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Study of the influence of tack resins on elastomeric formulations and applied evaluation methodologies

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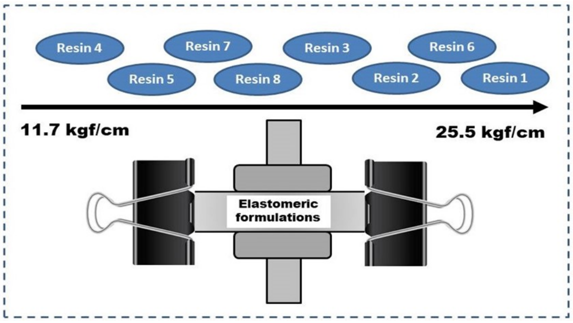

Abstract: In this study, we examined the impact of eight distinct types of tack resins sourced from diverse origins (phenol-formaldehyde: SP – 1068, CRJ – 418, Koresin and SRF – 1501; hydrocarbons: Unilene A – 90 and Plastack RB 809; coumarone and breu) in several elastomeric formulations. For this purpose, it was developed a simple, robust and easily reproducible tack test employing synthetic rubber to assess these formulations. The elastomeric compounds were characterized by attenuated total reflectance Fourier transform infrared spectroscopy, rheometry, Mooney viscosity, crosslinking density by swelling in solvent, hardness, abrasion wear, tensile and tear strength, followed by tack tests. Overall, the formulations exhibited similar physical-mechanical properties. Regarding the tack test results, SP-1068 (phenol-formaldehyde) and Plastack RB 809 (hydrocarbon) resins demonstrated higher values, whereas the resorcinol-type phenol-formaldehyde presented a reduction of 10.7 %. Furthermore, interference from the functional groups in the resin's chemical structure, difficulty in interacting with the matrix, and lower crosslink density were observed. In conclusion, the developed methodology proved satisfactory in yielding reliable and reproducible tack values.

Keywords: Elastomeric formulations, adhesion, tack, resins.

Introduction

Adhesion is the mechanical strength of the interface between the adhesive and the substrate, which can be interpreted as a bond between the adhesive film and the substrate under the action of light pressure. This term is known as instant adhesion and is usually called tack or stickiness. The expressions tack, tackiness and stickiness emerged with the rubber industry, specifically because natural rubber presents stickiness on its surface during prolonged storage [1]. Tack is one of the most important properties in the tire industry. There are some factors that influence this property, such as contact time, pressure exerted on the substrate with the adhesive material, and temperature. Another factor of great relevance for this property is related to the viscoelastic characteristics of the material [2].

It is currently classified into two types. The first is "specific adhesion", also known as chemical adhesion, and the second is "mechanical adhesion", which occurs with the penetration of the adhesive into the pores of the substrate, forming species of mechanical fastening hooks. Cohesion, on the other hand, can be defined as the internal resistance of the adhesive, which consists of the ability of the adhesive film to resist a peeling force [3]. Therefore, it is not possible to explain this mechanism separately, as it is the combination of several forces that overlap and influence each other [4]. In the industrial area, there are substances capable of improving adherence in rubber compounds. They are usually resins derived from different sources and classified according to their chemical origin, natural source and synthetic source. In this way, the resins have the function of promoting wetting and adhesion, contributing to the increase of tack and flexibility [5].

The literature provides different alternative methodologies and equipment options for determining tack measurements. In the method proposed by Forbes and McLeod [6], for instance, two circular rubber samples were initially prepared through compression in cylinders, subsequently cut and reassembled with a Teflon disc between them. After obtaining the samples cut in half with flat surfaces, they were compressed and separated in a universal mechanical testing machine (Instron). In an effort to ascertain the adhesion of fabric screens in the rubber factory, Pickup [7] successfully devised an instrument regarded as both simple and robust, where only a strip of the material to be tested was fixed to a wheel. This wheel was connected to a weight via a calibrated spring. The weighted wheel was positioned on a sheet of the material to be tested, ensuring that the two surfaces were pressed together. After a certain time of contact, the weight increased, causing the spring to extend and, at the moment the surfaces separated, the spring tension was obtained as a measure of adhesion. However, the reproducibility of this test was not considered very adequate due to the small surface area used in each test.

Despite the availability of these methods, achieving accurate and repeatable results for this property remains challenging. Numerous adaptations have been attempted on existing methodologies; however, there is still no certainty of obtaining reliable results for tack measurements. For this reason, it is important to carry out further studies to measure this property, which is so important in the elastomeric field. Taking into account all these aspects, the main goal of this study is to assess the influence of eight tack resins on the adhesion and cohesion properties of elastomeric compounds, and then to develop a reproducible methodology for determining tack in these materials.

Experimental Section

Materials

The materials and amounts used in this research were: 100 phr SBR-1502 polymer (Arlanxeo), 5 phr ZnO (Nexa Resource), 3 phr stearin (Baerlocher), 60 phr carbon black N-550 (Birla Carbon Brasil), 2 phr sulfur (Phelps), 1.5 phr cyclohexyl-2-benzothiazole sulfenamide accelerator (Shandong Yanggu Huatai), 0.15 phr diphenylguanidine accelerator (Zhedong Zhejiang), and 0.1 phr tetramethylthiuram disulfide accelerator (Shandong Yanggu). Eight resins were evaluated with the fixed amount of 8 phr in each formulation: SP-1068 named “Resin 1” (Crios Resinas Group), CRJ-418 named “Resin 2” (SI Group), Koresin named “Resin 3” (Basf SE), SRF-1501 named “Resin 4” (Crios Resinas Group), Unilene A-90 named “Resin 5” (Braskem), Plastack RB 809 named “Resin 6” (Henghe Materials and Science Technology), Coumarone named “Resin 7” (ChemiChem International), and Breu WW/SBT 100 named “Resin 8” (Química Ambiental Ltda).

Resin 1, derived from octylphenol and formaldehyde, is a thermoplastic resin commonly employed to enhance the grip on both natural rubber and synthetic elastomers. Resin 2, a thermoplastic alkylphenol resin supplied in flakes, serves primarily as a tackifier in NR (natural rubber) and SBR (styrene-butadiene rubber)-based rubber compounds. It is also employed in various cement applications for tire seams and technical rubber products. Resin 3 stands out as the sole formaldehyde-free phenolic compound available on the market, distinguished by its compatibility with all contemporary rubber formulations. Resin 4 is a standard Novolak resorcinol resin. When combined with a methylene donor and added to the rubber stock, this resin acts as an adhesion promoter, enhancing the adhesion of the rubber compound. Resin 5 is a C9 thermoplastic hydrocarbon derived from the catalytic polymerization of hydrocarbons through a continuous process. It exhibits compatibility with a wide range of solvents and polymeric materials, characterized by its hydrophobic nature and light-yellow pellet form. Resin 6 is a hydrocarbon resin, classified as a petroleum thermoplastic resin, soluble in solvents based on hydrocarbons. Resin 7 is characterized by its dark brown granular solid appearance and is derived from the C9 fraction, a by-product of ethylene, through thermal polymerization. It boasts excellent solubility, chemical stability, and offers strong adhesive properties along with lower heat conduction. Resin 8 is a solid resin, shaped into compact flakes with a distinct clear yellowish-amber color, and remains insoluble in water at 20 °C.

Attenuated total reflectance (ATR) Fourier transform infrared (FTIR) spectroscopy

The resins were characterized by ATR-FTIR spectroscopy employing a Perkin Elmer spectrometer in the region between 4000 and 500 cm^{-1} and with a resolution of 4 cm^{-1}.

Bands within the region of 3700-3500 cm^{-1} were selected for quantitative analysis using OriginPro® 9.0 software. Quantitative analysis was determined by dividing the area of the OH vibration peak by the total area encompassing all peaks within the entire spectrum.

Preparation of the elastomeric compositions

The formulations were prepared from a standard elastomeric formulation, varying the tack resin type sourced from different origins. The standard compound did not contain any adhesive resin. Initially, the mixing process was conducted in a closed mixer (Banbury, Copé) at 40 rpm and 160 °C. After 24 h, the mixture was accelerated in an open mixer (cylinders) at 60 °C for 180 s, with a friction ratio between the cylinders set at 1:1.25.

Characterization of the elastomeric compositions

Mooney viscosity and cure characteristics

Mooney viscosity analyses (Mooney MV 2000 - Alpha Technologies) were carried out according to ASTM 646-07 [8] at 100 °C and 1 + 4 min with a small rotor. The tests were performed in triplicate.

Curing tests were performed with an MDR 2000 moving die rheometer (Alpha Technologies Inc.) according to ASTM D 5289-12 [9]. The minimum torque (M_L), maximum torque (M_H), difference between M_H and M_L (ΔM), scorch time (t_{s1}), and cure time (t_{90}) were determined at 150 ºC for 10 min. These tests were also performed in triplicate.

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Tack test

The samples with a defined area were placed in the middle of the claws that were made and adapted to the Instron equipment (Figure 1). The contact between the samples occurred for a few seconds, and then the claws were separated to measure the force required to detach the samples (tack). For the tack test, three specimens were molded with an average thickness of 2.1 mm. The bottom (5.0 × 5.0 mm) and top (10.0 × 5.0 mm) of each specimen were then folded and placed in the same holder (detail in Figure 1A) as if was a sandwich sample - sample holder bottom - sample holder top - sample before the test. The rate of jaw separation was set at 500 mm min^{-1}.

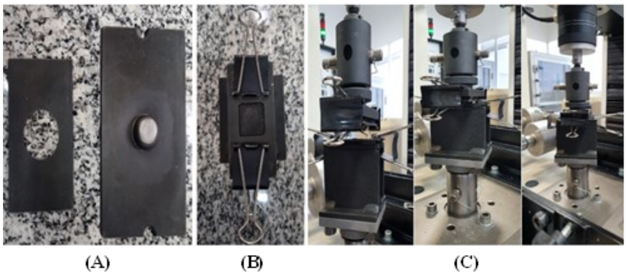

Figure 1. (A) Sample holder (bottom - left, and top part - right); (B) sandwich made with the two parts indicated in (A); (C) samples being tested.

Crosslinking density

The crosslinking density was performed by the swelling method. In this procedure, samples were immersed in heptane and maintained in darkness at 23 ± 2 °C for 120 h. The formulations were weighed before and after immersion, and the crosslinking density was calculated using the Flory-Rehner equation [10], with Kraus correction [11]. Density values (Alfa Mirage, MD-300S) were determined in accordance with ASTM D 297-13 [12].

Mechanical tests

The hardness tests were conducted using a Shore A Teclock durometer (Bareiss) following the specifications outlined in ASTM D 2240-05 [13]. The abrasion test was carried out using a MAQTEST abrasion meter in accordance with DIN ISO 4649-10 (method A) [14]. The tear strength test of the compositions was executed according to the ASTM 624-00 [15], while tensile strength tests were undertaken using an Instron2000 tensometer (Alpha Technologies) equipped with a 20 kN load cell and displacement rate of 500 mm min^{-1}, according to ASTM 412-06 [16]. All tests were performed in triplicate and the results were based on the median of the determinations.

Results and Discussion

ATR-FTIR

Figure 2 displays the ATR-FTIR spectra of the tack resins. The broad bands observed in the region between 3700 and 3000 cm^{-1} correspond to the axial OH deformation of phenols. These assignments were identified in the spectra of Resins 1, 2, 3, and 4, with a more pronounced intensity in Resin 4, possibly due to the presence of resorcinol (which contains two OH groups per structural unit). The appearance of minor peaks in the fingerprint region (1490-1030 cm^{-1}) is associated with C–O bonds attributed to phenolic groups. In the spectra of Resins 5, 6, 7, and 8, bands were observed in the region of 2990-2730 cm^{-1} corresponding to C–H stretching related to CH_2 and CH_3 groups. Moreover, bands at 700 cm^{-1} confirm the out-of-plane vibrations of the same hydrogens observed in the 2990 cm^{-1} region.

Quantitative analysis performed using ATR-FTIR after normalizing the spectra revealed the following sequence: 0.15, 0.11, 0.070, 0.33, 0.0016, 0.023, 0.0060, and 0.033, for Resins 1, 2, 3, 4, 5, 6, 7, and 8, respectively. These results confirm that Resin 4 is the most polar, while Resin 5 is the least polar among them all.

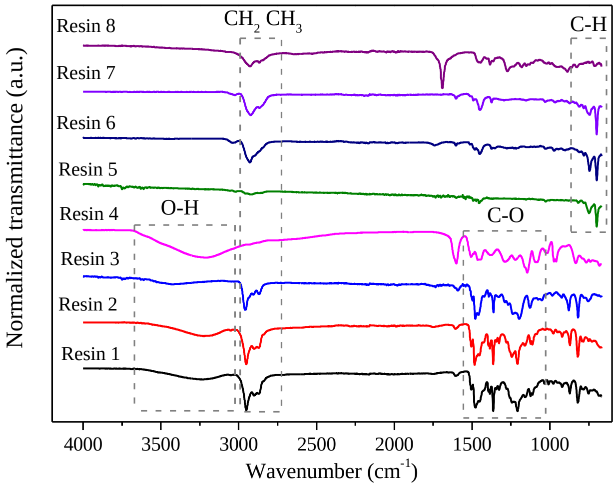

Figure 2. Normalized ATR-FTIR spectra with baseline correction of the evaluated resins.

Mooney viscosity and cure characteristics

Mooney viscosity and torque characteristics of the elastomeric compositions are shown in Figure 3. Mooney viscosity is associated with processability, extrusion, and molecular weight. In this study, the formulations with Resins 5, 6, and 7 exhibited similar values for this property. The formulation using Resin 4 displayed the highest value being 12.7 % higher than the standard formulation, whereas the remaining formulations yielded lower results (approximately 22.5 % reduction). The high viscosity values of Resin 4 can be attributed to the weak chemical affinity between the rubber and the resin, which originates from resorcinol. The presence of multiple polar OH groups in the resin structure hinders the miscibility of the polymer chains of both compounds, resulting in higher viscosity values.

Regarding minimum torque (M_L), all formulations showed similar values, which are directly related to the viscosity in the initial stage of the vulcanization process, where crosslinking has not yet occurred. Therefore, the inclusion of tack resins likely did not influence the M_L values before vulcanization, also indicating little variation in the processability of the formulations. This result is expected since M_L was measured during the initial stage of the vulcanization process [17].

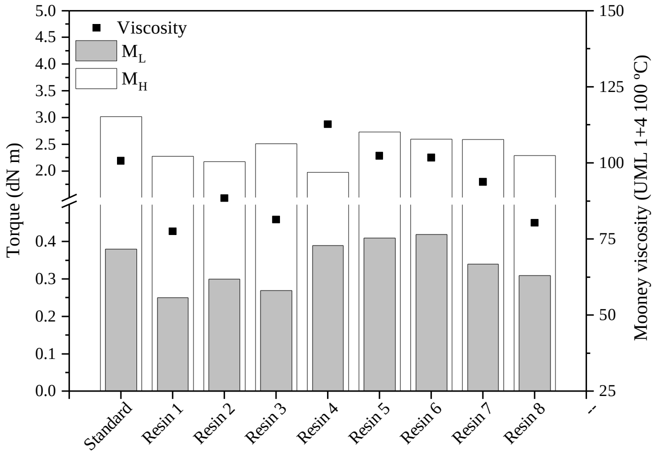

Figure 3. Mooney viscosity and torque characteristics of the studied elastomeric formulations.

In the case of maximum torque (M_H), which can be related to the material's stiffness, it was noted that, overall, formulations with different resins experienced a reduction in M_H compared to the standard formulation. This suggests that the incorporation of resins is hindering the formation of the crosslinking network. Furthermore, the internal lubrication effect of the resins on the chains may also impede crosslinking formation. Formulations using Resins 3, 6, and 7 presented similar values to each other, with reductions of 16.9, 13.9, and 14.2 %, respectively, compared to the standard formulation. Conversely, formulations with Resins 4 and 5 experienced the highest and lowest reductions (34.5 and 9.7 %, respectively).

Table 1 presents the results obtained for ΔM. As can be observed, the standard formulation has the highest number of crosslinks.

Table 1. Results of ΔM for the elastomeric formulations developed with different types of resins.

Formulation	ΔM (dN m)
Standard	2.6
Resin 1	2.0
Resin 2	1.9
Resin 3	2.2
Resin 4	1.6
Resin 5	2.3
Resin 6	2.2
Resin 7	2.2
Resin 8	2.0

The formulation with Resin 4 had the lowest ΔM value, indicating a lower quantity of crosslinks. Elastomeric formulations with Resins 1, 2, and 3, on the other hand, showed similar values, as did the formulations with Resins 5, 6, 7, and 8. Thus, it can be concluded that the resins used in the formulations were influencing the curing reaction of the developed elastomeric compounds, as well as the quantity of crosslinks. These results are supported by the swelling results that will be discussed later.

Regarding the process safety time (t_{s2}) (Figure 4), the formulations with Resins 5, 6, 7, and 8 had values similar to the standard formulation, indicating that the use of these resins does not change the safety of the process [17]. On the contrary, the formulations with Resins 1, 2, and 3 showed a reduction of 41.4 %, while the formulation with Resin 4 presented an even lower value, with a 53.5 % reduction rate. This result indicates that resins derived from hydrocarbons, coumarone and natural do not interfere in the vulcanization process, while resins derived from phenol-formaldehyde considerably reduce the safety of the process. In the optimal vulcanization time (t_{90}), we observed similarity in most samples, indicating that, regardless of the type of resin, there was no interference in the vulcanization time of the compounds. Only the formulation using Resin 3 showed a reduction of 35.5 %, while the formulation using Resin 4 showed a higher t_{90} value (an increase of 89.5 %). In these two formulations, antagonistic effects were observed, indicating that Resin 3, as it belongs to the group of resins derived from phenol-formaldehyde, accelerates the vulcanization process and Resin 4 (resorcinol resin) delays it, possibly due to parallel reactions of the accelerators with this resin.

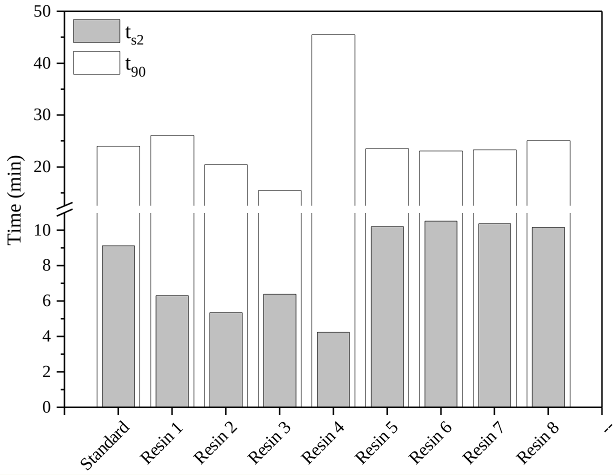

Figure 4. Cure results of the studied elastomeric formulations.

Concerning the curing tests (Figure 4), the formulation using Resin 4 showed a lower t_{s2} compared to the standard formulation (53.5 %) and an increase of 89.5 % in t_{90}. This result suggests that this resin absorbs from the accelerator system and does not vulcanize, as well as the influence of the pH of the medium (acidic character of resorcinol) deactivates the accelerators and delays the curing of the material [18]. For the formulations containing Resins 5, 6, 7, and 8, t_{s2} and t_{90} showed values similar to the standard formulation and this can be explained by the fact that aromatic resins are highly compatible with synthetic elastomers [19].

Tack and crosslinking density

Tack is one of the most important properties in the tire industry, as it is necessary to avoid prior deformation of the product, i.e., to maintain its shape so as not to deform before vulcanizing. The tack value is defined as the force required to dislodge uncured compounds on contact. Table 2 presents the results of the tack test and crosslink density for the eight formulations.

Table 2. Tack and crosslinking density results of the studied elastomeric formulations.

Formulation	**Tack ± SD** (kgf/cm)	**Crosslinking density** (mol/cm^3 ± SD) (×10^{-4})
Standard	13.1± 5.2	5.48 ± 0.09
Resin 1	25.5 ± 8.1	3.61± 0.02
Resin 2	19.4 ± 2.8	3.30 ± 0.04
Resin 3	19.4 ± 2.2	3.40 ± 0.02
Resin 4	11.7 ± 1.9	1.12 ± 0.05
Resin 5	14.2 ± 2.2	4.63 ± 0.08
Resin 6	23.9 ± 0.8	4.20 ± 0.09
Resin 7	14.5 ± 2.4	4.24 ± 0.03
Resin 8	15.3 ± 2.5	3.34 ± 0.01

As can be observed, the formulation prepared with Resin 1 had the highest tack value (25.5 kgf/cm) while the formulation with Resin 4 had the lowest value (11.7 kgf/cm), indicating that this formulation does not have tack since the formulation value was 13.1 kgf/cm. The other formulations showed similar values; however, in smaller quantities. Formulation prepared with Resin 8, with a low value, still guarantees the tack property, as rosin resins make the compound sticky as they are associated with the elastomeric phase [20].

The complete association between the tack resin and the rubber directly reflects on the final properties, as there must be compatibility at the molecular level between the components of the mixture, especially for hydrocarbon resins that have higher tack values [21]. The resins that presented the best tack values were those that presented the best compatibility with the matrix; therefore, more homogeneous mixtures demonstrate better tack values. The results obtained for the crosslink density (Table 2) corroborate the results for the M_H, where a reduction in the ΔM values as the resins change, indicates that a higher crosslink density may be related to greater rigidity of the material, thus causing higher values of M_H.

Hardness, abrasion and tear strength

The mechanical properties of the studied elastomeric formulations are shown in Figure 5. In the hardness tests, considering the standard deviations, a similarity in the values according to the resin type was observed.

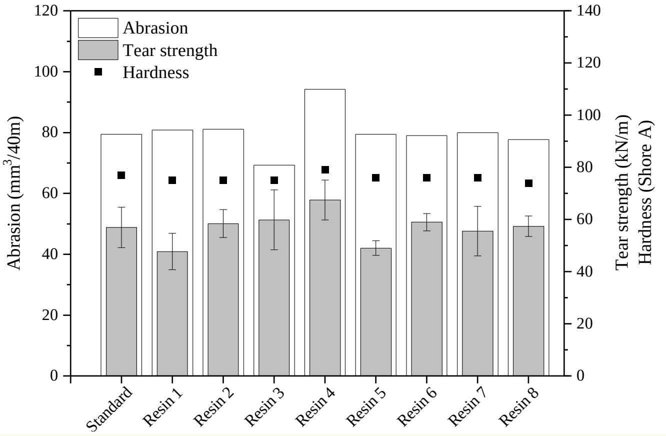

Figure 5. Hardness, abrasion and tear strength of the studied elastomeric formulations.

In general, for the abrasion property, the formulation with Resin 4 showed a higher value (18.6 % of increase), indicating a lower durability of the compositions using this resin. Formulation with Resin 3 showed a lower value (13 % of reduction), indicating a trend towards greater durability of compositions with this resin. These results are related to the stiffness of the material and, consequently, the crosslinking density, where an increase in stiffness provides less wear due to abrasion. Veiga et al. [22] observed similar results, where the abrasion wear values decrease with increasing material stiffness. Regarding the tear strength property, the formulations with Resins 2, 3, and 6 were similar to the standard formulation. The formulation prepared with Resin 4 presented the highest value with an increase of 18.4 %, while for the formulations with Resins 1 and 5 there was a reduction of 16.3 % and 14 %, respectively.

Tensile strength

Strength and elongation at break and modulus at 200 % results are displayed in Figure 6. As can be noted, the formulation with Resin 4 exhibited a 40.26 % decrease in modulus at 200 % compared to the standard formulation. This may be related to the lower formation of crosslinks due to the presence of resorcinol, or that it hinders the vulcanization of the material and influences the rigidity of the material. For the results of the strength at break property, we noticed that all formulations presented higher values than the standard formulation. Formulation with Resin 4 displayed a high elongation value, suggesting an interference of resorcinol in the vulcanization process, which results in fewer crosslinks and, consequently, higher elongation values. Regarding the property of strength at break, no significant variations were observed among the results for the formulations with different types of resins.

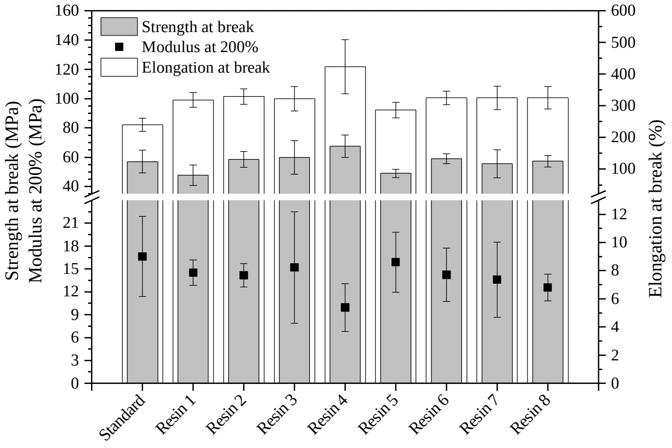

Figure 6. Mechanical properties of the studied formulations.

Conclusion

In this study, we prepared and assessed the influence of tack resins on the adhesion properties of elastomeric compounds, as well as developed an easy and reproducible methodology to determine tack values in the materials studied. The tack resins showed different behaviors, according to their chemical structure and functional groups, while the phenol-formaldehyde-derived resins showed satisfactory values indicating affinity with the matrix, except for the resole-type resin. Coumarone and rosin resins showed consistent values for the evaluated property, indicating compatibility with the elastomeric matrix. When compared with the tack value found for the standard formulation, higher values were obtained for formulations with Resins 1 and 6 (94.6 % and 82.4 % of increase, respectively). However, it was observed that the standard deviation values of the sample with Resin 1 were considerably high; therefore, additional tests must be carried out before confirming this result. The lowest value was presented by formulation with Resin 4 (10.7 % of reduction). In conclusion, the methodology developed proved to be efficient for obtaining the results, being simple to apply and reproducible.

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Authors Contribution

A. Moresco: Methodology, Validation, Formal Analysis, Writing-Original Draft, Writing-Review & Editing; N. B. Guerra: Writing-Original Draft, Writing-Review & Editing; M. Giovanela: Writing-Original Draft, Writing-Review & Editing; S. Moresco: Conceptualization, Methodology, Formal Analysis, Writing-Original Draft, Writing-Review & Editing; J. S. Crespo: Conceptualization, Writing-Original Draft, Writing-Review & Editing, Supervision, Project Administration, Funding Acquisition. All authors have approved the final version of the manuscript.

Conflicts of Interest

The authors have declare no conflicts of interest.

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