is one of the key issues which must be addressed for the utilization of flax fiber over glass fiber in a composite material [39,40]. Having a low amount of moisture present in natural fibers will result in higher quality composites with increased mechanical properties as high as 25% [39].

There have been endeavors for single fiber tensile test trials of flax strands which have had the capacity to fit the Weibull appropriation for their quality [41–45]. Flax tows are available for composite application as spools or in a woven architecture [46]. It has been demonstrated that complex shapes can be accomplished with flax tow based woven textiles by sheet framing processes. However, the challenge remains that there has been no standard by ASTM for the tensile test of bast fiber tows. The most relevant standard is ASTM D3822/D3822M (Standard Test Method for Single Textile Fibers).

The scope of this standard is applicable to continuous (filament) and discontinuous (staple) fibers or filaments taken from yarns or tow. Hence, a test method specific to testing tows must be developed independently to characterize the mechanical properties of bast fiber tows.

Moisture desorption and sorption by flax fiber mats as a source of reinforcement in composites and the rate that water diffuses through the mats could affect the mechanical performance of the ensuing composites [40]. There is a requirement to find a drying method of the fiber mat which is being used prior to the reinforcement to reduce the moisture content in the mat. However, the lack of enough supporting data to draw the correlation between different variables such as temperature, areal density and moisture desorption rate in flax fiber is a huge hindrance in achieving this goal.

The desorption of water through different flax fiber mats, and the effect of temperature on desorption also needs research to ensure better mechanical performance in fiber reinforced composites as well as a consistent manufacturing quality of fiber-reinforced composites. In this study, the diffusion behavior of water in flax fiber mats and using the moisture retention as an advantage is studied. In addition, tensile tests are performed on flax fiber tows in wet and dry conditions to provide additional data to understand the affecting mechanism of moisture absorption by flax fiber as well as support development of an independent future ASTM tensile test procedure for natural fiber tows.

2. Materials and Methods

2.1. Moisture Diffusion Testing

The moisture absorption behavior of plant-based and textile fibers in general and flax fiber in specific has been defined by previously developed models [47–49]. The moisture diffusion in and out of natural fiber mats can be determined using second Fick's law and the behavior is referred to as Fickian behavior [50].

Untreated non-woven flax fiber mats with three different areal densities of 228 gsm, 300 gsm, and 400 gsm were chosen to study the process of desorption for the maximum removal of moisture. The experiments followed the guidelines of standard SR ISO 6741-1/1998. At a minimum, three samples of each areal density were cut to the size of 50.8 mm × 25.4 mm × 6.3 mm as shown in Figure 2 and were immersed in distilled water for 2 h to permit the continuation of water sorption until saturation limit was reached.

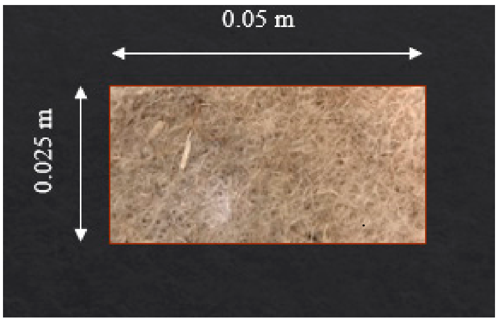

Figure 2. Non-woven flax fiber mat cut to size before the immersion into distilled water.

The initial moisture content of the samples was measured using an AZI moisture analyzer (Computrac®® MAX®® 4000XL, Arizona Instrument, Arizona, US). The initial mass of fiber mats was recorded and fibers were placed in a convection oven (Model 1370FM, VWR Co., Randorm, PA, USA) at 55 °C and 80 °C. At intervals of 10 min, the mass of the fiber mats was recorded and experiments were carried out until three consecutive measurements were recorded to be the same within less than

2%. Each experiment was repeated three times to ensure reproducibility of results. The amount of the water content, *M(t)*, at instantaneous time was determined using the following equation [39].

$$M(t) = \frac{M_t}{M_0} \tag{1}$$

where M_t is the moisture content at time t, and M_0 is initial moisture content the fiber. The desorption diffusion co-efficient, D, (i.e., the rate at which moisture diffuses through the mat) was calculated using the following equation [51]:

$$\frac{M_2 - M_1}{\sqrt{t_2} - \sqrt{t_1}} = \frac{4M_m\sqrt{D}}{h\sqrt{\pi}} \tag{2}$$

where M_m is the maximum moisture content of the mat at an instant, h is the thickness of the sample, and M_2, M_1 are moisture content at times t_2, t_1, respectively.

2.2. Tow Tensile Testing

A woven flax fiber mat with plain weave was used to carefully extract fiber tows for tensile testing. The woven mat procured from Composites Evolution, Chesterfield, UK (Biotex flax) has the same number of threads in both warp and weft directions and all tests specimens were pulled out from warp direction of the fabric. The plain-woven mat as well as pulled-out fiber tows are shown in Figure 3.

(**a**) (**b**)

Figure 3. (**a**) Woven mat of the flax fibers (plain weave), (**b**) flax fiber tow pulled out of the weave.

An Instron 5567 universal tensile testing machine equipped with filament grips was used to perform tensile tests according to ASTM D3822 at room temperature and 65% relative humidity. A minimum of five tow samples were made of multiple strands whose ends were cast with epoxy resin for better clamping in filament grips for each test. The set-up is shown in Figure 4. A crosshead speed of 0.01 mm/min was set for all tests, and tests were carried out at different gauge lengths of 153 mm, 175 mm, 200 mm, 220 mm and 242 mm. Each test was done at 5%, 25%, 60% and 80% moisture content.

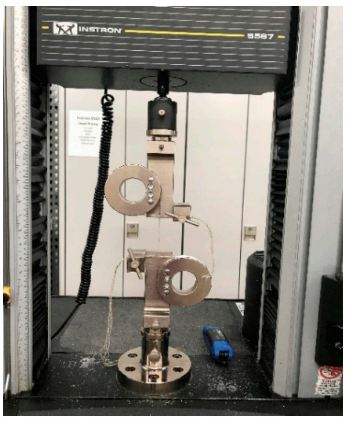

Figure 4. Flax fiber tows in filament grips of the Instron 5567 universal testing machine.

3. Results

3.1. Moisture Desorption in Flax Fiber Non-Woven Random Mats

The change in moisture content was plotted against square root of time and results are shown in Figure 5. Statistical analysis was performed using ANOVA with 95% confidence interval to examine the significance of the results. The *p*-value of the results was compared to the significance level to assess the null hypothesis. As expected, temperature had a significant effect on moisture desorption. The least significant mean for 55 °C was significantly different from least significant mean for 80 °C. On the other hand, there was no significant difference found between any two areal densities of flax fiber mats tested. The variation of the means of diffusion coefficient for 55 °C was in the range $1.98 \times 10^{-8} \pm 4.75 \times 10^{-9}$ m^2/s. This variation was very similar in the range of diffusion of water through the different areal density flax fiber mats tested. The variation of the means for 80 °C was in the range $4.05 \times 10^{-8} \pm 8.67 \times 10^{-9}$ m^2/s. Similarly, the variation of the means for the different areal density flax fiber mats tested was not significantly different, implying that that fiber concentration was not affecting the moisture diffusivity in a significant way.

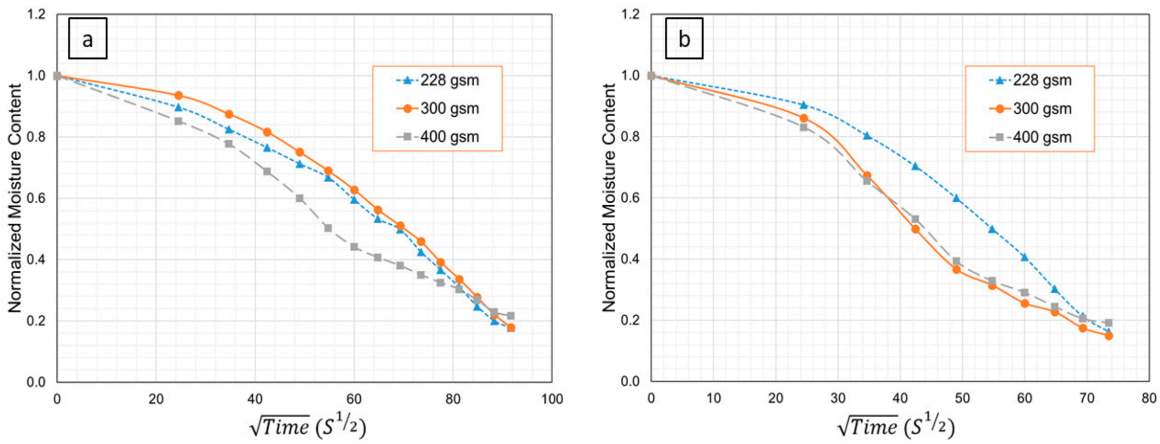

Figure 5. Desorption curves for moisture through non-woven random flax fiber mats for different areal densities: (**a**) at 55°C, (**b**) at 80 °C.

3.2. Influence of Moisture on Tensile Properties of Flax Fiber Tows

A typical load-displacement curve for the flax fiber tows tested is presented in Figure 6. The tensile properties were found to be independent of the gauge length for the range investigated in this study. The first non-linear region (at small cross-head displacement) corresponds to the stage where the fibers or fiber bundles within the tow arrange themselves (i.e., align) with the loading axis during the tensile loading. The maximum tensile force was recorded as the peak point of the curve according to ASTM D2256 standard [25]. The maximum force is divided by the linear density of the fiber bundle to calculate the maximum breaking tenacity (in centinewton per tex, cN/tex) [52]. In addition, the slope of the linear elastic portion of the load/displacement curve was used to calculate the stiffness (N/mm) of each fiber bundle.

Statistical analysis of the tensile results using one-way ANOVA method proves the significant variation between different moisture levels while there was no statistically significant effect observed from variation in gage lengths. Table 1 presents the average results of the tensile tests at different moisture content of flax fiber tow. Distribution of breaking tenacity and stiffness of the samples are presented in Figure 7.

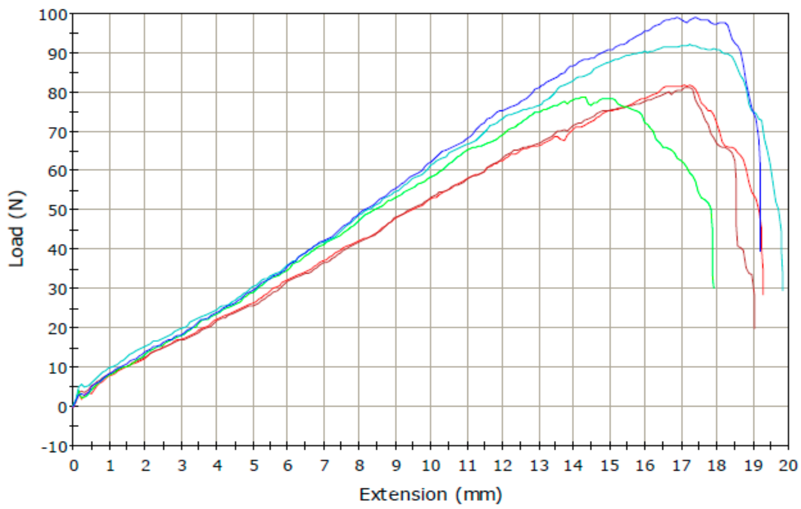

Figure 6. Typical load-displacement curve for flax fiber tow.

Table 1. Average results of tensile test on flax fiber tows with different moisture contents.

Moisture (%)	Tenacity (cN/tex)	Stiffness (N/mm)	Elongation at Failure (%)
5	7.18 ± 0.94	4352.80 ± 495.69	12.78 ± 0.98
25	9.83 ± 0.21	3342.28 ± 208.27	22.74 ± 3.67
60	4.13 ± 0.29	3141.40 ± 100.95	21.49 ± 3.23
80	3.06 ± 0.07	2700.88 ± 135.42	28.27 ± 0.34

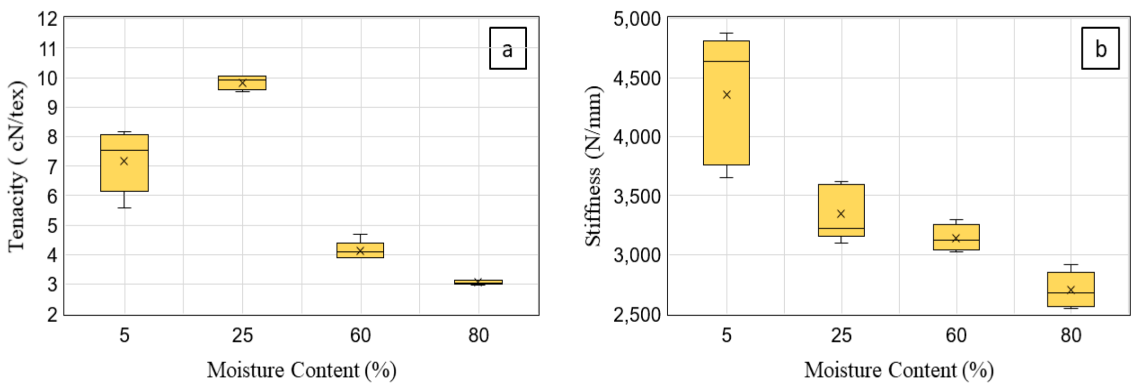

Figure 7. (**a**) Distribution of tenacity for various tow moisture levels, (**b**) distribution of stiffness for various tow moisture levels.

4. Discussion

4.1. Moisture Desorption in Flax Fiber Non-Woven Random Mats

Based on presented results of moisture desorption, the diffusion process follows Fickian behavior [45] in the linear region of the plot which allows the calculation of the diffusion co-efficient. According to second Fick's Law, the rate of the mass of water that passes through a cross-section unit of fiber is proportional to the concentration gradient of moisture (dM/dx) by the proportionality constant, i.e., diffusivity coefficient, D (cm^2/s). Diffusivity is slow at lower moisture contents and increases as the moisture content increases.

According to studies done by Stamboulis et al. [47,50] and evidence provided by Feng et al. [53], moisture diffusion in flax fiber occurs by three mechanisms and interactions; first, diffusion through the air to the fiber bundle surface; second, diffusion of moisture in between fiber bundles and to the single fiber surface; and third, diffusion of water from surface of single fiber to the inner structure and absorption by cellulose chains both by penetrating the space between micro-fibrils and through chemical bond with hydroxyl group [50]. Since there is no chemical reaction happening in the first two stages, it is assumed the moisture can leave the space between fiber bundle and surface of the fiber by the drying process (increasing temperature). Validity of this assumption is demonstrated in a study conducted by Feng et al. [53].

The initial moisture content at 55 °C was 5.23% and the final moisture content was 1.46% and for 80 °C the initial moisture content was 6.12% and the final moisture content was 1.12%. The initial moisture content varied between 4% and 6% and the final moisture content varies between 1% and 2.5% for all specimens. This shows that there is some moisture remaining in the mats after drying that cannot be removed very easily. This is the water content that is held by fiber due to presence of swelling stresses [49] as well as the water held by strong bonds with the cellulose chain. The maximum moisture retention for the flax mat was found to range from about 14% to 19%.

4.2. Influence of Moisture on Tensile Properties of Flax Fiber Tows

Increase in moisture content of flax fiber tows increased the breaking tenacity to a certain extent, passed which tenacity greatly dropped. Similar behavior was observed by Warner [54] where increases in moisture content, resulted in plasticizing effect and as a result of this effect he observed an increase in strength of the tow (i.e., tenacity). However, similar to the current results of this study, strength increases up to certain moisture content, in this case, 25%. This is attributed to the mechanism of moisture absorption and plasticizing effect of moisture and presence of water in between fibrillary chains of cellulose [54]. The addition of moisture to the fiber after this point will result in increases in bonding water and therefore reduce the plasticizing effect and consequently reduction in tenacity [47,50].

On the other hand, examining the stiffness of the flax fiber tows, however, reveals that there is an opposing effect of decreasing rigidity with increase in moisture content. A similar trend in decrease of fiber stiffness by increase in moisture was observed and discussed by Stamboulis et al. [50]. As previously mentioned, flax fiber exhibits its rigidity from rigidity of cellulose chains and micro-fibrils in the secondary layer of the fiber. Closely packed cellulose chains as well as the micro-fibrilar angle is of the most important factors affecting rigidity of the fiber. Water absorption affects this in two ways; first, water molecules force themselves between micro-fibril chains and push the chains apart from each other; second, the mass of cellulose is affected, and they become prone to changing shape by application of loading [48]. In addition, as suggested by Stamboulis et al. [50], swelling occurs in an anisotropic fashion, meaning that swelling in the fiber due to moisture is larger in the direction perpendicular to the cellulose chains compared to the direction parallel to the fiber axis. This could affect the micro-fibrilar angle and consequently, the rigidity of the fiber.

5. Conclusions

This study has produced physical and mechanical results explaining how flax fiber tows behave when they undergo tensile loading and how the flax fiber mats allow moisture to diffuse through them. The results obtained in this study are relative to the specific areal densities for flax fiber mat and specific moisture levels and gauge lengths for tensile testing only. Varying results might be obtained for other areal densities besides those considered for this study for the diffusion experiment as well as for different gauge lengths for tensile experiment results considered.

The desorption of moisture in the flax fiber mats studied were shown to have significantly different diffusion coefficients at 55 °C as compared to the mats tested at 80 °C However, the diffusion coefficients calculated were independent of areal density. The tensile testing of flax fiber tows resulted in values which are stable for their use in structural applications of high-performance composites. The maximum tensile breaking tenacity of the flax fiber tows increased with moisture content up to 25% but then was found to decrease under very wet conditions (i.e., 80%). Stiffness of the fiber tows were found to decrease while elongation at break where found to increase with increasing moisture content.

Future work in this area should be focused on better understanding the influence of flax fiber moisture content on composite properties as well as the diffusion of moisture into a composite composed of flax fiber reinforcement. In addition, more research into the influence of strain-rate, gauge length, and fiber bundle or tow size needs to be tested in order to help continue to establish an ASTM test standard for tensile testing bast fibers.

Author Contributions: S.S.R. prepared the specimens, performed the experiments, analyzed the results, and wrote a majority of the paper. A.A. analyzed the results, provided discussion and elaboration of results, wrote and edited portions of the paper. C.A.U. conceived the research idea and experiment design, analyzed the results, provided discussion and elaboration of results, wrote and edited portions of the paper.

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