

# Evaluation of an ammonia-free natural rubber latex adhesive

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## Abstract

Natural rubber latex (NRL) within the adhesives industry has had limited applications because of poor water and chemical resistance; additionally, conventional preservation methods with ammonia produce poor adhesive properties and introduce toxicity. Today, solvent-based adhesives dominate the market because of their superior mechanical properties; however, they negatively affect the health of users depending on the application due to the toxic chemicals. In this study, we investigated using a novel NRL-based ammonia-free adhesive as primary materials with bio-additives, specifically cellulose, and collagen, to enhance their mechanical property provide the industry with an adhesive safe to handle. The peeling strength of an ammonia-free NRL, 60% dry rubber content (DRC) NRL, with cellulose and collagen (Industrial Adhesive) were compared to high-performance adhesives used within the footwear and construction industry to prove these adhesives direct applicability within these industries. Our findings showed that the ammonia-free NRL Industrial Adhesive exhibited superior mechanical properties when compared against resilient flooring adhesives and footwear adhesives. Additionally, the peeling strength of an ammonia-free 35% DRC with only cellulose (Office Adhesive) was compared to PVA-based Elmer's Glue and it was observed that the ammonia-free NRL adhesive outperformed Elmer's Glue by almost reaching four times the peeling strength of Elmer's Glue.

## KEYWORDS

ammonia-free, bio adhesive, natural rubber latex, sustainability

## 1 | INTRODUCTION

Among the numerous classes of adhesives, natural rubber adhesives have been in use for almost 200 years.<sup>1</sup> Natural rubber is sourced from the sap of rubber trees that grow primarily in tropical regions. This sap, known

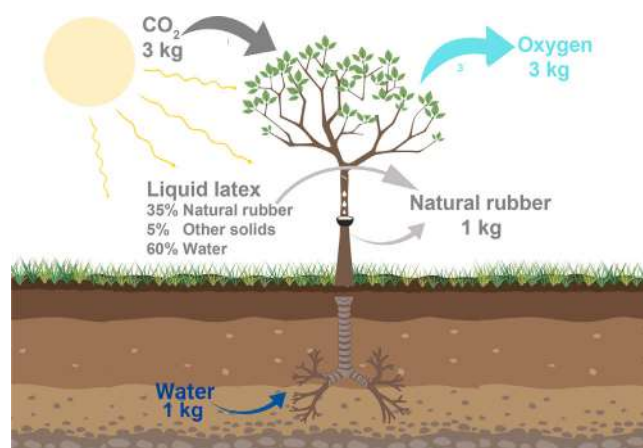
as natural rubber latex (NRL), is collected by making small incisions on the tree's bark through tapping. The sap is then collected in small cups attached to the tree and processed to create a wide variety of natural rubber products.<sup>2</sup> NRL can rapidly decompose through the balance of acidification and alkalization reactions caused by

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the bacteria and other microorganisms.<sup>3</sup> Therefore, before processing NRL, ammonia is traditionally used to inhibit coagulation<sup>4</sup> and as a biocide to protect and hinder decomposition.<sup>5–7</sup> In addition to ammonia, existing natural rubber latex adhesives contain other chemicals, such as tetramethylthiuram disulfide (TMTD) and zinc oxide (ZnO) to help stabilize and preserve the liquid natural rubber latex. These chemicals harm tree tappers and workers handling natural rubber.<sup>8–10</sup> As stated, the onset of putrefaction triggers an unwanted coagulation process, which renders the latex malodorous, challenging to handle and process. Hence, the natural rubber producer is forced to use chemicals that, once added, render the NRL stable and in a processable condition.

Of the above-mentioned chemicals, ammonia possesses a grave threat to living organisms due to its highly hazardous and toxic nature, whereas recent studies have shown that NRL membranes processed with ammonia may exhibit cytotoxic and genotoxic effects in cultures, and increased inflammation in implanted mouse. Research conducted on mouse fibroblast cells, where they were implanted with ammoniated and ammonia-free NRL membranes, revealed significant differences in cell viability within 72 h. The ammoniated NRL membrane exhibited a threefold decrease in cell viability compared to the ammonia-free NRL membrane. Moreover, the ammoniated membrane was found to be genotoxic, showing a 15-fold increase in damage compared to the ammonia-free membrane. These results strongly suggest that the ammoniated NRL membrane is not biocompatible and poses a hazardous risk. Furthermore, subjects injected with the ammoniated NRL membrane experienced a substantial delay in wound healing and an acute inflammatory response in the tissues surrounding the implant site. These findings indicate the potential risks associated with using ammoniated NRL membranes and raise concerns about their suitability for biomedical applications.<sup>5,11</sup> Despite the devastating consequences of ammonia-based latex, natural rubber continues to be a valuable resource in producing a wide range of everyday products, from tires and hoses to gloves and adhesives.<sup>12–15</sup> The natural rubber tree produces the quintessential biopolymer with the largest molecular weight, resulting in a material with exceptional mechanical properties that outperform those of its synthetic counterparts.<sup>16</sup> The added benefit of the natural rubber tree is that it also serves as a sustainable polymerization plant that can approximately provide us with 1 kg of natural rubber, all while sequestering 3 kgs of CO<sub>2</sub>, releasing 3 kgs of clean oxygen into the atmosphere, and consuming 1 kg of water (Figure 1).



**FIGURE 1** A rubber tree depicted as a natural polymerization plant where the tree can provide us with 1 kilogram of natural rubber, all while intaking 1 kg of water, sequestering 3 kg of CO<sub>2</sub>, and releasing 3 kg of clean oxygen.

To meet the growing demand for sustainable alternatives, biopolymers such as natural rubber can be considered.<sup>17,18</sup> There has been an increased interest in sustainable materials such as natural rubber. Areas of interest include stabilization techniques,<sup>17,19</sup> as well as life cycle analyses that show how natural rubber is superior to any synthetic materials it replaces.<sup>20</sup> As stated, natural rubber has been used in the adhesives industry, however, most natural rubber adhesives are solvent-based adhesives with a wide variety of formulations and properties.<sup>12–15</sup> To facilitate the development of such adhesives, organic solvents are commonly used to dissolve or disperse the adhesive components.<sup>21</sup> Proper dispersion of components promotes a homogeneous adhesive mixture, ultimately ensuring optimal performance by eliminating defects that may produce stress concentrations. In addition, solvents improve the adhesive's viscosity profile, facilitating the adhesive's application, and serve as a method to soften the substrate surface, promoting diffusion and proper bonding.<sup>19</sup> However, a detrimental characteristic of solvents is that they also rapidly vaporize upon application, producing volatile organic compound emissions (VOC), leaving the user prone to respiratory problems and other health issues.<sup>22</sup> Numerous studies have shown the direct health implications caused by solvent-based adhesives, particularly in factory workers exposed to those compounds over their careers.<sup>23–27</sup> Heuser and coworkers showed genetic damage in Brazilian footwear manufacturing workers exposed to solvent-based adhesives. The researchers compared them with workers who handled water-based adhesives and revealed that workers exposed to solvent-based adhesives for 5 years showed significant DNA

damage and mutations, including nasal cancer and leukemia.<sup>28</sup>

Furthermore, the strict regulations for treating contaminated water make it difficult for small plantation owners, which represent the majority of the operations,<sup>29,30</sup> to comply, inadvertently blocking the progress of initiatives put in place. To compound the issue, using ammonia in natural rubber processing produces a strong odor and fosters allergenic molecules. Researchers have demonstrated that the addition of ammonia causes protein breakdown, leading to the formation of allergenic proteins, specifically the shortest chain proteins found in the serum. The study also offers evidence comparing ammoniated and non-ammoniated latexes using ELISA testing to highlight the distinctions in allergenic proteins, such as Hev b 1, Hev b 3, Hev b 5, and Hev b 6.02,<sup>16,20,31–33</sup> making it unattractive for various industries,<sup>4</sup> despite its superior mechanical properties. It is also vital to state that the environmental friendliness of natural rubber latex adhesives is significantly compromised by using toxic organic solvents, even though NRL is a sustainable alternative with excellent properties for the adhesives industry. Therefore, mitigating detrimental health effects to adhesive handlers needs to be addressed by searching for alternative additives capable of performing as well as solvent-based adhesives.

There are NRL-based adhesives that include alternative additives, such as formaldehyde instead of solvents, however, it has yet to perform as well as solvent-based adhesives.<sup>31</sup> Traditionally, solvent-based adhesives have outperformed biobased adhesives, but they have done so at the expense of the users. To enhance their strength, the natural rubber must be chemically modified or blended with synthetic resin.<sup>34,35</sup> This corroborates with current research, as it has been primarily shown that the properties of NRL-based adhesives can be improved by incorporating other bio-based polymers as fillers. For example, starch-reinforced NRL-based adhesives have been found to enhance the morphologies and mechanical properties of the overall adhesive system.<sup>32,34</sup> Additionally, modification to synthetic polyurethane systems have also been done to achieve improved peel strength characteristics, but still do not reach solvent-based adhesive properties.<sup>36</sup> It is hypothesized that the limitations of natural rubber latex adhesives within industry is primarily due the presence of water within its composition. Additives commonly used within the adhesives industry are not soluble in water, therefore, making it challenging to adapt current solvent-based formulations to natural rubber latex formulations. Additionally, natural rubber latex is susceptible to coagulation if an additive alters the zeta potential of the overall blend. Therefore, one must navigate through the multivariable balance between surfactants and additives to ensure solubility is optimized and stabilizing is maintained.

This study presents a novel NRL-based adhesive that outperforms ammonia-based and solvent-based adhesives by utilizing the novel ammonia-free natural rubber latex.<sup>16,20</sup> There are multiple formulations of this natural rubber latex that is preserved in a cationic or acidic environment.<sup>16</sup> The adhesives tested in this study are based on one of these ammonia-free liquid latex systems produced using the components listed in Table 1 and has been in a stabilized state for over 4 years.

## 2 | MATERIALS AND METHODS

### 2.1 | Material composition

Five commercially available adhesives, highly relevant in various industries, were evaluated and benchmarked against the two adhesives utilizing the ammonia-free natural rubber latex as its primary raw material. This is to provide proof of this sustainable adhesives' ability to outperform adhesives highly utilized in industry. Tables 2–4 below presents each adhesive along with its material composition, and classification information regarding toxicity. To note, SOAN Laboratories bio adhesives are produced using Alfapreno, one of their recently patented ammonia-free natural rubber latex (AF Latex) as its

**TABLE 1** Composition of the ammonia free natural rubber latex from SOAN Laboratories, specifically the latex grade named: Alfapreno.

| Component                              | Amount [%vol] |
|--|---------------|
| Natural rubber latex                   | 98.6          |
| Ethoxylated tridecyl alcohol           | 1             |
| 50/50 hydrofluoric acid/water solution | 0.4           |

**TABLE 2** Material composition of the SOAN laboratories AF latex industrial adhesive.

| Component  | Amount [%wt] |
|--|--------------|
| Centrifuged alfapreno natural rubber latex (60% DRC) | 99           |
| Hydroxyethyl cellulose                               | 0.5          |
| Collagen   | 0.5          |

**TABLE 3** Material composition of the SOAN laboratories AF latex office adhesive.

| Component                                | Amount [%wt] |
|--|--------------|
| Alfapreno natural rubber latex (35% DRC) | 99.2         |
| Hydroxyethyl cellulose                   | 0.8          |

TABLE 4 The material composition of the solvent-based adhesives benchmarked against the AF Latex Industrial and Office adhesive.

| Adhesive name                          | Component       | Amount [%wt] | Classification   |
|--|-----------------|--------------|--|
| Pattex contact adhesive <sup>37</sup>  | Acetone         | < 50         | May cause organ toxicity and serious eye irritation.   |
|  | Solvent naphtha | < 30         | Suspected carcinogen, flammable and aspiration hazards   |
|  | Ethyl acetate   | < 25         | Highly flammable liquid and vapor. Causes eye irritation and may cause drowsiness or dizziness.            |
|  | Zinc oxide      | < 0.25       | May cause acute hazards and chronic hazards.   |
| Angelus contact adhesive <sup>38</sup> | Ethyl acetate   | < 35–60      | Highly flammable liquid and vapor. Causes eye irritation and may cause drowsiness or dizziness.            |
|  | Heptane         | < 12–25      | May cause hallucinations, distorted perception, and change in blood or tissue levels.                      |
|  | Cyclohexane     | < 12–25      | May cause changes in kidney, ureter, and bladder health.   |
| Carpet Parabond M-260 <sup>39</sup>    | n-hexane        | 25–50        | Highly flammable with aspiration hazard and reproductive toxicity. May cause skin irritation upon contact. |
|  | Toluene         | 5–20         | Reproductive toxicity, specific target organ toxicity skin irritation and aspiration hazard.               |
|  | Naphthalene     | *            | Suspected carcinogen, flammable and aspiration hazards   |

\*Compound identified within safety data sheet without mention of content.

matrix of choice.<sup>16,20</sup> To draw conclusions between ammoniated vs. ammonia-free, an adhesive based on traditional ammoniated natural rubber latex technology was acquired from Continental de Pegantes y Soluciones S.A.S. and evaluated. No information is available regarding the material compositions besides that it comprises ammoniated natural rubber latex.

It can be appreciated from Tables 2 and 3 above that AF Latex-based bio adhesives primarily contain the natural rubber latex with minuscule amounts of additives which do not pose a risk to the user and manufacturer. In contrast to the compounds seen in Table 4 below, these adhesives include highly toxic chemicals which require users to be within well-ventilated areas.

The AF Latex office adhesive was compared to Elmer's Office glue, where Elmer's is the leading adhesive brand in the United States with 23.6% sales share.<sup>40</sup> Elmer's office glue is a proprietary Polyvinyl Acetate based adhesive which has no composition information within their material data sheet as it declares a 100% mixture of proprietary non-hazardous ingredients within the material data sheet.<sup>41</sup>

## 2.2 | Performance evaluation methodology

### 2.2.1 | T-Peel test—ASTM standard F2256

An MTS Criterion Model 43 tensile testing machine equipped with a 50kN load cell (MTS Model LPS.504) was employed for conducting peeling strength tests.

To ensure proper load transfer, the Advantage™ wedge action grips (Model no. 50, 2716-015) from MTS Systems Corporation were used. The action grips were instrumental for ensuring that the sample remained locked in position and that slippage was minimized. Sample specimen were cut from natural grain cow leather provided by Tannery NYC sheets with a width and length dimension of 2.54 cm and 15 cm, respectively. Two specimens were then bonded by 12.5 cm of the total 15 cm length and a constant force was applied to the specimen for 24 hours to promote full adherence. An exemplary illustration of the T-Peel test can be seen in Figure 2 below, in which the arrows indicate the direction of deformation. Pursuant to the ASTM F2256 standard, the conditioned sample was deformed using a crosshead speed of 250 mm/min.

Calculation of the average T-Peel strength, also known as the interfacial toughness  $T$  of the adhesive is reported as follows

$$T = \frac{F}{w},$$

where  $F$  is the force in Newtons and  $w$  is the width in centimeters.

### 2.2.2 | Carpet adhesive shear strength test—ASTM Standard D6004.

Similar to the T-Peel Test mentioned above, the MTS tensile testing machine setup was utilized for the

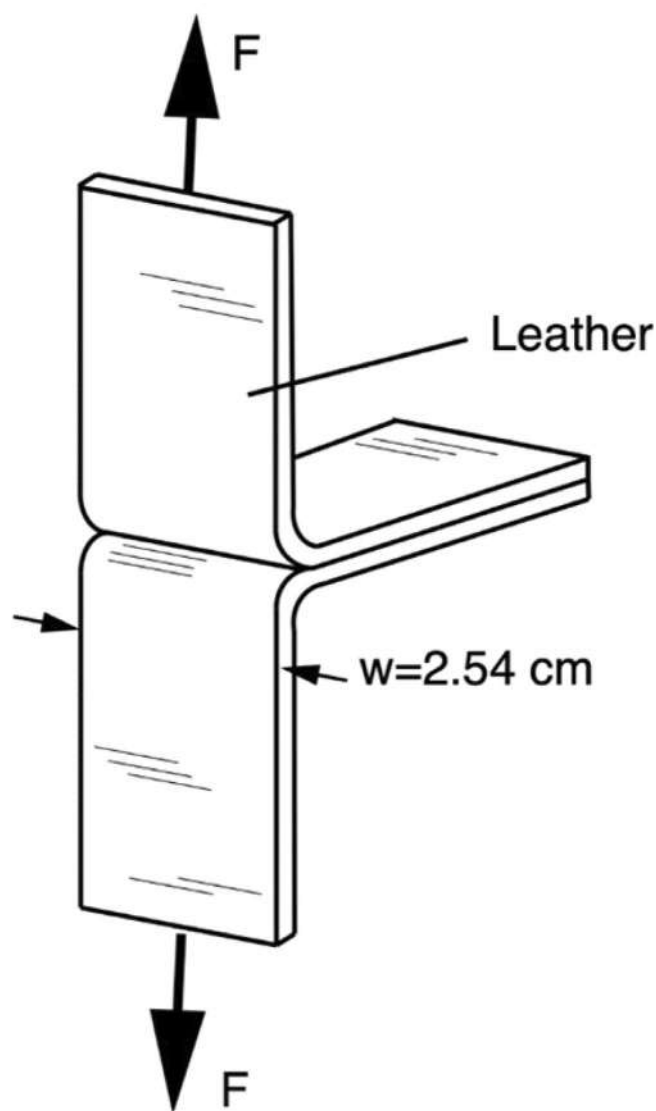


FIGURE 2 An illustration of the T-Peel test with arrows pointing in the force direction.

evaluation of the carpet adhesive. The carpet backing test specimen Canvas Duck Natural, Fabric by the yard sold by FoamSpot were cut to a 2.54 cm width and bonded to the Sande plywood substrate, resulting in a total bonding length of 7.62 cm (3"). Figure 3 below depicts the testing schematic, where it can be seen that one of the tensile grippers holds onto the carpet backing while the other grippers sustain the plywood substrate commonly used in the construction industry.

In accordance with the ASTM D6004 standard, cross-head speed is set to 2.54 cm/min, whereas the interfacial shear strength of the adhesive  $S$  is reported as follows,

$$S = \frac{F}{L \cdot w},$$

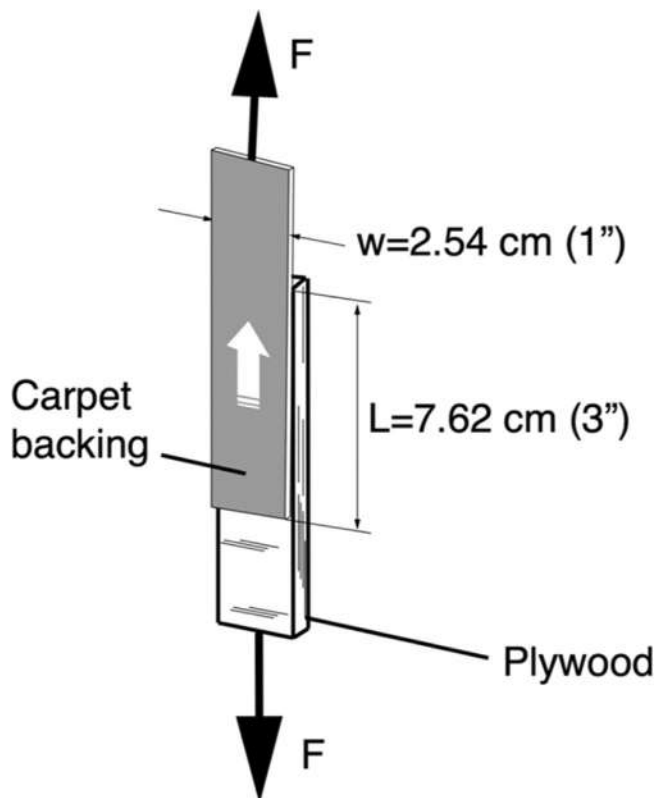


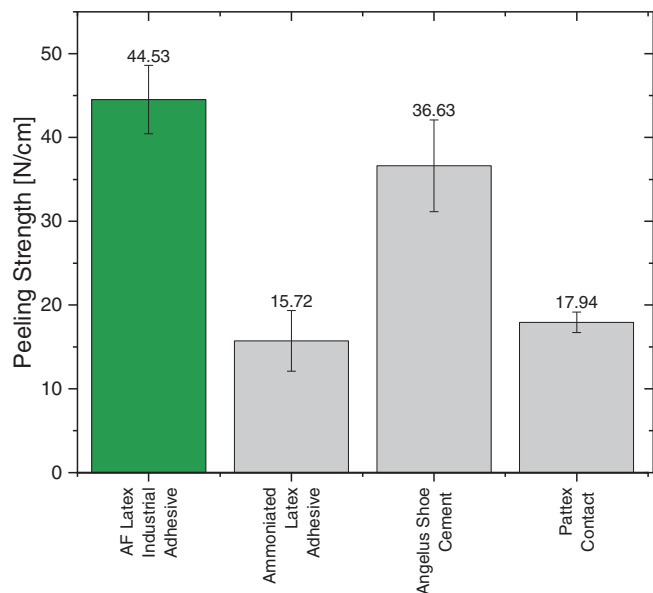
FIGURE 3 Illustration of the testing schematic for the carpet adhesive testing with arrows pointing in the direction of the force.

where  $F$  is the force in Newtons,  $w$  is the width of the specimen in meters and  $L$  is the length of the specimen bonded onto the substrate in meters.

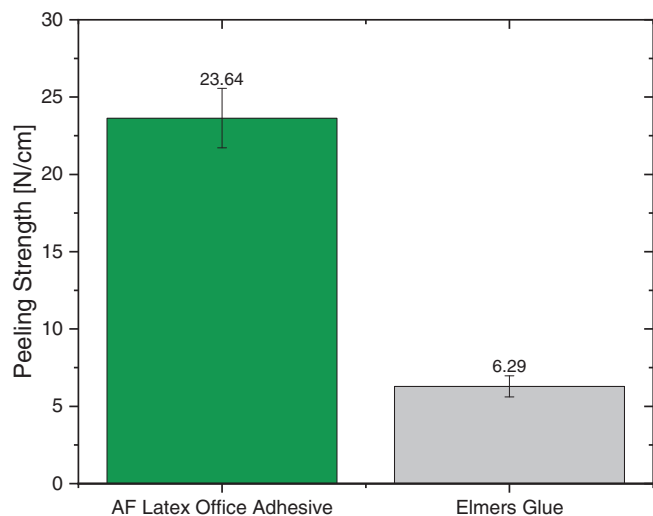
### 3 | RESULTS AND DISCUSSION

#### 3.1 | AF latex industrial adhesive evaluation

A minimum of six T-Peel tests were conducted to ensure reproducibility in results by using the ASTM F2256 standard. As seen in Figure 4 below, it can be concluded that the AF Latex industrial adhesive outperforms all other adhesives which are produced using toxic chemicals from non-renewable sources. When comparing the ammonia-free natural rubber latex adhesive with the ammoniated version, we see that the ammonia-free bio adhesive is 2.83 times stronger. The influence of protein content on natural rubber material behavior and theorized that higher modulus and larger tear strength in natural rubber is associated with greater protein content.<sup>42</sup> Although ammonia successfully preserves and stabilizes the natural rubber latex, it does so at the expense of the proteins and



**FIGURE 4** T-Peel tests for leather-based adhesives in accordance to ASTM F2265.

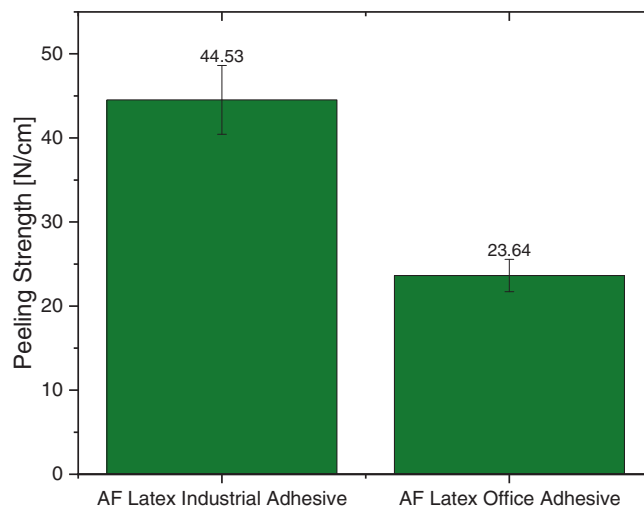


**FIGURE 5** The peeling strength results of AF Latex office glue is 3.75 times larger than traditional office glue, tested in accordance to ASTM F2256.

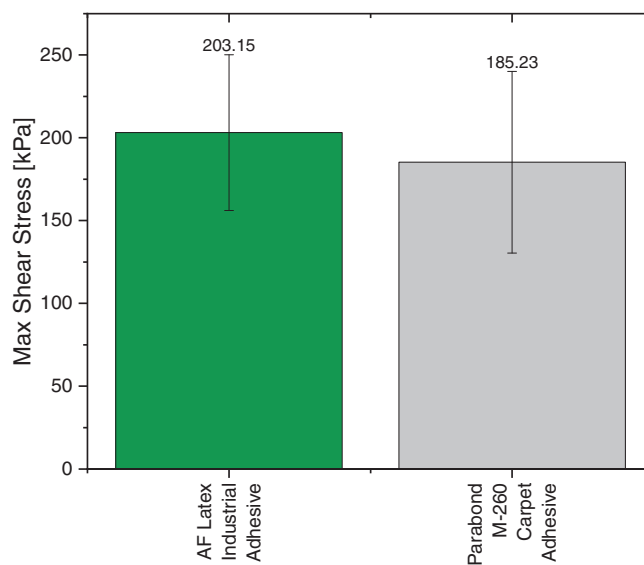
many living organisms in charge of enhancing mechanical performance.

### 3.2 | AF latex office adhesive evaluation

The T-Peel tests of office adhesives were performed using the ASTM F2256 standard and it can be concluded (Figure 5) that natural rubber latex based adhesive results in adhesive which is 3.75 times stronger than the Elmer's Polyvinyl Acetate based adhesive. This comparison is



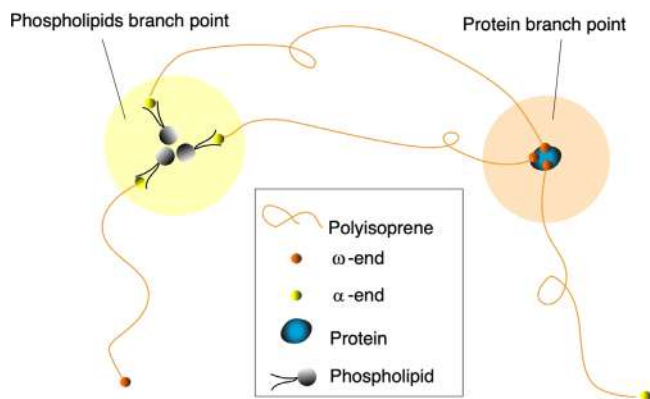
**FIGURE 6** A comparison in peeling strength between both AF Latex adhesives grades, whereas the key differentiator is the dry rubber content present in the latex.



**FIGURE 7** A comparison in shear strength performance between the AF Latex Industrial Adhesive and the traditional solvent-based carpet adhesive.

crucial as it provides proof that a natural rubber latex adhesive which has not been centrifuged has the ability of surpassing one of leading adhesives within the office supplies industry.

Upon inspection of Tables 2 and 3 above, the key difference in composition between both AF Latex adhesives is that the AF Latex Industrial Adhesive is made from Alfapreno natural rubber latex which was centrifuged to increase the dry rubber content from 35% to 60%, while the AF Latex Office Adhesive was formulated using non-centrifuged Alfapreno natural rubber latex with the



**FIGURE 8** Natural rubber molecular network with protein and phospholipids branch points. It is presumed that natural rubber forms a network structure through the aggregation of proteins and phospholipids in polyisoprene segments, where the ends of main chain natural rubber molecules interact with phospholipids and proteins, respectively.<sup>20</sup>

original 35% dry rubber content. Assuming the rules of mixtures applies to this application, doubling the quantity of rubber content essentially results in twice the amount of material capable of sustaining stresses.<sup>43</sup> For that reason, it is hypothesized that having twice the amount of dry rubber content within the mixture results in an adhesive with two times the peeling strength. This can be appreciated in Figure 6 below where the AF Latex Industrial Adhesive and the AF Latex Office Adhesive results in a peeling strength of 45.53 N/cm and 23.64 N/cm, respectively.

### 3.3 | Carpet adhesive evaluation

With the construction industry ranking 2nd on the list of the 10 biggest industries in the world in 2021, resulting in approximately 13% of the world's GDP,<sup>44</sup> it is vital to replace adhesives used within the construction with a more sustainable alternative capable of at least performing on par to traditional adhesives used currently. The AF Latex Industrial Adhesive underwent shear strength tests in accordance with the ASTM D6004 standard to evaluate its applicability within the construction industry, specifically within the carpet industry. The above-mentioned ASTM standard is used to measure shear strength of adhesives used to bond resilient flooring and carpet to selected substrates. It can be appreciated in Figure 7 below how the ammonia-free natural rubber latex adhesive outperforms the Parabond M-260 carpet adhesive traditionally used in the United States carpet industry.

## 4 | CONCLUSIONS

Contrary to what has been described in the literature, a fully bio-based adhesive is capable of outperforming solvent-based adhesives. Figure 8 presents a schematic of a polyisoprene molecular network with phospholipid and protein branch points.<sup>20</sup> In contrast to this novel natural rubber liquid latex stabilization and preservation technique which leaves this network intact, an ammonia-based stabilization and preservation process destroys the protein and phospholipids which serve as branch points, leading to a rubber material with lower mechanical properties. For that reason it is theorized that one of the reasons why the ammonia-free natural rubber latex adhesive outperforms most technologies is because these branch points serve as a natural reinforcement.

T-Peel tests confirmed that the AF Latex Industrial Adhesive had a peeling strength of 44.53 N/cm while the best solvent-based adhesive reached a peeling strength of 36.63 N/cm. Furthermore, the AF Latex Office Adhesive was tested against Elmer's Glue, whereas the AF Latex Office Adhesive reached a peeling strength of 23.65 N/cm, while Elmer's Glue resulted in a peeling strength of 6.29 N/cm. Finally, AF Latex Industrial Adhesive was benchmarked against an adhesive traditionally used in the carpet industry. In accordance with the ASTM D6004 standard, the AF latex Industrial Adhesive reached a max shear stress value of 203.15 kPa while the Parabond M-260 carpet adhesive reached 185.23 kPa.

Additional AF Latex-based adhesives are in the development stage as there exists various grades of ammonia-free natural rubber latex, such as, Betapreno, Gammapreno, Epsilonpreno, Thetapreno, all which offer a wide variety of chemical bond types. Furthermore, the above mentioned Alfapreno grades are only two formulations using cellulose and collagen, which opens the doors to a variety of future adhesive blends that have the ability of providing a wide range of sustainable adhesives with comparable strength to solvent-based adhesives.

### DATA AVAILABILITY STATEMENT

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

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