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Cosmetic gloves from natural rubber latex for upper limb prostheses: Preparation and physicochemical, mechanical and biological characterization

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A B S T R A C T

Upper limb amputations are frequently the result of traumatic events, often associated with car accidents or incidents involving industrial machinery. The development of prosthetic technologies is essential to provide physical assistance to amputees and offer psychological support. In such circumstances, cosmetic gloves can be utilized to improve personal and social comfort by enhancing visual characteristics similar to those of a biological hand. In this work, we developed formulations based on natural rubber latex (NRL) with antibacterial properties by the incorporation of an additive consisting of zinc oxide coated with silver nanoparticles to manufacture cosmetic gloves designed for upper limb prostheses. The dispersion efficiency of the additive was investigated by microscopic techniques. Tensile and tear strength tests, Shore A hardness measurements, swelling indices, hydrophobicity and inhibition of bacterial growth were also conducted to ascertain the suitability and adaptability of the developed material for integration into the daily activities of patients. Moreover, processing parameters for the immersion molding production of cosmetic gloves were assessed and validated, highlighting the crucial relationship between manufacturing techniques and material properties. Overall, the results indicated that the combination of the additive with a high crosslinking density of NRL effectively improved the material mechanical properties, resulting in a tensile strength of up to 20.7 MPa and a tear strength of approximately 40 kN m^{-1}. In addition, and according to the antibacterial tests, the gloves exhibited bacteriostatic effects against *Staphylococcus aureus* and bactericidal effects against *Escherichia coli*. As a conclusion, the dataset indicates that the manufactured cosmetic gloves can be used to covering upper limb prostheses. Additionally, it underscores the considerable potential for the cosmetic glove manufacturing industry through the use of appropriate formulations and processing techniques for NRL.

1. Introduction

The hand is at the distal end of the upper limb, which allows the possibility of execution activities that need to reach distant points, and due to the arm joints, such as the elbow and the shoulder, this movement occurs in different positions and orientations. The absence of this member has consequences not only for physical abilities but also for the emotional health of individuals [1,2]. According to the United Nations,

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more than 1 billion people worldwide currently need assistive technology, and studies indicate that this number should double in the next decade due to the aging of the population [3]. Among upper limb prostheses, myoelectric prostheses are widely used due to their physiologically natural operation and the lower force required for movement [2]. Despite the various functionalities promoted by their use, high abandonment rates of prostheses are observed [4].

The absence of anthropomorphism is one of the main discomfort factors related to the use of hand prostheses, so covering them with cosmetic gloves is an alternative to promoting a biological appearance. A cosmetic glove is defined as a covering that can be added over a prosthetic hand. The use of these gloves, which have an appearance and color similar to those of the lost limb, is important for restoring the confidence and social comfort of patients [5]. In addition to visual applications, cosmetic gloves also promote increased friction between the prosthesis and objects, as the materials used in their production have a higher coefficient of friction, increasing the safety of handling daily items [6]. Another benefit of using gloves is their ability to protect the internal mechanisms of the prosthesis against external agents such as dirt, humidity, corrosion or even water [7].

Considering these aspects, natural rubber latex (NRL) is an interesting material for use in this area because it is an elastomer with a combination of unique properties, such as mechanical strength, elasticity, softness, and resilience. These varied properties make NRL a material for use in a wide scope of applications, ranging from small sealing components to gigantic machine tires, which are present in human life in several ways [8]. NRL also has great potential for use as a biomaterial in applications such as the production of catheters, bioactive dressings, and surgical gloves [9–12]. NRL is a colloidal dispersion in which rubber particles are surrounded by proteins, lipids and extensive chains of fatty acids, thus conferring a negative charge and the consequent colloidal stability of the medium [8–10,13]. Although synthetic isoprene rubbers are widely developed and applied, their behavior and properties differ from those of natural rubbers, confirming the importance of polymers obtained from rubber trees [8,14,15]. The development of cosmetic gloves with greater resistance, flexibility and durability is essential for reducing the frequency of change, as formulations based on NRL are efficient and low-cost alternatives [16]. Moreover, the use of NRL has been prioritized not only for its attractive mechanical and chemical properties for a wide range of applications but also due to its renewable source [17,18].

The incorporation of additives with antimicrobial properties into cosmetic gloves is an alternative to the absence of touch for the perception of dirt on the glove and to the difficulty of cleaning them with conventional methods, such as soap and water [19]. Silver is an example of a material that has been widely used to prevent the growth and proliferation of pathogens [20,21], and silver nanoparticles (AgNPs) have particularly been used in a wide range of products in the food industry or in the covering of hospital instruments. Interestingly, in addition to its antibacterial properties, many studies have demonstrated its effectiveness against certain protozoa, fungi and viruses [12,22–24]. Like AgNPs, metal oxides are also recognized as antimicrobial agents and exhibit minimal toxicity to humans [22]. Among the main known metallic oxides, zinc oxide (ZnO) is one of the most attractive because it can also act by damaging the surface of bacterial cells [25,26]. Additionally, ZnO is considered nontoxic to humans and is commonly used in medical devices [27]. The incorporation of ZnO in natural polymers, such as NRL, is commonly carried out during the vulcanization process; however, it can also act against bacteria such as *Staphylococcus aureus* (*S. aureus*) and *Escherichia coli* (*E. coli*) [25]. With the aim of enhancing the antibacterial properties of NRL films for subsequent application in the manufacturing of cosmetic gloves, we specifically chose to incorporate an additive consisting of ZnO coated with AgNPs into various formulations for this purpose. The selection of an additive with this particular composition was based on the antibacterial activity of the individual components, as well as their potential for effective dispersion within the latex matrix.

Considering all these aspects, the objective of this study was to develop NRL-based compounds employing the mentioned additive to manufacture cosmetic gloves designed for upper limb prostheses, and subsequently characterize them to determine the most suitable formulation for the intended application. Additionally, this research also aimed to evaluate different production parameters of cosmetic gloves using an immersion molding process.

2. Materials and methods

2.1. Materials

Centrifuged NRL (60 *wt*% dry rubber in ammonia solution) was purchased from RubberSul (Estância Velha – RS, Brazil). The components used for the preparation of the formulations were ZnO (Votorantim S/A, São Paulo – SP, Brazil), zinc ethylphenyl dithiocarbamate (ZEPC) vulcanization accelerator (Yasho Industries, Mumbai, India), sulfur (Basile Química Indústria Comércio, São Paulo – SP, Brazil) and phenol-styrene antioxidant (Proquitec Indústria de Produtos Químicas, Vargem Grande Paulista – SP, Brazil), which were provided by the Vipal Borrachas (Nova Prata – RS, Brazil). Potassium hydroxide was purchased from Dinâmica Química Contemporânea Ltda. (Indaiatuba – SP, Brazil), sodium lauryl sulfate was manufactured by CRQ Produtos Químicos (Diadema – SP, Brazil), poly(acrylic acid) was produced from Polysciences, Inc. (Warrington – PA, USA), and the surfactant Tween 80 (polyoxyethylene sorbitan monolaurate) was obtained from Sigma Aldrich (São Paulo, SP – Brazil). The additive was acquired from S^3nano (Florianópolis – SC, Brazil). A brown pigment fabricated by Salisil Química Ltda. (Porto Alegre – RS, Brazil) was also used. Ethanol (Dinâmica Química Contemporânea Ltda., Indaiatuba – SP, Brazil), calcium chloride dihydrate (Merck S/A, São Paulo – SP, Brazil), and calcium carbonate (Dinâmica Química Contemporânea Ltda., Indaiatuba – SP, Brazil) were used in the coagulant dispersion. The chemicals used were all of analytical grade.

2.2. Additive characterization

The silver content was quantified by inductively coupled plasma–optical emission spectrometry (ICP–OES) using a Thermo Scientific ICP-OES ICAP 7000 spectrometer. The analyses were carried out according to method 3120-B described in the Standard Methods for Examination of Water and Wastewater (2017).

The surface morphology was analyzed using a Tescon MIRA 3 LMH scanning electron microscope (SEM). The source voltage used was 15 kV. The composition was evaluated by energy dispersive spectroscopy (EDS) using the same equipment. The size distribution of both ZnO and AgNPs was assessed using transmission electron microscopy (TEM, model JEM 1011 JEOL), operating at an acceleration voltage of 80 kV. Additionally, the particle size was determined using ImageTool software by counting approximately 100 particles.

2.3. NRL formulations

The composition of the seven formulations developed in this study is detailed in Table 1. Component quantities were determined based on literature and expressed in parts per hundred of rubber (phr) [16,25,28,29].

After mixing the components, the mixtures were dispensed into 140 mm diameter Petri dishes for subsequent drying. Once the drying process was complete, the NRL films were removed from the dishes using distilled water heated to 60 ± 2 °C, followed by vulcanization at 110 °C for 45 min. Subsequently, the films were cut and analyzed through different techniques.

L1, L2, L3, and L4 formulations were evaluated to ensure they possess appropriate mechanical and thermal properties for

Table 1
Composition of the developed formulations.

	Pure latex (phr)	L1 (phr)	L2 (phr)	L3 (phr)	L4 (phr)	L5 (phr)	Vulcanized latex (phr)
NRL[1]	100.0	100.0	100.0	100.0	100.0	100.0	100.0
Additive[2]	–	0.07	0.13	0.25	0.50	1.50	–
KOH[3]	–	0.03	0.05	0.10	0.20	0.20	0.20
Surfactant[4]	–	0.07	0.13	0.25	0.50	0.50	0.50
Antioxidant[5]	–	0.07	0.13	0.25	0.50	0.50	0.50
ZnO[6]	–	0.02	0.05	0.10	0.25	0.00	0.75
ZEPC[7]	–	0.07	0.13	0.25	0.50	0.50	0.50
S[8]	–	0.09	0.18	0.35	0.75	0.75	0.75

[1] natural rubber latex.
[2] compound consisting of zinc oxide coated with silver nanoparticles.
[3] potassium hydroxide.
[4] sodium lauryl sulfate.
[5] phenol-styrene antioxidant.
[6] zinc oxide activator.
[7] zinc ethylphenyl dithiocarbamate vulcanization accelerator.
[8] sulfur.

manufacturing cosmetic gloves. However, to confirm the effectiveness of the antibacterial additive, two additional formulations were prepared: one with a high quantity of the additive (L5) and one without the additive (vulcanized latex).

Regarding the L4, L5, and vulcanized latex formulations, different quantities of the additive were evaluated. Since this compound consists of ZnO coated with AgNPs, the zinc oxide activator (indicated by the number 6 in Table 1) was not added in the L5 formulation. In the vulcanized latex formulation, by other hand, the ZnO content corresponds to the sum of the additive and zinc oxide activator added to the L4 formulation. These adjustments were made to solely assess the antimicrobial efficacy of the AgNPs. This approach was used to prevent any potential interference from ZnO, which also has antimicrobial properties, thus ensuring the accuracy of the results.

2.4. NRL formulations characterization

The films resulting from formulations were characterized by SEM coupled with energy dispersive spectroscopy (EDS) using a Tescon electron microscope (MIRA 3 LMH), with a source voltage of 15 kV. Before visualization, the samples were sputter-coated with gold.

Hardness was measured with a Shore A hardness tester (Mainard, model M-709) according to ASTM D2240. As hardness is an indicator of crosslink density, the optimal vulcanization time (45 min) of the NRL films was determined based on the constant Shore A hardness. The NRL films were cut into dumbbells, and the tensile strength was measured according to ISO 37:2017(E) using a universal EMIC testing machine with a maximum capacity of 30 kN and a speed of 500 mm min^{-1}, where an increasing uniaxial load was applied to the sample. The tear resistance of the samples was assessed following the ASTM D624–00 standard. In this procedure, five specimens were subjected to longitudinal force until tearing occurred, utilizing a universal EMIC testing machine. All mechanical tests were conducted in quintuplicate.

To determine the resistance of the NRL films when exposed to solvents commonly used for hygiene, swelling index tests were performed employing adaptations of the ASTM D3616-95 standard. The analyses were conducted using two different everyday solvents: ethanol 70 % (*v/v*) and water with neutral soap (2.5 % (*v/v*), pH ~ 6.5). The samples were allowed to contact the solvent for 18 or 40 h, and each test was replicated three times.

A thermogravimetric analyzer (Shimadzu equipment, model TGA-50) was used to study the thermal stability of the NRL filmset temperatures ranging from 25 to 600 °C at a heating rate of 10 °C min^{-1} under a nitrogen flow of 50 mL min^{-1}.

The hydrophilicity of the NRL films with two different concentrations of silver was investigated through the evaluation of contact angles. For this, a drop of distilled water was placed on the surface of the samples at three different points on the specimen. Ten contact angle measurements were performed for each drop inserted with the aid of Phoenix equipment, model 150. The test was performed in triplicate.

To ensure the production of gloves with superior durability, accelerated aging in an oven was conducted on select NRL formulations. This process was undertaken to facilitate subsequent assessment of the mechanical properties. The samples were kept in an oven with air circulation at 55 °C for 20, 40 or 80 days, which corresponded to a shelf period of 6 months, 1 year or 2 years, respectively [30].

2.5. Antibacterial tests

For the antibacterial tests, the microorganisms *E. coli* and *S. aureus* were employed. Each microorganism was evaluated using diffusion test in agar with Müller–Hinton (MH) broth in both semisolid and liquid media. Bacterial cultures were transferred to a phosphate-buffered saline (PBS) solution and adjusted for turbidity with a 0.5 McFarland scale. Subsequently, the absorbance was spectrophotometrically measured between 0.1 and 0.5 at 600 nm, which indicates an approximate concentration of 1.0×10^8 colony-forming units per milliliter (CFU mL^{-1}). The suspension was subsequently diluted at a ratio of 1:100 to achieve a concentration of 1.0×10^6 CFU mL^{-1} for incorporation into the agar diffusion test culture medium. Additionally, the solution was further diluted at a ratio of 1:100 to attain a concentration of 1.0×10^4 CFU mL^{-1} for cultivation in MH broth. In the agar diffusion test, plates were individually inoculated with *E. coli* and *S. aureus*. The samples were subsequently placed on the respective culture media and incubated during 24 h at 37 ± 1 °C.

For analysis in liquid medium, 1.0 mL of MH broth and 50 μL of the solution containing 1.0×10^4 CFU mL^{-1} were added to each well. The NRL films were then placed in the wells and kept in an oven at 37 ± 1 °C for 24 h. After this period, 60 μL of 0.01 % (*w/v*) resazurin was added to each well containing MH broth and placed in a shaker under stirring at 15 rpm and 37 °C for 1 h. Finally, a bluish medium, similar to the negative control without inoculum, was reinoculated into semisolid medium at 37±1 °C for 24 h. This evaluation was performed in liquid medium followed by inoculation in agar to determine whether the samples exhibited bacteriostatic activity, preventing bacterial growth and reproduction, or whether the NRL films demonstrated bactericidal properties by killing the bacteria [31].

2.6. Glove preparation: processing and immersion molding

To fabricate NRL gloves by the immersion process, molds of dental plaster and acrylonitrile-butadiene-styrene (ABS) were used. The molds were initially cleaned with water with neutral soap and then dried in an oven (Fig. 1).

Then, the molds were immersed in a coagulant dispersion (see Table 2) for 1.5 min and returned to the oven at 60 °C until completely dry. The coagulant solution enhances the coagulation capacity of NRL, leading to the production of thicker gloves. Its application is essential for enhancing mechanical properties [32]. Different immersion times in the formulated latex were evaluated (1, 2, 5 and 10 min). The NRL gloves was also dried in an oven (60 °C) for 6 h. After drying, the gloves were demolded from the molds with the aid of a release solution (10% (*w/v*) calcium carbonate). The gloves were then leached for 3 min in distilled water heated to 60 ± 2 °C to eliminate excess chemicals from the surface. Vulcanization was carried out at a temperature of 110 °C for 45 min until a constant hardness was achieved.

Table 2
Composition of the coagulant dispersion.

Chemical	Concentration (% *w/w*)
Distilled water	32.5
Ethanol 70 % (*v/v*)	32.5
Calcium chloride	32.6
Calcium carbonate	1.3
Bentonite	1.1

3. Results and discussion

3.1. Characterization of the additive

The ICP–OES results indicated that the additive contains 0.024 % (*w/w*) of silver. This concentration is suitable for applications in NRL to promote antibacterial properties since very low concentrations of this element (approximately 1 ppm or 0.0001 %) can already be considered effective against bacteria [31,33].

The morphology of the additive, along with its chemical composition, is depicted in Fig. 2. As can be observed, SEM micrograph at higher magnification (Fig. 2b) revealed distinct hexagonal structures of ZnO (highlighted by red arrows) with varying sizes [27,34]. Regarding the EDS analysis, the spectrum highlights the presence of Zn, O, Na, C, Si and Ca (Fig. 2c). These signals were expected since, according to the manufacturer, the additive is stabilized with long-chain polymers and silica. The absence of Ag in the EDS spectrum is attributed to its low concentration in the additive, as confirmed by the ICP-OES analysis, and by the limitations of the technique.

The choice of this additive was motivated by the necessity for proper dispersion within the NRL matrix, as agglomerations could generate possible points of mechanical fracture in the material. Thus, as ZnO has good dispersion in the NRL, it can carry AgNPs and facilitate their efficient dispersion. Characterization by TEM analysis revealed that AgNPs were randomly dispersed on the surface of ZnO particles (Fig. 3a) without noticeable agglomeration (Fig. 3b) (highlighted by red arrows). Furthermore, these AgNPs exhibited a spherical morphology with an average size ranging between 4 and 15 nm (Fig. 3c). Similar results have been reported by other authors who have also studied AgNPs, corroborating our findings [24,35,36].

3.2. Characterization of the NRL films

3.2.1. SEM-EDS analysis

Fig. 4 presents the chemical composition maps of the NRL films vulcanized with 0.5 phr (L4) and 1.5 phr (L5) of the additive obtained by SEM−EDS. As can be observed, ZnO was dispersed over the entire surface of the NRL films and appears in high concentrations because it was also used as a vulcanization agent. Distinctly, we did not observe Ag distribution because its concentration was lower than the detection limit of the EDS technique. Moreover, satisfactory dispersions of sulfur and zinc, found in ZnO and ZEPC, were also observed, indicating, as reported in the studies of Hu et al. [37], that a good dispersion of vulcanization agents helps to increase crosslinking and consequently improve mechanical properties.

3.2.2. Mechanical and physical properties

Fig. 5 shows the results of the mechanical analyses for the samples with different chemical quantities in the vulcanized NRL films. We observed that as the concentration of the chemicals lowers, the maximum strength gradually decreased. A reduction to 1/2, 1/4 or 1/8 (L3, L2, and L1 formulations, respectively) resulted in a reduction of 55.1, 88.4 and 96.1 %. This behavior was consistent because the chemicals influence the latex vulcanization system, and curing with fewer cross-linkages diminishes the ability of the material to withstand mechanical stress. Notably, the L4 formulation exhibited a tensile

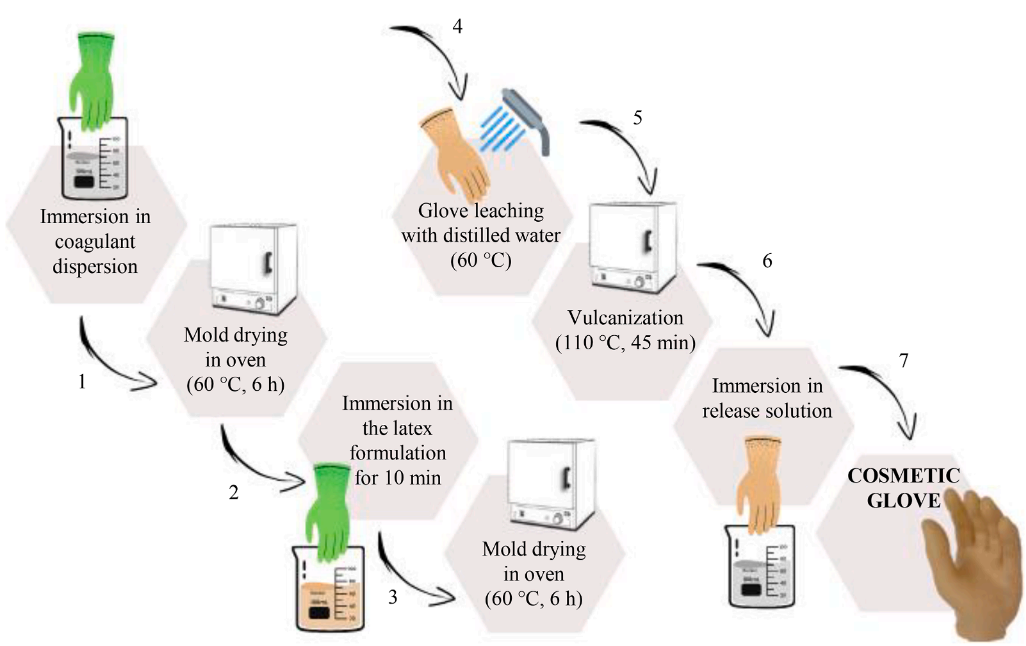

Fig. 1. Schematic representation of the sequential stages involved in the production of NRL gloves through immersion molding.

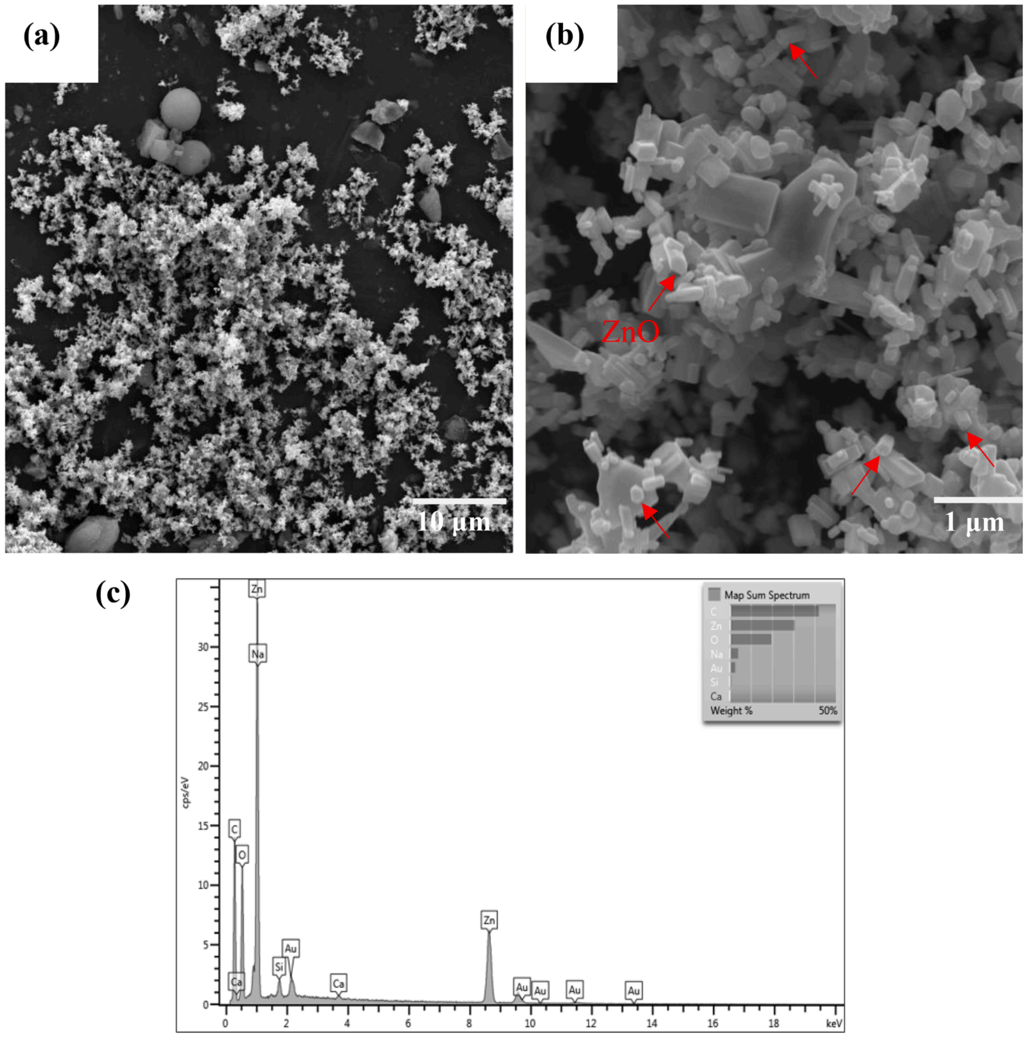

Fig. 2. SEM micrographs at (a) 5,000 × and (b) 50,000 × magnifications, and (c) EDS spectrum of the additive.

strength ~ 20.7 MPa, establishing it as a viable alternative. Likewise, Li et al. [38] developed antimicrobial materials with a NR matrix containing silver and graphene oxide for glove production, achieving a tensile strength of up to 21.2 MPa.

As in tensile strength tests, diminishing the concentration of chemicals causes a decrease in the ability of the material to elongate before breaking. The L4 formulation does not have the average of its maximum elongation at break reported in Fig. 5 since the universal test equipment track ended before the specimens were broken. The results indicated that the chemical concentrations used for this formulation were the most common for the other characterizations and consequent production of cosmetic gloves.

Studies on the development of materials to produce gloves have also assessed the maximum elongation at break. Yabuki et al. [39] evaluated a thermoplastic elastomer, achieving a maximum elongation of 1300 %. Conversely, Li et al. [38] developed gloves with an NRL matrix, reporting elongations of less than 600 %. Comparatively, our work achieved a maximum elongation at break of approximately 3000 %, indicating that the developed material is very promising for the manufacture of cosmetic gloves.

To corroborate the results obtained from the analysis of tensile strength with maximum elongation at break, tear resistance tests were also performed on NRL films with reduced chemical concentrations. Once again, a notable decrease in tear strength of approximately 31.2 % was noted when the quantity of chemicals in the L4 formulation was halved (L3 formulation). This percentage increased to 70.3 % when the amount was reduced by one quarter (L2 formulation) and further to 85.6 % when the amount was reduced by one eighth (L1 formulation). The L4 formulation showed tear resistance of 40 kN m^{-1}, surpassing the values reported by other authors for cosmetic gloves made of thermoplastic elastomer and silicone rubber [39].

Similar tear resistance results, between 23 and 37 kN m^{-1}, were also achieved by Hu et al. [37] after the incorporation of modified cellulose nanocrystals in a vulcanized NR matrix. These authors concluded that their cosmetic gloves adequate mechanical properties, even without the incorporation of reinforcing fillers. The chemicals employed in latex vulcanization had a greater influence on the hardness than on the tensile and tear strength of the produced NRL films.

As illustrated in Fig. 5, with a low concentration of chemicals, such as in the L1 formulation, the vulcanized NRL displayed an increase of 16.5 % in resistance to localized deformation compared to that of pure latex. Similar results were observed for the L4 formulation and in the study of Hu et al. [37], in which vulcanized rubber reinforced with nanocellulose exhibited a Shore A hardness ranging between 35 and 45. Consistent with the results of the mechanical properties, an increase in chemical concentration results in increase material hardness, indicating that the L4 formulation are the most appropriate for the production of the gloves.

3.2.3. *Thermal stability*

The thermal stability of the NRL films was evaluated via

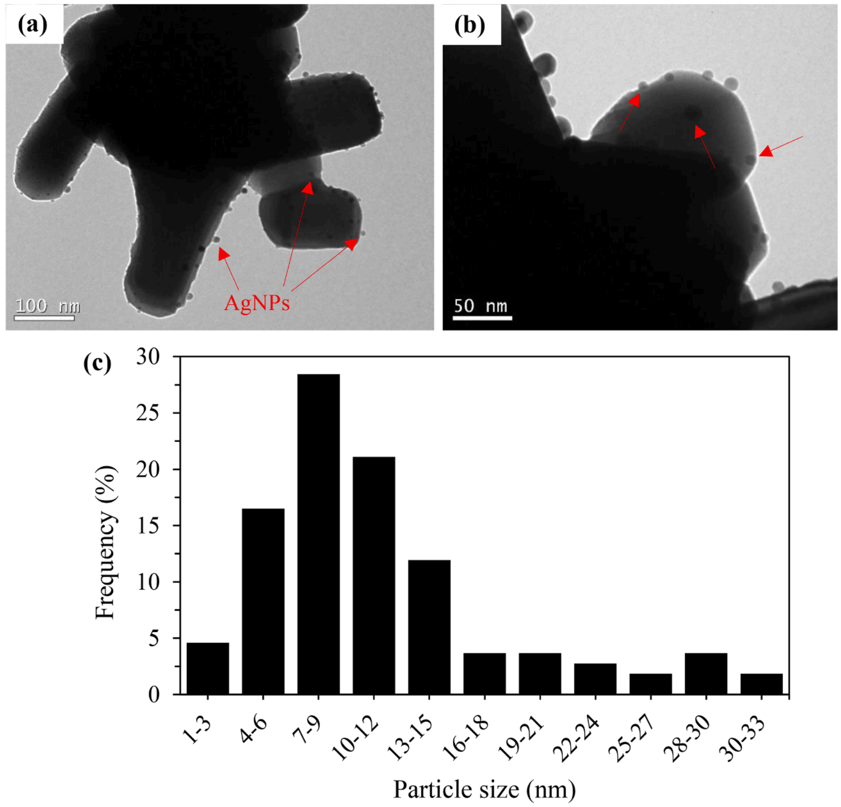

Fig. 3. TEM micrographs (a and b) of the additive and (c) size distribution diagram of the AgNPs.

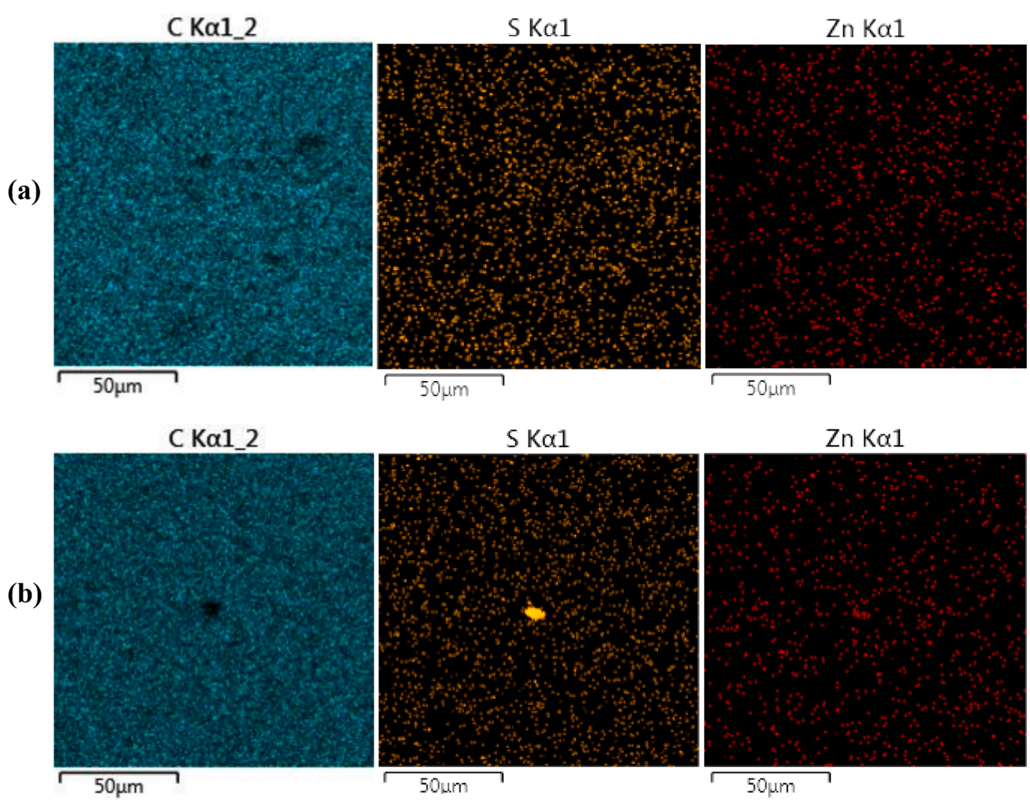

Fig. 4. Chemical composition maps of the NRL films vulcanized with (a) 0.5 phr (L4) and (b) 1.5 phr (L5) of additive obtained by SEM–EDS.

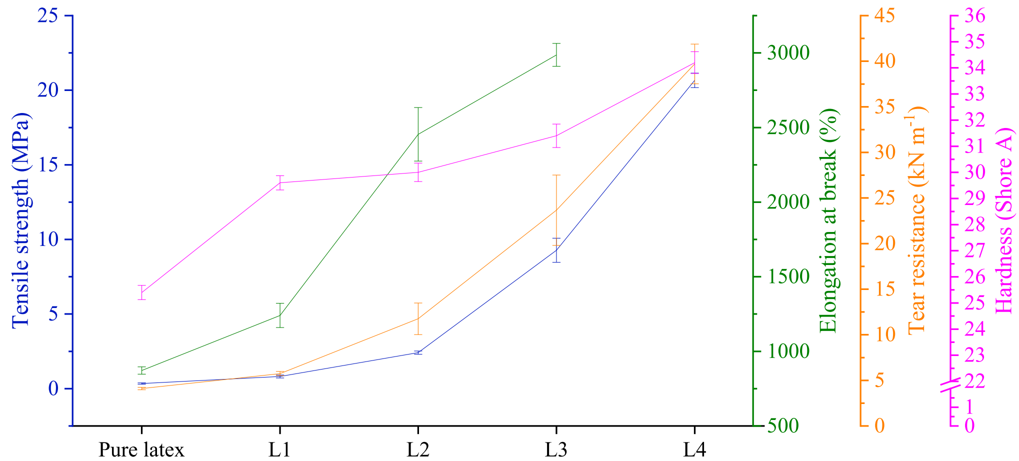

Fig. 5. Tensile strength, elongation at break, tear strength and Shore A hardness analyses of pure latex and NRL films.

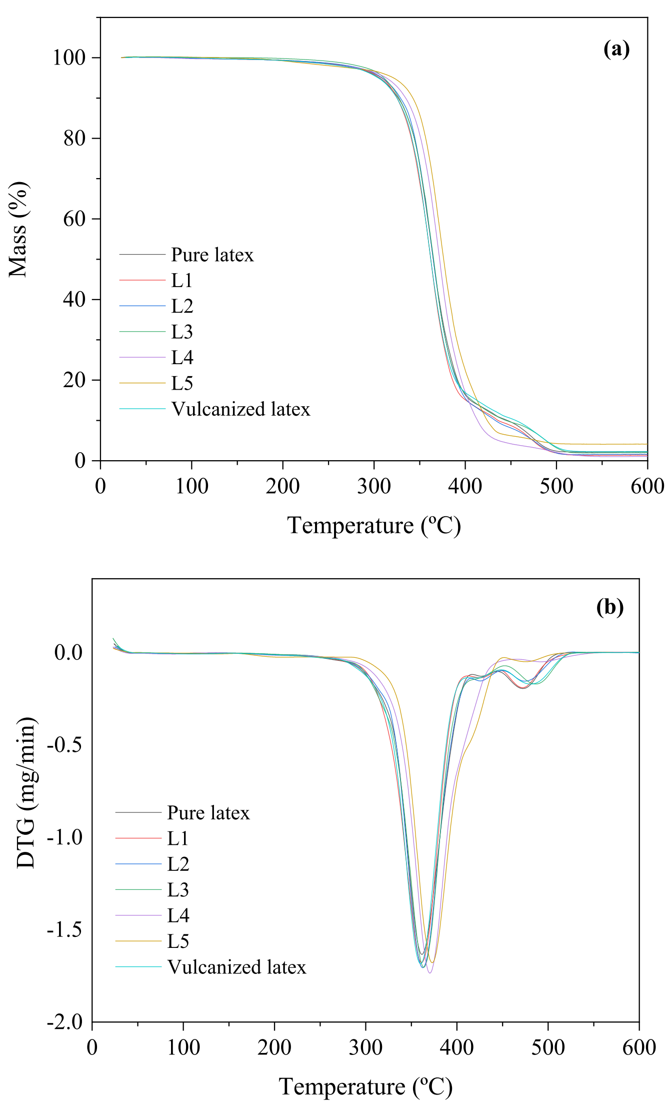

Fig. 6. Thermogravimetric analysis of the pure latex, NRL films and vulcanized latex: (a) TGA curves; (b) DTG curves.

thermogravimetric analysis (Fig. 6). The findings revealed that the onset of material degradation occurs at a temperature nearing 360 °C. The thermal decomposition of NRL films can start within the range of 200–500 °C or may occur in two distinct steps, extending from 200 to 500 °C and from 500 to 700 °C [40]. It is known that some of the organic components existing in the rubber matrix, such as polyisoprene molecules, proteins, aldehydes, ketones, and carboxylic acids, undergo complete degradation at approximately 375 °C. At higher temperatures, highly cross-linked polymeric molecules degrade.

Causin et al. [40] indicate that the thermal degradation and behavior of polymeric materials can be altered at high temperatures due to their formulation and slight variations in additives. It is assumed that the thermal stability of the studied samples was adequate, as temperatures close to 50 °C are sufficient to cause burns on human skin within minutes; therefore, since this material is not subjected to higher temperature conditions, it meets the necessary safety and usability standards.

3.2.4. Swelling index

The analysis of resistance to solvents commonly used in daily cleaning revealed that the NRL films can be effectively used as barriers against water and humidity. This was evidenced by the fact that the swelling observed after both 18 and 40 h was consistently less than 5 % (*w/w*) for all the samples tested, regardless of the solvent used (Fig. 7). Moreover, the low swelling indices indicated that the mechanical resistance of the NRL films was not significantly reduced after contact with the solvents. This suggests that the crosslinks in NRL films were effective at preventing the ingress of solvents, as demonstrated by these results.

3.2.5. Contact angle

Hydrophobicity is a desirable characteristic for the development of a material for application in the coating of myoelectric prostheses since it must also act as a barrier against environmental agents, such as humidity or even water, that may eventually fall on the prosthesis. For contact angle, different concentrations (0.5 and 1.5 phr – L4 and L5 formulations, respectively) of the additive were investigated. This variation was chosen based on the finding that the L4 formulation was the most suitable for producing cosmetic gloves among those assessed in Table 1.

The hydrophobicity of the NRL films was assessed by investigating the contact angles with the incorporation of two different concentrations of the additive (Fig. 8). The vulcanized latex sample has a contact angle of 77.5 ± 0.7°, indicating a hydrophilic nature, although it approaches the hydrophobic range (contact angles greater than 90°) [41–43]. Upon the addition of 0.5 phr of additive, a contact angle of 80.0 ± 0.6° was observed, indicating a slight increase in hydrophobicity. Nevertheless,

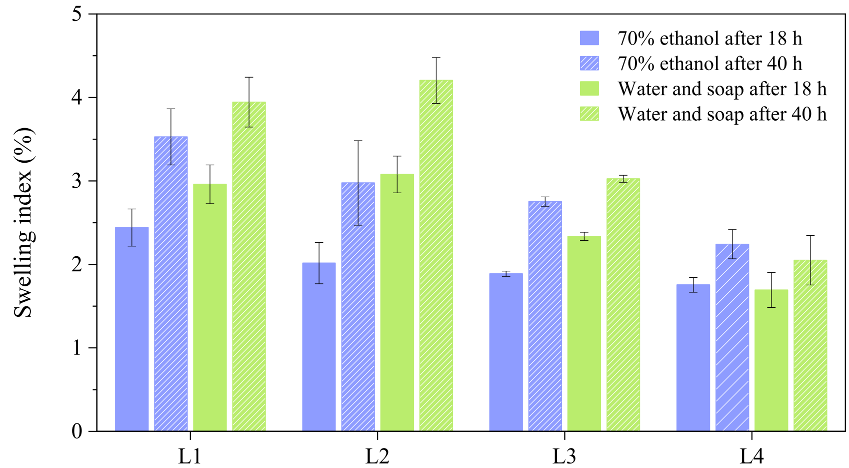

Fig. 7. Swelling indices of the NRL films in daily cleaning solvents.

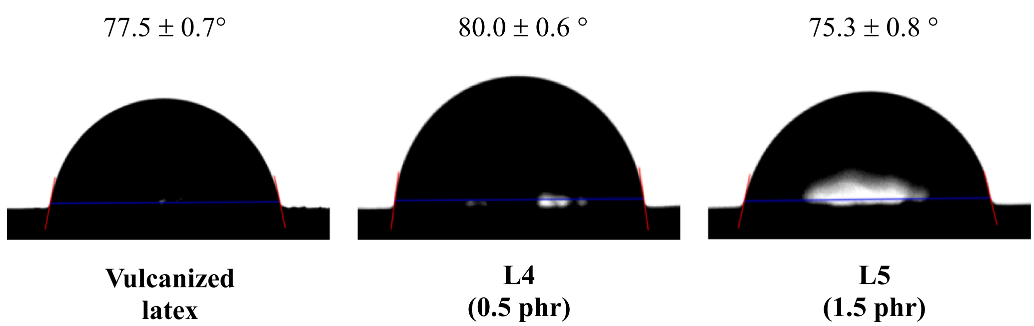

Fig. 8. Contact angles of the vulcanized latex and NRL films containing 0.5 (L4) and 1.5 phr (L5) of additive.

at higher concentrations, such as 1.5 phr of this additive, the hydrophobicity of the NRL film decreases (75.3 ± 0.8°), possibly due to the excess metallic oxide since ZnO structures possess polar terminations that tend to interact more with water molecules, thereby reducing the contact angle [44]. Furthermore, although a near-hydrophobic character is desirable for avoiding contact between water and humidity and the internal mechanism of the prosthesis, the scientific literature indicates that hydrophilic surfaces contribute to reducing the fouling of microorganisms, which is a desirable characteristic of cosmetic gloves [33].

3.2.6. Shelf life – accelerated aging

Fig. 9a shows the assessment of the tensile strength of the aged NRL films, which indicates that the maximum tensile strength decreases consistently across all the materials as the aging time increases. However, the decrease in tensile strength for each material was not uniformly pronounced, suggesting efficient cross-linking of the NRL chains and slow degradation. As reported by Rattanawongkhae et al. [21], the presence of AgNPs could hinder the mechanical performance of NRL films since a lower resistance was observed in all our materials that used silver. Moreover, this phenomenon may be attributed to the NPs acting to initiate a fracture in the material if they fail to interact effectively with the NRL matrix.

Contrary to these findings, our study did not observe such behavior. With respect to both the L4 and L5 formulations, the tensile strength exceeded that of the NRL film without the additive. This difference may be attributed to the presence of ZnO in the additive, which contains smaller particles, ranging between 300 and 500 nm, than ZnO activator.

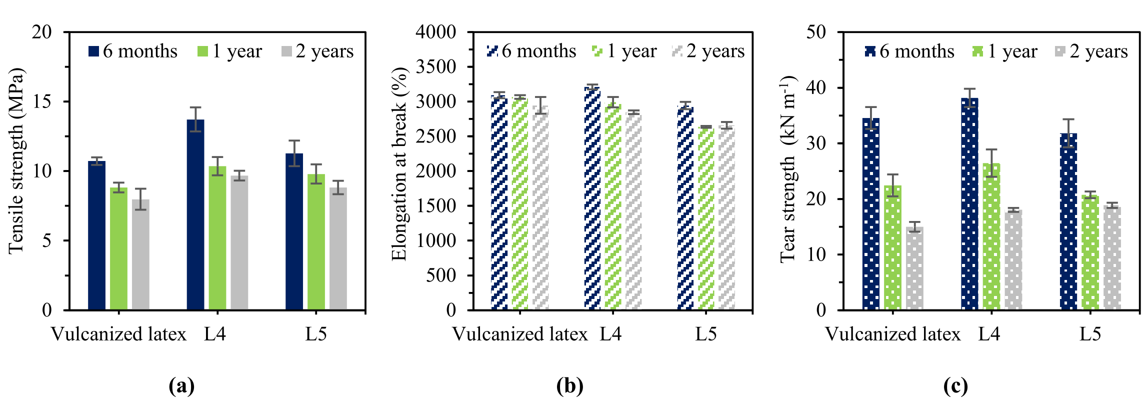

Fig. 9. (a) Tensile strength, (b) elongation at break, and (c) tear strength results for the vulcanized latex and NRL films containing 0.5 (L4) and 1.5 phr (L5) of additive after accelerated aging.

Nanometric ZnO has demonstrated favorable interactions with vulcanization accelerator, thereby enhancing the efficiency of the process [45].

When contrasting these two formulations, L5 exhibited a lower tensile strength, likely because the excess additive was not fully incorporated into the matrix. Additionally, for the L4 formulation the maximum tensile strength before accelerated aging (Fig. 5), revealed a reduction in tensile strength of 43.8 % after 6 months, 50.0 % after 1 year, and 53.2 % at the end of 2 years. Biermann [7] reported that the minimum strength stress for cosmetic gloves was 850 psi (approximately 5.9 MPa). Therefore, despite the significant reduction in this mechanical property, the tensile strength of the L4 formulation remains relatively high, suggesting its viability for application in the production of cosmetic gloves.

In contrast to the tensile strength data, there is no maximum elongation at break data available for the L4 formulation before accelerated aging since the equipment reached its limit before the samples of this formulation could break. Thus, comparisons between different aging times are feasible. Fig. 9b illustrates that, across evaluated materials, even after aging for 2 years, significant flexibility can be sustained in cosmetic gloves, potentially reducing the need for frequent replacements. Furthermore, a minimum elongation requirement of 850 % is required for materials used in cosmetic glove production [7], indicating that all NRL films meet this demand.

Among the mechanical properties examined, tear strength was the property that was most influenced by aging time (Fig. 9c). After 6 months, the L4 formulation resulted in a 4.5 % reduction compared to the preaccelerated aging result (Fig. 5). After 1 year, the same formulation had a 44 % reduction, and at the end of 2 years, it had decreased by 55 %. Nevertheless, even after 2 years, the tear strength remains satisfactory, approximately 18 kN m^{-1}. Yabuki et al. [46] noted that Japanese records suggest that cosmetic gloves are typically replaced every two years; hence, the NRL film must be sufficiently resistant to endure at least this duration. Thus, after the accelerated aging tests, it can be concluded that the evaluated NRL films are durable for up to 2 years for use as cosmetic gloves.

3.2.7. Antibacterial analysis

Many studies have shown the antibacterial efficacy of AgNPs against Gram-negative bacteria, such as *E. coli*, and against Gram-positive bacteria, such as *S. aureus* [24,38,47]. In the agar diffusion tests performed with *E. coli* (Fig. 10a), no inhibition halo was observed in the NRL films lacking the additive. Conversely, in both samples containing 0.5 and 1.5 phr of the additive, an inhibition halo was evident, indicating the antibacterial activity. The darker zones surrounding the samples represent the protection zones. The diameters of the halos formed with both concentrations of the additive were similar, suggesting that large quantities of the additive are unnecessary for observing activity against Gram-negative bacteria such as *E. coli*. Similar behavior was also observed for the tests against *S. aureus* (Fig. 10b), confirming that the incorporation of additive inhibits the growth of Gram-positive bacteria [38]. According to the data reported in other studies, the larger zones of inhibition in these bacteria are attributed to thicker cell walls, which contain more peptidoglycan layers. These layers are more susceptible to damage by AgNPs, leading to enhanced inhibition [24,48].

To determine whether the NRL films exhibited bactericidal or bacteriostatic properties, tests were conducted in liquid medium (MH broth), followed by inoculation of this medium onto Petri plates containing agar. In the evaluation involving the bacteria *E. coli* (Fig. 11a), NRL films containing AgNPs demonstrated bactericidal properties, as evidenced by the appearance of a blue color similar to that of the negative control following the addition of resazurin dye. Moreover, the inoculation of NRL films from liquid media onto solid agar media resulted in minimal colony growth. However, in this experiment, we observed that higher concentrations of the additive led to more pronounced bactericidal effects. While the sample containing only NRL exhibited total bacterial growth, the NRL film with 0.5 phr (L4) of the additive had only four colonies, and no colonies were identifiable in the NRL film with 1.5 phr (L5) of the same additive. Despite the presence of a slightly violet color in the resazurin test, when the evaluation was conducted with *S. aureus*, all materials exhibited bacteriostatic properties. Following the inoculation of the liquid media onto solid agar media, complete microorganism development occurred (Fig. 11b).

Finally, we confirmed that the NRL films containing silver presented enhanced antimicrobial activity against *E. coli*. Similarly, Mam and Dangtungee [49] assessed the antimicrobial activity of NRL foams containing AgNPs and reported that these materials were more effective against a Gram-negative strain (43 %) than against a Gram-positive strain (25 %). This behavior may be attributed to the stronger electrostatic attraction between the cell membranes of negatively charged bacteria and positively charged AgNPs. From the antibacterial test results, we observed that NRL films containing the additive can be effectively used in the production of cosmetic gloves to prevent microbial growth. This approach is particularly crucial because conventional hand

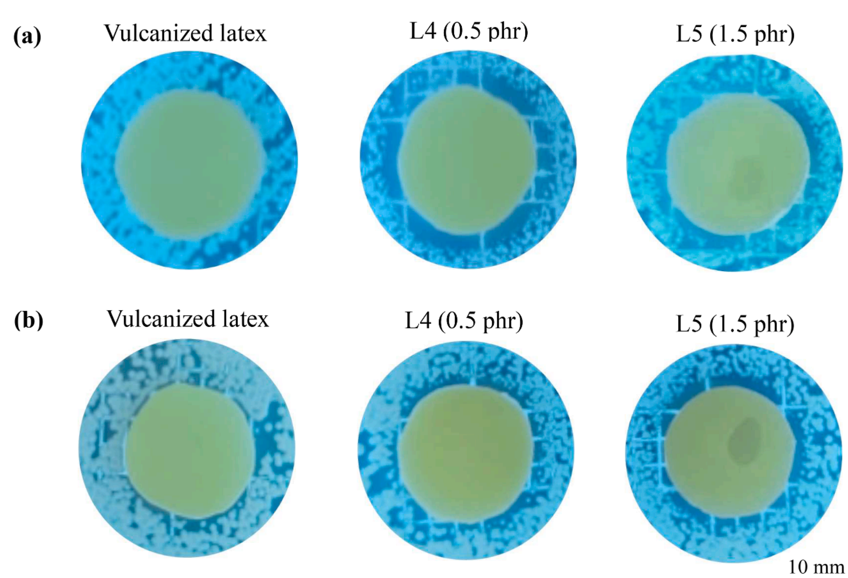

Fig. 10. Growth inhibition tests with vulcanized latex and NRL films containing 0.5 (L4) and 1.5 phr (L5) of additive against (a) *E. coli* and (b) *S. aureus*.

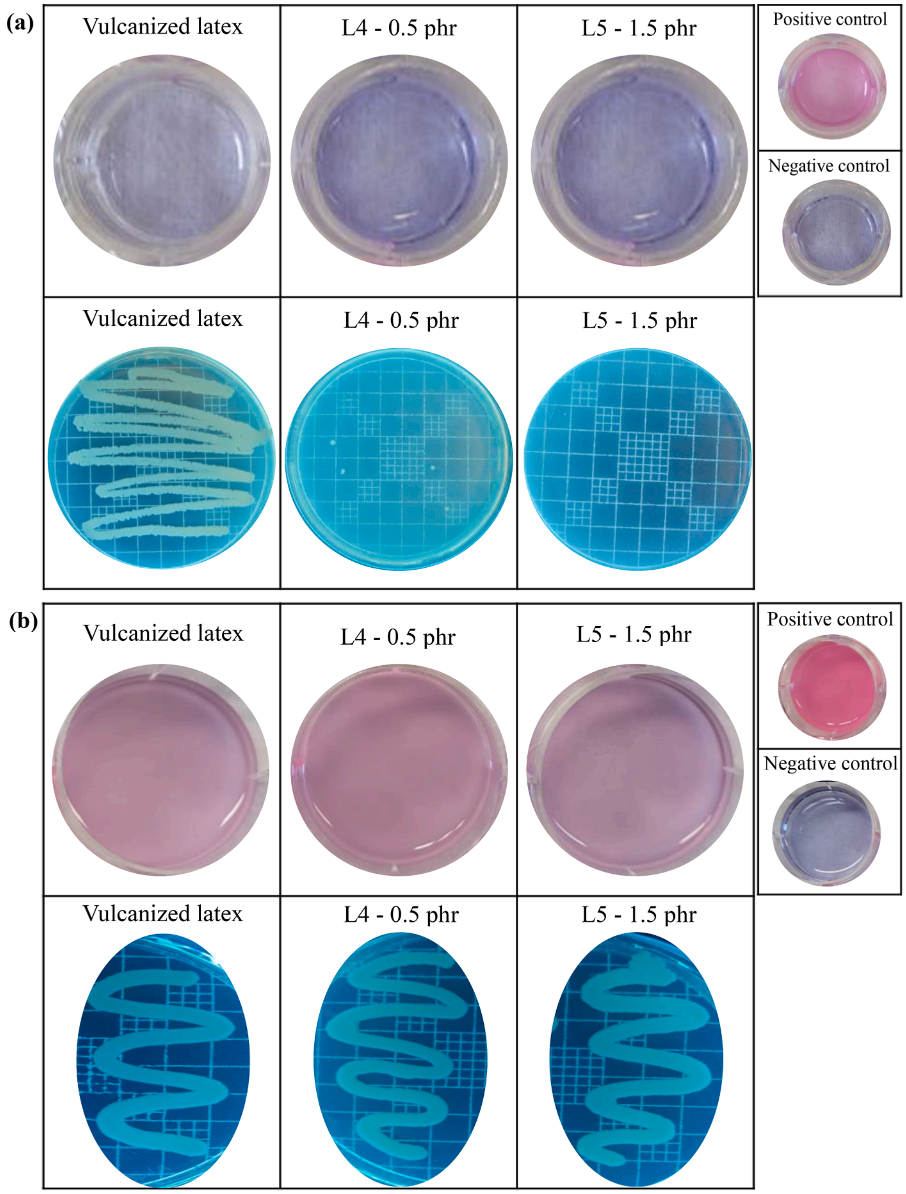

Fig. 11. Inhibition of bacterial growth of (a) *E. coli* and (b) *S. aureus* promoted by vulcanized latex and NRL films containing 0.5 (L4) and 1.5 phr (L5) of additive in liquid media and agar diffusion tests.

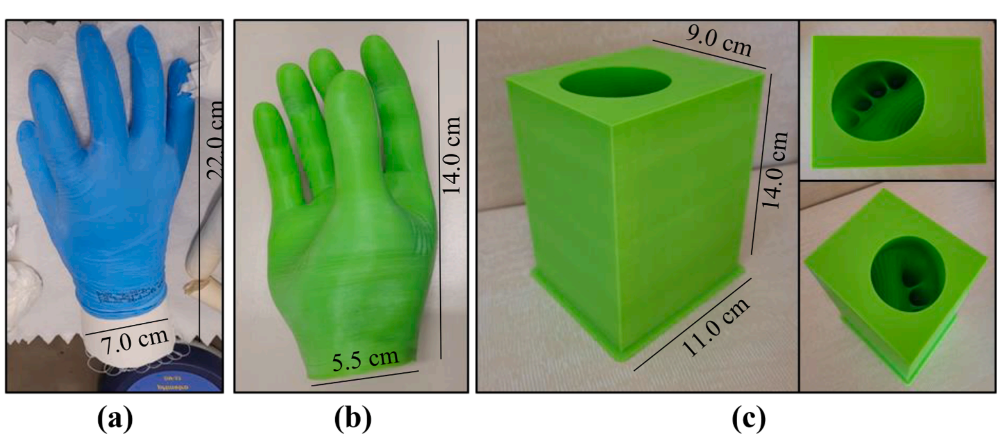

Fig. 12. Photographs of the molds made of different materials and formats evaluated for use in producing gloves: (a) positive plaster cast covered with a nitrile glove; (b) positive ABS mold; (c) negative ABS mold.

hygiene methods involving soap and water cannot be applied directly to gloves or upper limb prostheses.

3.3. Molding processing

The development of cosmetic gloves requires a personalized approach, considering the individual characteristics of amputee patients, such as age, skin color, and hand size, which greatly influence the characteristics of the glove. It is imperative to devise glove production methods that can be readily adjusted by qualified prosthetists to accommodate these unique personal attributes.

In our study, various molds were tested. The initial mold was crafted with a dental plaster base (Fig. 12a) and then coated with a nitrile glove. This coating was essential because the high porosity of the plaster hindered the deposition of the coagulant solution required in this process. To achieve a glove better suited for the formats of claw-type upper limb prostheses, we assessed the use of a new mold (Fig. 12b) fabricated with ABS through 3D printing. This mold offered lower porosity, eliminating the need for nitrile glove coverage. Before employing this mold, others based on ABS (acrylonitrile butadiene styrene) were also produced using a 3D printer. Nevertheless, molds with excessively thin walls experience significant deformation when subjected to drying in an oven at 60 °C and, more prominently, during vulcanization at 110 °C. Additionally, a new ABS mold was employed, which served as a negative control mold, as depicted in Fig. 12c. The objective was to diminish the volume of the NRL required to immerse the mold, as a considerably smaller volume is adequate for coating its inner walls.

After employing the three different molds, several advantages and disadvantages were noted. The plaster mold coated with the nitrile glove was easy to demold and offered sufficient mass for stability during the immersion period. However, plaster is inherently brittle, limiting its reusability. Additionally, a plaster also requires the use of a nitrile glove covering due to its high porosity, which absorbs the coagulant solution, preventing the glove from forming. Conversely, employing a positive ABS mold facilitated a more anthropomorphic thumb position owing to the mold's ability to incorporate finer details during printing. However, the presence of voids within the mold compromises its stability during immersion and renders it susceptible to deformation when exposed to heat in an oven. To address these issues, the mold was filled with a cement material, which was chosen for its suitability for the intended purpose. Additionally, to minimize the amount of NRL needed, casting molding was explored using a negative ABS mold. However, this method significantly hindered the release of the gloves and resulted in the entrapment of air bubbles between the mold and the NRL, compromising the appearance of the glove.

This study conclusively establishes that employing a positive ABS mold represents the most optimal approach. Moreover, this mold is straightforward to prepare and manage. Importantly, compared with plaster molds, gloves with a more anthropomorphic design are produced, as the thumb position in the plaster mold results in an unnatural accumulation of the NRL in the glove during prosthesis movements.

3.4. Preparation of cosmetic gloves

The feasibility of using immersion molding for producing NRL gloves was evaluated through a test involving the L4 formulation (Fig. 13). Latex gloves are typically manufactured for hospital or general protection purposes and are intended to be disposable. Therefore, their thickness does not need to be substantial. This is in contrast to latex gloves with varying nanocellulose contents produced by Blanchard et al. [50], which exhibited thicknesses ranging from 0.18 to 0.36 mm after 40 s of immersion in the NRL formulation. In this study, different immersion times of the mold in the NRL formulation were examined (1, 2, 5 and 10 min), and the corresponding thicknesses of the produced gloves are displayed in Table 3.

The thickness achieved after 10 min of immersion was deemed suitable, with the glove averaging approximately 1.6 mm in thickness. Yabuki et al. [39] obtained variable thicknesses between 1.5 and 5 mm of cosmetic gloves manufactured from a thermoplastic elastomer and silicone from injection molding. As reported in other studies, the crosslinking density of NRL gloves increases with increasing dwell time, and consequently, the mechanical strength of the gloves increases [51]. Gloves manufactured at lower speeds lack structural stability, rendering them unsuitable for the intended application.

Finally, to broaden the range of color options available for the gloves, color tests were conducted using mixtures of dyes typically used in latex-based paints, namely, yellow ochre and brown. Fig. 13 displays a concise palette of colors intended for use in the production of cosmetic gloves. The customization of cosmetic gloves must be executed in the most efficient manner possible, as it serves as a key motivator for their usage and facilitates more comfortable social interaction. Therefore, personalization through both format and color is of utmost importance.

4. Conclusions

In this study, NRL formulations were assessed for their ability to produce cosmetic gloves with suitable mechanical properties which require frequent removal and replacement. Additionally, an additive consisting of ZnO coated with AgNPs was incorporated to enhance the antibacterial properties of the NRL. The use of larger quantities of vulcanization agents, as seen in the L4 formulation, proved to be more effective at attaining a tensile strength exceeding 20 MPa, tear strength of 40 kN m^{-1}, and elongation at break exceeding 3000 %. The NRL films also exhibited thermal stability suitable for everyday tasks. Evaluation

Table 3
Glove thicknesses for different immersion times using the L4 formulation.

Immersion time (min)	Thickness (mm)
1	0.02 ± 0.01
2	0.12 ± 0.04
5	0.74 ± 0.17
10	1.60 ± 0.12

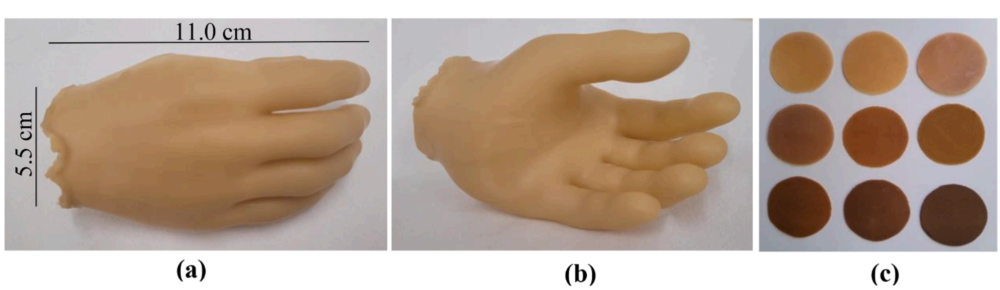

Fig. 13. (a and b) Vulcanized cosmetic gloves made from L4 formulation by an immersion molding process; (c) NRL glove color palette.

of the contact angle and swelling in polar solvents revealed that NRL exhibited near-hydrophobic characteristics and displayed minimal swelling in common solvents such as soap, water, and ethanol 70 % (*v/v*). These attributes are significant for the utility of the material as a prosthetic covering. The developed materials demonstrate a durability of up to 2 years, as determined through tensile and tear strength assessments conducted following accelerated aging of the samples. The antibacterial properties were found to be effective, as the materials containing the additive exhibited bactericidal activity against *E. coli* and bacteriostatic activity against *S. aureus*. In summary, the production of cosmetic gloves demonstrated that the material possesses favorable properties and processing capabilities, making it suitable for use as a coating for upper limb prostheses. Moreover, the NRL formulations can be readily dyed, simplifying the customization process for prosthetists. This customization aspect holds significant importance in restoring the self-esteem of patients who have undergone amputation.

CRediT authorship contribution statement

Danielle Restelatto: Writing – review & editing, Writing – original draft, Validation, Methodology, Investigation, Formal analysis, Data curation, Conceptualization. **Jordana Bortoluz:** Writing – review & editing, Writing – original draft, Methodology, Formal analysis. **Paula Sartori:** Writing – review & editing, Writing – original draft, Methodology, Formal analysis. **Nayrim B. Guerra:** Writing – review & editing, Writing – original draft, Methodology, Formal analysis. **Cristiani C.P. Cid:** Writing – review & editing, Writing – original draft, Formal analysis. **Robinson C.D. Cruz:** Writing – review & editing, Writing – original draft. **Noel M. Gately:** Writing – review & editing, Writing – original draft. **Declan M. Devine:** Writing – review & editing, Writing – original draft, Supervision. **Marcelo Giovanela:** Writing – review & editing, Writing – original draft, Conceptualization. **Mariana Roesch-Ely:** Writing – review & editing, Writing – original draft, Conceptualization. **Janaina S. Crespo:** Writing – review & editing, Writing – original draft, Supervision, Project administration, Funding acquisition, Conceptualization.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

Data availability

Data will be made available on request.

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