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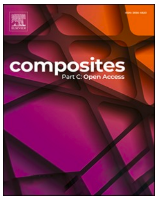

Improved properties of high-density polyethylene by integrating high content of bio-fillers based on green nanolignin for applications in plastic industry

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ABSTRACT

A study on the processability and physical and mechanical properties of different thermoplastic-based green nanocomposite materials for applications in injection processes is presented: commercial high-density polyethylene acting as the polymeric matrix and high contents of two types of modified nanolignins serving as bio-based fillers. It is a novel procedure to obtain good dispersibility of high loadings of bio-fillers in polyethylene which is difficult to achieve according to literature. The inclusion of green fillers brings property enhancements that are normally seen with expensive nanofillers like carbon nanotubes or graphene: improvement of mechanical properties while keeping good thermal stability and antioxidant properties has been obtained. To compare the effect of the inclusion of the two different nanofillers at 15, 25 and 35 % wt loadings on the characteristics of polyethylene, physical, mechanical, and morphological tests were performed. Injection pellets were fabricated and used to fabricate coupons to evaluate the mechanical features by performing both flexural and tensile tests. Remarkable enhancements (up to 60 % with respect to the neat polymer) of certain mechanical properties were observed for composites with 35 % wt loading of nanofillers. Moreover, investigating the effect of the bio-based nanolignins inclusion at high loadings on both crystallization and melting temperature suggested the robust thermal stability of the presented composites. As nanolignin is a renewable filler-type, it will bring properties like low-cost, low toxicity and partially replace petroleum-based stabilizers or other additives. Polyethylene with nanolignin becomes a “partially bio-based” high-performance material without changing the polymer backbone.

1. Introduction

The integration of biomass derived components such as processed lignin or nanolignin into blends of high-density polyethylene (HDPE) represents a forceful advancement in sustainable materials science nowadays. The global demand in bio-based alternative materials market was valued at approximately 1.32 billion EUR in 2024 and is projected to grow at a CAGR of 4.54 % from 2025 to 2030. This growth is driven by regulatory pressures and the adoption of new Circular Business Models (CBMs), especially in Europe [1]. Despite its abundance, only a small portion of lignin is currently valorized into high-value applications. From a freedom-to-operate perspective, the patent landscape for lignin-polymer composites is still in an emerging phase. This combination of robust market growth and needs along with a strong regulatory alignment is expected to position nanolignin-reinforced HDPE as a strategic material for future research and industrial driven applications.

From another perspective the broad use of polymers and plastics in different industrial sectors has not stopped growing in the last century [2]. The adaptability of these materials to many different fabrication processes is key to understanding this growth potential. Considering this, numerous methodologies to modify physical and chemical properties of polymers have been successfully developed and applied. The

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preparation of polymeric compounds by reinforcing them with fillers and nanofillers is one of the most versatile strategies to provide plastic materials with new properties [3,4] or to reduce, if convenient, their presence in formulations of new advanced composite materials. Several different aspects need to be considered when a nanofiller based polymeric composite is proposed. Under optimized loads of reinforcements, polymeric compounds may present improved mechanical properties like mechanical strength [5], stiffness [6] and toughness [7] since they provide reinforcement at the nanometer scale [8]. Also enhanced thermal stability can be achieved under controlled addition of nanofillers by increasing heat resistance which leads to materials more suitable for applications requiring elevated temperature performance [9]. On the other hand, polymeric based composite materials can also become multifunctional when carefully selecting nanoparticles to be integrated. Hence, increased electrical conductivity [10], enhanced optical properties [11], better wear resistance [12] or lower permeability [13] are examples of material properties and functionalities that can be conveniently tuned by developing polymeric compounds integrating nanofillers. There are also relevant research lines like the one presented by Junchao Ren et al. developing a novel approach to create nanocomposite films entirely from biomass-derived materials exhibiting promising properties such as mechanical strength and environmental sustainability [14] and an in-plane thermal conductivity and electromagnetic interference (EMI) shielding effectiveness [15]. Other works present biobased polymers like PLA to investigate the incorporation of carbon quantum dots (CQDs) into composite and develop nanocellulose based films with enhanced degradation, thermal, and mechanical properties since the addition of CQDs improves the dispersion of cellulose nanofibers within the PLA matrix [16]. It is important that all advantages brought by nano compounding science to materials can be obtained while keeping lightness of polymeric structures which is crucial in many of the most important industrial sectors like aeronautics of wind energy [17].

Beyond these advancements, current standards and regulations demand green transition to new ecological alternative materials, thus reducing the use of non-environmentally friendly traditional materials. To this extent, in the last decades numerous works dedicated to the development of fibers and nanoparticles from biobased sources have been published [18,19]. A wide range of potential advantages are gained. Not only do these types of components contribute importantly to reduced environmental footprints by being derived from renewable, biodegradable, or non-toxic sources but also, they bring significant improvements concerning low toxicity, recyclability, reduced emissions while matching regulatory compliance and market trends developed in the last years.

One of the polymeric matrices that has shown better processability performance and ability to integrate functional bio-fillers is HDPE. Recent published works show relevant advancements. A. Qingfa Zhang et al. obtained HDPE based composites reinforced with biochar derived from agricultural waste improving mechanical strength, elasticity, and thermal stability [20]. On the other hand, considering the use of bio-fillers influence in processability and properties of HDPE, works have been published concerning the use of natural fibers, especially in those obtained from wood. The study of Muhamad Shafiq Sukiman et al. [21] shows results concerning the potential use of wood–plastic composites manufacturing formwork structures through thermoforming, also by integrating wood based particles Demagna Koffi al[22] obtained a reduction of the specific heat capacity of HDPE indicating that wood fillers act as partial thermal insulators and also Fouad Erchiqui et al. [23] published a very important paper which demonstrates that composites with moderate wood content ($\leq$ 40 %) offer a balanced compromise between stiffness and thermoformability of HDPE, whereas higher loadings lead to poor deformation and excessive internal stresses. But also, in another work [24], the same author proves that the inclusion of the wood flour content in HDPE composites makes the material more elastic, reducing its viscoelastic deformability. In another interesting work by K.K. Agbozouhoue et al. [25] the use of ZnO to treat fibers effectively prevented fungal growth, demonstrating antifungal properties in PE based fibers composites whereas M. Tazi et al. [26] conveniently investigated the impact of softwood filler content on the biodegradability and morphological properties of wood–polyethylene composites. As well the reduction of the damage and durability of plant fiber-reinforced HDPE are key topics in the recent studies. In their paper, Alencar Bravo et al. [27] evaluate the fatigue life and thermomechanical behavior of novel green and bio-composite gears reinforced with birch fibers, comparing them to high-performance thermoplastics. The same author presents a comprehensive model for assessing fatigue life and damage modes of plastic gears [28]. Demagna Koffi et al. [29] investigated the impact of cooling holes on the thermal and mechanical performance of plastic gear teeth.

In the presented work, two distinct types of nanolignin were dispersed into HDPE. The inclusion of this type of particles within this polymer means an interesting research route in materials science since it combines the convenience of renewable resources with the high-performance of HDPE and applications in biodegradable plastics [30, 31].

Lignin, which is derived from a complex polymer in plant cell walls, is a promising additive used to improve the sustainability of polymers when compounding together [32,33]. In the case of HDPE, several advances can be obtained while maintaining this improved sustainability like enhanced UV resistance and better chemical barriers [34]. Nevertheless, some important challenges and drawbacks are yet to be overcome. On the one hand developing a homogeneous dispersion of lignin nanofillers within the polymeric matrix during industrial processes like extrusion could be a challenge due to the natural hydrophobicity of polyethylene and the polar nature of lignin. An effective adjustment of process parameters such as temperature or processing time must be considered to reduce the possibility of particles aggregations formation. Whereas the effect of nanolignin on the mechanical properties of polyethylene has not been widely reported yet, there are some studies reporting increases in flexural strength when lignin is chemically modified that are very interesting. Laura Dehne et al. [35] reported an enhancement in the flexural strength up to +30 % vs unmodified-lignin blends based in Polyethylene with 50 wt % loadings of lignin. Furthermore, there are works emphasizing the importance of lignin modification to achieve a good interaction with polymers to get good mechanical properties as in the work published by Majid Kiaei et al. [36]. Other study to consider is the one published by R.R.N Sailaja et al. [37] where improvements in different mechanical properties of LDPE (low density polyethylene) were achieved by blending this polymer with up to 40 % wt loadings of esterified lignin.

If research continues improving compounding techniques, the inclusion of nanolignin in polyethylene will acquire a key role in the development of greener, environmentally friendly polymeric multifunctional materials in the future. Indeed, the results presented below cover the growing necessity of developing sustainable materials to protect environment and contribute positively to the global climate crisis. By incorporating biobased additives as lignin, the use of fossil-fuel derived and widely use PE is reduced. Nanolignin is presented as well as a green alternative to other non-green silica, clay or carbon black, the improvement of certain properties, explained below, is an important achievement. Concerning the innovation and the novelty of this research, the conventional lignin particles present problems to integrate in polymers. Due to high surface area nanolignin interacts more effectively with PE leading to better interfacial adhesion and to improved mechanical and thermal properties. The presented control on particle size, dispersion, and loading percentage of nanolignin allows fine-tuning of composite performance which is a key innovation area with potential important applications in packaging materials with antimicrobial properties or sustainable composites for construction or automotive uses among others.

2. Materials & methods

2.1. *High-density polyethylene*

PE-Liten-PEHex (purchased from Orlen Unipetrol) was supplied by SILON plastic compounding company and used as received. Kraft lignin (BioPiva 100) was supplied by UPM (Finland); all other chemicals were purchased from commercially available vendors within the EU and used without further purification. RO water was used for the preparation of nanolignins. The two types of nanolignins were provided by Creative Nano PC, prepared from commercially available Kraft lignin either via chemical modification (LigAc) or via an in-house developed physico-mechanical method using ultrasonication (NK) [38].

2.2. *FTIR*

Fourier-Transform Infrared Spectroscopy (FTIR) was recorded on a Tensor 27 spectrometer, a Bruker instrument fitted with a diamond Attenuated Total Reflection (ATR) accessory at a specific resolution of 4 cm^{-1} in the wavenumber range from 600 to 4000 cm^{-1} in transmittance mode. The baseline of all presented spectra was corrected, and the spectra was normalized.

2.3. *DLS*

Dynamic Light Scattering (DLS) was used to evaluate the particle size of NK and LigAc additives using a Litesizer 500 particle analyzer (Anton Paar, Graz, Austria). Prior to analysis, a dispersion in water was prepared at approximately 70–200 ppm to obtain 30–40 % transmittance. All measurements were performed no less than three times.

2.4. *TGA*

A Q50 TGA model from TA Instruments was used to perform Thermogravimetric Analysis (TGA). The samples were heated from ambient temperature to 750 °C in a 40 mL/min flow of air at a heating rate of 10 °C/min.

2.5. *DSC*

A Q20 DSC model from TA Instruments with Heat Flux was used to perform Differential Scanning Calorimetry analysis (DSC). It is equipped with Tzero® Technology for enhanced signal and baseline performance, dual digital mass flow controllers and compatibility with autosamplers and multiple cooling systems. Important thermal transitions, including crystallization temperature (Tc) and melting temperature (Tm) are achieved. In the experiments presented in this work, a double heating ramp from 20 °C to 240 °C with double cooling at a heating/cooling rate of 10 °C/min was used.

2.6. *SEM*

The morphology and dispersion of fillers within the HDPE polymeric matrix of the developed composites were studied by Scanning Electron Microscopy (SEM). Samples were embedded in an epoxy resin before being carefully cut using a low-speed saw and subsequently polished to preserve the fractured surfaces. The samples were then mounted onto the sample holder using double-sided carbon tape. To ensure the required conductivity of HDPE-LigAc & HDPE-NK composites necessary for SEM high-quality imaging, all composites were gold coated using a SC7620 ‘Mini’ Sputter/Glow Discharge System (Quorum Technologies LTD, UK). SEM images were recorded on a TESCAN VEGA COMPACT instrument, which provided detailed surface images for understanding the distribution of components and interactions within the HDPE-lignin hybrids.

2.7. *Contact angle*

Water contact angle measurements were performed using an optical tensiometer, One Attention (Biolin Scientific, Espoo, Finland). Measurements were performed in quadruple and averaged.

2.8. *UV-vis*

An Agilent Cary 60 UV–vis instrument run in absorbance mode was used to record UV–vis spectra. The antioxidant activity of the HDPE composites with NK and LigAc fillers at different filler loadings were evaluated by monitoring the reduction rate of the DPPH radical in the antioxidant’s (NK/LigAc) presence via UV–Vis spectroscopy. This technique measures the ability of a substance to scavenge free radicals by observing the decrease in absorbance of a DPPH solution after incubation with the test sample. A 0.079 mM DPPH solution in EtOH was prepared and stored in the dark for 16 h at room temperature. The prepared films with the same dimensions (1 cm × 1 cm) were immersed in 3 mL of the DPPH/EtOH solution at room temperature and kept in the dark. The composites’ antioxidant capacity was determined by measuring the absorption decay at 517 nm at regular time intervals via UV–Vis. The residual DPPH content in the solution was calculated using Eq. (1):

$$\text{Residual DPPH content (\%)} = 100 - 100\,(A_0 - A_1/A_0) \tag{1}$$

where A_0 is the absorbance of the control sample and A_1 is the absorbance in the presence of the films.

2.9. *Rheometer*

Rheometric measurements were taken using an Anton Paar MCR 102e rheometer operating in both oscillatory and rotational modes. It was used to determine both deformation and fluidity in all types of tests. In this case, a plate-to-plate geometry with a diameter of 25 mm and a gap between plates of 1 mm was used. The samples were tested in oscillatory mode, always below the viscoelastic limit, with a constant strain of 0.1 %. Both storage modulus and viscosity values were acquired using rheometric measurements.

2.10. *Mechanical tests*

Tensile tests were performed in a universal machine, SHIMADZU AG-X PLUS 5 kN, following standard UNE-EN ISO 527–2:2012. Fabricated specimens were subjected to tensile testing at a speed of 1 mm/min up to 2 % strain to determine elastic modulus and at a speed of 20 mm/min until failure to determine tensile strength. The same machine was used to perform flexural tests under standard UNE-EN ISO 178:2020 under a force load of 1.5 N.

2.11. *Extrusion process*

Fig. 1 presents pictures corresponding to the different steps of the extrusion process related to the compounding development of this work. Figs. 1a and b depict pictures of as-received NK powder and commercial PE-Liten-PEHex polyethylene pellets. No treatment of these two materials has been necessary prior to the mixing process. The different reinforced polymeric composites were manufactured using a THERMO FISHER Haake Polylab QC conical twin screw extruder, which can be adapted to a mixer chamber to produce the blends and to an extrusion chamber to produce filament. Weighted amounts of thermoplastic pellets of HDPE and lignin (Figs. 1c and d) were hand mixed prior to being gravity fed to the mixer barrel through a hopper. Within the barrel the blend melts and the fillers are homogeneously dispersed within the polymeric matrix. Fig. 1e shows the uniform aspect of LigAc15 that was observed during the mixing process. During the process, torque is

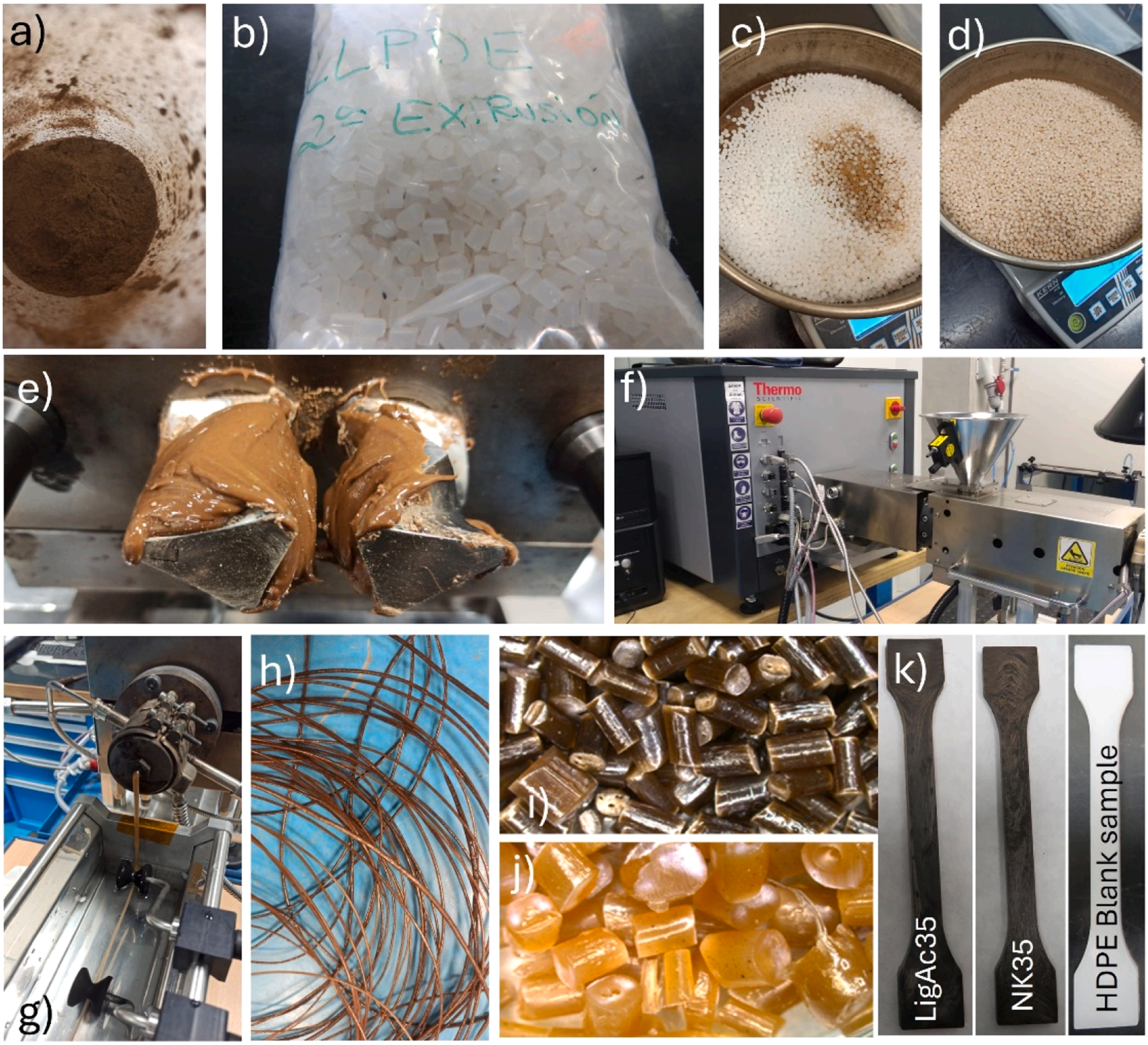


Fig. 1. a) NK powder as received from Creative Nano. b) Neat pellets of commercial PE-Liten-PEHex Polyethylene. c) HDPE and LigAc "as received" materials before and d) after being manually mixed. e) Melted state of the HDPE-LigAc masterbatch within the mixing chamber. f) THERMO FISHER Haake Polylab QC conical twin screw extruder. g) Compounded filament when being extruded. h) Final filament manufactured. i) and j) final produced pellets from HDPE reinforced with 15 % of LigAc and NK fillers respectively. k) Injected coupons of HDPE containing 35 %wt of LigAc and NK as well as a reference coupon of the neat HDPE polymer.

continuously monitored to determine when the blend achieves suitable homogeneity (torque stabilizes at 10 Nm in all cases). Once the blend is well mixed, the resulting composite is transferred to an extruder barrel (Fig. 1f), where it is mixed and melted again to produce filament. To ensure that the final material will not degrade or become weakened, it is crucial to control the temperature; the extruder barrel has 4 electrical heating zones. The temperatures are set in an increasing profile from the rear to the front: 1) 150 °C; 2) 160 °C; 3) 170 °C; and 4) 180 °C, the molten extrudate flows to the discharge end of the barrel. These temperatures were selected considering that the nanolignins may degrade at higher temperatures, leading to water release, which may lead to a significant degree of porosity and in the subsequent deterioration of the composite's mechanical properties. When homogeneity is sufficient, the blend is forced out through the head of the extruder, which dictates the shape and size of the final filament (1,75 mm, see Fig. 1g). Afterwards, the extruded filament is rapidly cooled in air to solidify (see Fig. 1h). This step is crucial for obtaining the desired properties of the final injection pellets. Then, the filament is cut into small pellets using a pelletizer MP 10 Lab from KFM at a speed of 5 kg/hour. The obtained pellets are approximately 2 mm in size, which was deliberately controlled to facilitate the subsequent injection procedure. Figs. 1i and 1j illustrate obtained pellets for composites LigAc15 and NK15 respectively.

2.11.1. *Injection process*

Pellets obtained from the aforementioned extrusion process were used to fabricate coupons by injection molding for flexural and tensile tests. Briefly, pellets were fed into the injection molding machine (Engel VC40 Tech), a 40-ton injection molding machine with a 30 mm screw diameter and an injection volume of 99 cm^3; it injects up to 120 *g* of material per cycle under 1451 bar. The blend is then melted by heat under pressure in a heated barrel. Then the melted material was injected into molds, corresponding to the two different types of specimens of interest, flexural and tensile, under high pressure. Fig. 1k shows obtained tensile coupons integrated with different loads of LigAc on the left and on the right a coupon with neat HDPE. In this present work, the injection molding process was optimized by adjusting the following parameters: speed 22 cm^3/s, filling time 1,06 s; filling pressure 1203 bar and shear rate 0,94 /s.

2.12. *Hot press machine*

Samples to evaluate angle contact results were fabricated using a hot press machine EPROPR_002 - Prensa ISOJET PRESSE at 180 °C.

3. Results & discussion

3.1. *Properties of nanolignins*

The different nature of the two nanolignin fillers are expected to alter the properties of their corresponding HDPE composites. To better understand this influence, characterization studies were performed on the neat nanolignin fillers. Fig. 2a illustrates the FTIR spectra of NK and LigAc, whereas Fig. 2b lists all their corresponding band assignments. From DLS measurements it is apparent that the NK additive had a small standard deviation in particle size with a measured HDD= 498±37 nm and PDI= 27 %, as illustrated in Fig. 2c. LigAc, on the other hand, had a smaller particle size with a measured HDD= 224±27 nm and PDI= 21 %. Finally, thermogravimetric analysis (TGA) was employed to study the thermal profile of the NK and LigAc fillers. Fig. 2d depicts TGA curves of NK and LigAc; thermal decomposition takes place after 180 °C. A 50 % mass loss occurs at 447 °C and 483 °C for NK and LigAc, respectively. Complete thermal decomposition is observed above 590 °C with a low ash content of 2–4 wt % for both nanolignins. These results reveal that both types of selected green additives are suitable candidates to conveniently tune properties of polymers like HDPE and can be employed in both extrusion and injection processes since their respective process windows lie out of the degradation temperature range of HDPE.

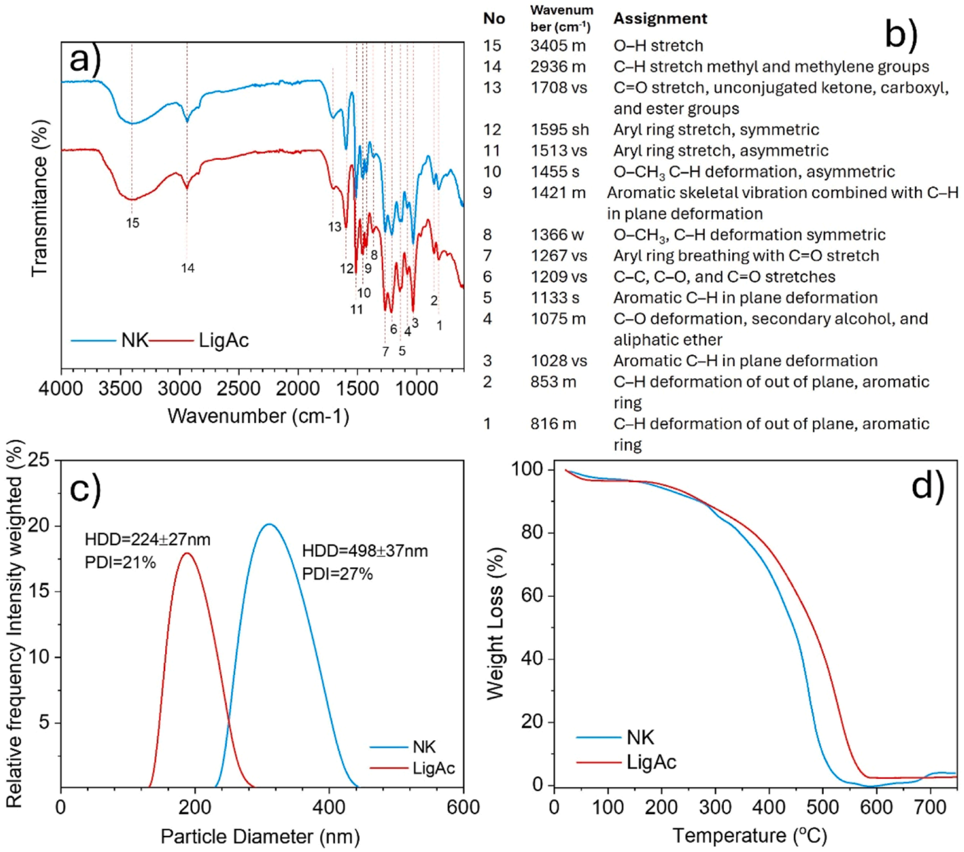
3.2. *Compounding development procedure*

Polymers and fillers were compounded together using a double-phase process consisting of a mixing procedure and an extrusion procedure aiming to optimize both processes and compounding parameters. This process has been explained in detail in Section 2. Two sets of composites, one for each nanolignin filler with three different % loadings each, were prepared. Firstly, composite masterbatches containing 35 wt % of each nanolignin were produced. Subsequently, to obtain composites with different filler content, the masterbatches were diluted with HDPE to achieve the targeted loads of 15 % and 25 %. Table 1 summarizes all the composites prepared.

The interaction between the polymer matrix and nanolignin fillers is primarily governed by physical dispersion, with interfacial adhesion enhanced through secondary interactions, such as van der Waals forces or hydrogen bonding, rather than by covalent chemical bonding. This is due to the mismatch in polarity between the polymer matrix and nanolignin fillers. HDPE is a nonpolar semicrystalline polymer which is

Table 1
Summary of all HDPE-nanolignin blends prepared in this study.

Filler loading	LigAc	NK
15 wt %	LigAc15	NK15
25 wt %	LigAc25	NK25
35 wt %	LigAc35	NK35



No	Wavenumber (cm^{-1})	Assignment
15	3405 m	O–H stretch
14	2936 m	C–H stretch methyl and methylene groups
13	1708 vs	C=O stretch, unconjugated ketone, carboxyl, and ester groups
12	1595 sh	Aryl ring stretch, symmetric
11	1513 vs	Aryl ring stretch, asymmetric
10	1455 s	O–CH_3 C–H deformation, asymmetric
9	1421 m	Aromatic skeletal vibration combined with C–H in plane deformation
8	1366 w	O–CH_3, C–H deformation symmetric
7	1267 vs	Aryl ring breathing with C=O stretch
6	1209 vs	C–C, C–O, and C=O stretches
5	1133 s	Aromatic C–H in plane deformation
4	1075 m	C–O deformation, secondary alcohol, and aliphatic ether
3	1028 vs	Aromatic C–H in plane deformation
2	853 m	C–H deformation of out of plane, aromatic ring
1	816 m	C–H deformation of out of plane, aromatic ring

Fig. 2. a) FTIR-ATR spectra of NK and LigAc in transmittance mode. Table in b) Assignment of bands in FTIR spectra of nanolignins: vs very strong; s strong; m medium; w weak; sh shoulder relative to other peaks in the spectra. c) Particle size distribution of LigAc and NK as measured by DLS. d) TGA graph of NK and LigAc, heated at a rate of 10 °C/min under air flow.

hydrophobic and has low surface energy. Which explains why it presents a poor chemical affinity for polar fillers like nanolignin which are rich in functional polar groups and thus hydrophilic. Nanolignin particles are dispersed into the HDPE matrix, but since HDPE is nonpolar, the molecular compatibility between both phases is low, and thus the nanoscale size of the filler is particularly important to achieve a uniform dispersion, reducing agglomeration and improving mechanical and thermal properties. On the other hand, concerning interfacial interactions, these are determined by a secondary mechanism: Van der Waals forces between HDPE chains and the hydrophobic regions of lignin. In the presented report, the effects of this mechanism on key properties of compounding are discussed: dispersed nanofillers function as mechanical reinforcement, increasing stiffness and sometimes tensile strength, and considering thermal stability, nanolignin can improve or keep the thermal resistance of the polymer. In the presented work, agglomeration problems have been overcome by using a twin-screw extruder with optimized screw elements to produce a pre-blend or masterbatch. Another potential limitation of the presented methodology is the possibility of lignin degradation under certain temperature conditions. Lignin begins to degrade between 200 and 250 °C and the typical PE extrusion temperatures (160–220 °C) which is close to this range. It is important to keep the processing temperature as low as possible for the selected material (<200–210 °C if possible) to avoid darkening/carbonization, odor, release of volatiles, reduction of molecular weight, or deterioration of mechanical performance.

To select the range of interest in terms of filler dispersion and filler content maximization within the polymeric matrix and develop a more efficient and effective formulation, rheometric measurements were conducted. This technique provides a quantitative route to estimate the optimum filler load in a polymer matrix by evaluating the influence of several compounding characteristics, such as viscosity, which can be estimated as a function of oscillatory frequency (ω) and temperature (T) during the rheology measurements from the direct measure of storage and loss modules (G' and G''). In the first place, the basic relationships of the main rheological variables should be explained:

$$G^*(\omega)=G'(\omega) + i\cdot G''(\omega) \quad (2)$$

$$\eta^*(\omega)=\eta'(\omega) - i\cdot\eta''(\omega) \quad (3)$$

From these definitions the expressions corresponding to the dependance of modules with oscillatory frequency are:

$$G'(\omega)= \omega\cdot\eta''(\omega) \quad (4)$$

$$G''(\omega)= \omega\cdot\eta'(\omega) \quad (5)$$

The expression corresponding to the dependance of modulus with the temperature is:

$$G^*(\omega,T)= (1/aT) \cdot \eta^*(aT\cdot\omega,Tref) \quad (6)$$

Where ω is the oscillatory frequency, T is the temperature, G* is the complex modulus, G' is the storage modulus, G'' is the loss modulus, η is the viscosity, and aT is the horizontal shift factor. In the Supplementary Information section, the rheological curves corresponding to viscoelastic moduli G' and G'' in function of angular frequency for temperature values ranging from 190 °C to 220 °C and for both types of analyzed nanolignins are plotted. These results provide a sensitive means to assess the homogeneity of a compound since well-dispersed mixtures exhibit consistent and reproducible G' and G'' profiles across samples, whereas significant variations or irregular trends indicate potential agglomeration, uneven filler distribution, or incomplete mixing. Thus, G' and G'' analysis serves as an efficient, indirect indicator of mixture uniformity. In the case of this study, there are not any significant differences between the modules that could indicate a poor dispersion of the fillers.

The storage modulus plays an important role in understanding how the fillers determine the final processed compounding properties. Storage modulus increases with the load up to a certain value where the polymer becomes too stiff and additional fillers will compromise properties like flexibility and processability of the developed compounds. This inflexion point indicates the optimum weight percentage of fillers within the matrix. In Figs. 3a and b, the evolution of the storage modulus in relation to the filler content for NK and LigAc, respectively, is plotted. These rheology measurements indicate that the percolation threshold lies between 30 % wt and 40 % wt for both fillers. In rheology science the value of storage modules indicates the interaction between fillers. In the case of the present study, three distinct stages are identified as shown in the graphs. An initial plateau corresponding to lower filler loads appears due to the isolation of particles. As the percentage of nanolignin fillers increases, these begin to develop an internal network, which will confer new properties to the final compounding, but no aggregates affecting quality are yet formed. When loads of fillers overcome the percolation threshold, particle aggregation appears, giving rise to another plateau in the storage modulus representation.

3.3. SEM

Fig. 4 illustrates SEM images of the cross section of the composite tensile coupons. For neat HDPE (top left) a smooth surface is observed whereas the HDPE-nanolignin composites (all other SEM images) seem to have a 'roughened' surface. It is noteworthy that even at high filler loadings (NK35 & LigAc35) a homogeneous dispersion of the nanolignin fillers in the HDPE matrix was observed.

3.4. Contact angle

Considering the potential industrial applications of HDPE composites with improved features, contact angle analysis provides insights into the surface properties of the material, such as wettability, which is critical for various applications. To produce thin film samples for contact angle measurements (Fig. 5a), a hot press machine was used following the procedure described in Section 2. The sessile water droplet method was used to investigate the hydrophilicity of all HDPE composite films. Measurements illustrated in Fig. 5b were performed in quadruple.

Although there is certain standard deviation in some of the measurements, a general trend of increasing hydrophilicity with increasing lignin filler loading may be deduced. Neat HDPE is more hydrophobic than all the composites with a contact angle of 95.8° HDPE-LigAc composites are more hydrophobic compared to HDPE-NK composites, which may be explained by the partial alteration of hydroxyl groups in LigAc, which occurs during its synthesis. Since hydroxyl groups enhance hydrophilicity, HDPE-NK has more free hydroxyls and thus more wetting capability. Obtaining HDPE-based composites with improved hydrophilicity might be of special interest in the coating industry.

3.5. Antioxidant activity

Antioxidant activity of HDPE composites containing different loadings of nanolignin fillers was studied using the DPPH method (Section 2). The results are shown in Fig. 6.

It is apparent that the neat HDPE sample has negligible antioxidant activity; however, a substantial increase is observed when NK and LigAc lignins are incorporated into HDPE as additives, which in turn further increases with higher additive loading. Unsurprisingly, the HDPE-NK35 composite shows the greatest antioxidant activity, followed by HDPE-NK25 and HDPE-Lig-Ac35. The antioxidant activity of nanolignins arises from the free phenolic hydroxyl groups, which are abundantly present; the partial alteration of hydroxyl groups in LigAc could explain its reduced antioxidant activity compared to NK.

3.6. Viscosity and thermal properties

To obtain a better understanding of how the integration of green fillers affects the properties, such as the thermal stability of the final

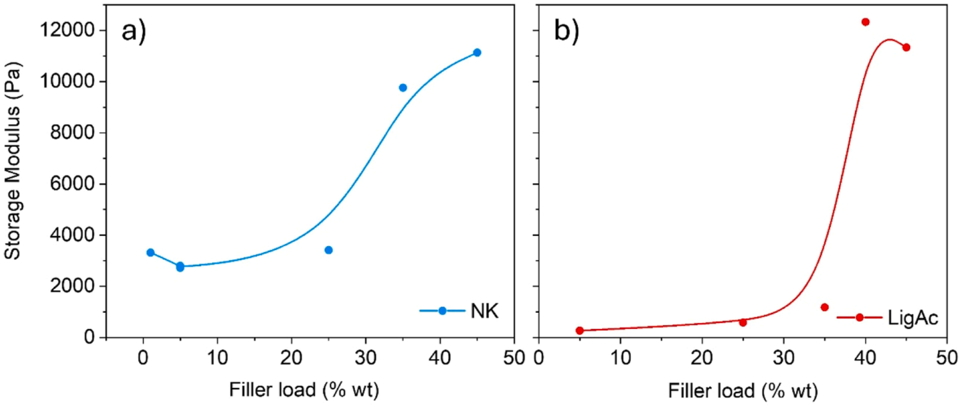


Fig. 3. Evolution of the storage modulus with the fillers content for NK (a) and LigAc (b) respectively.

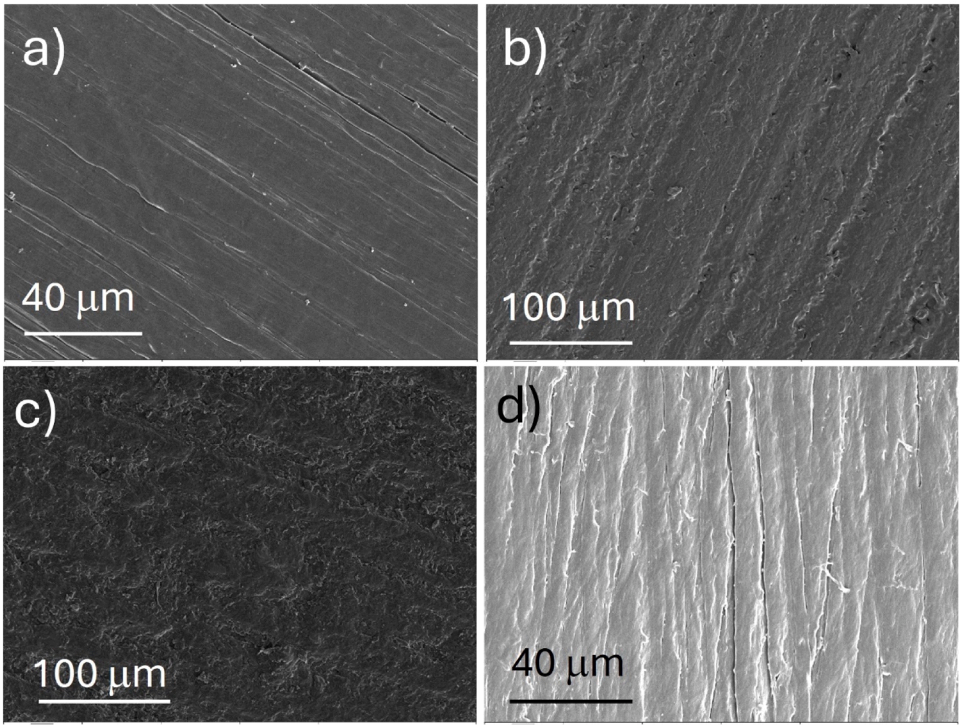


Fig. 4. SEM images of cross sections of composite tensile coupons. Neat HDPE (top left), NK35 (bottom left), LigAc35 (top right) and LigAc15 (bottom right). Respective scale bars are plotted in the bottom-left corner.

composites, some additional studies were performed. DSC tests were conducted following the procedure described in Section 2. The most relevant results are displayed in Figs. 7a and b, which compile values corresponding to the melting temperatures (Tm) and the crystallization temperatures (Tc) of all composites prepared. It is apparent that Tm slightly increases for 15 % filler loading in both cases, whereas further increasing the filler loading to 25 % results in composites with a Tm like neat HDPE. Interestingly, at 35 % NK loading, the Tm is increased to 138.4 °C, while in contrast, 35 % LigAc loading decreases the Tm to 131.8 °C. This may be due to weak interactions with the polymer or minimal nucleation effects. These results are relevant for applications in the injection molding and/or extrusion fields. On the other hand, Tc remains constant with increasing filler loading up to 25 % in both cases. After this, LigAc35 shows a slight increase in Tc while NK35 shows a slight decrease. The physical dispersion of particles within the polymeric matrix explains the obtained thermal stability. Lignin contains aromatic rings and crosslinked structures, which are thermally stable. When nanolignin is well-dispersed, it can function as a thermal barrier, slowing down polymer degradation by acting as a radical scavenger during decomposition and delaying heat transfer through the matrix.

Fig. 7c depicts the evolution of the viscosity of the HDPE composites as nanolignin filler loading increases gradually. These values have been estimated by performing rheometric measurements as described in Section 2. Even though adding reinforcements usually increases viscosity, a good dispersion of fillers in the polymer may allow the polymer chains to move more freely, which can reduce viscosity. However, if large clumps of nanofillers form, this could increase flow resistance and raise viscosity, especially when the filler content exceeds 30 % by weight, as observed in the case of LigAc. Fig. 7d shows the decrement of enthalpy of crystallization, estimated from DSC measurements previously explained, with the addition of nanofillers. In both cases, the evolution is quite abrupt, meaning that the crystallization degree of the composites is reduced, thus losing the capacity of being compressed, which might affect different polymeric processes. Dispersibility also

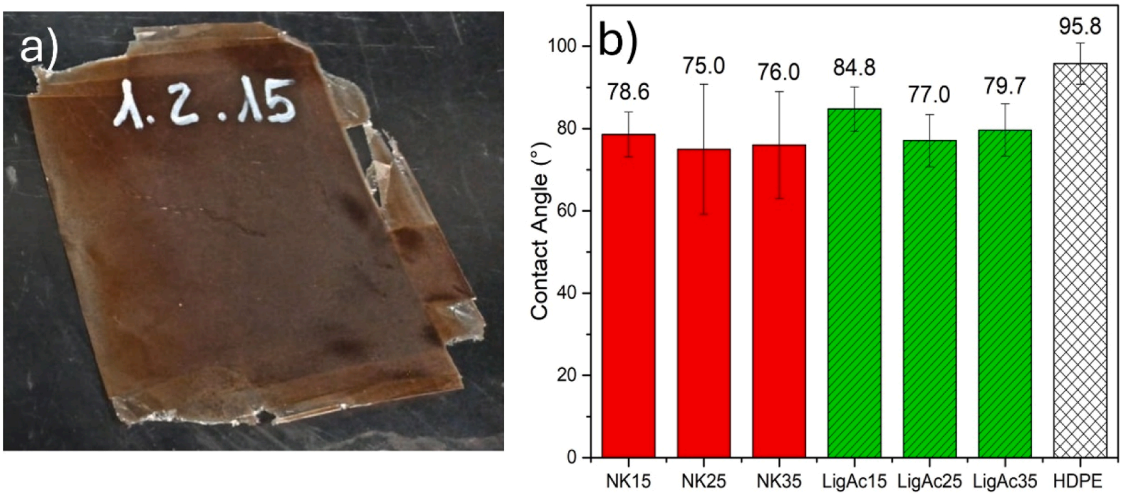


Fig. 5. a) Hot press sample corresponding to HDPE integrating 15 % wt of LigAc b) Contact angle measurements of HDPE-NK and HDPE-LigAc composites at different loadings.

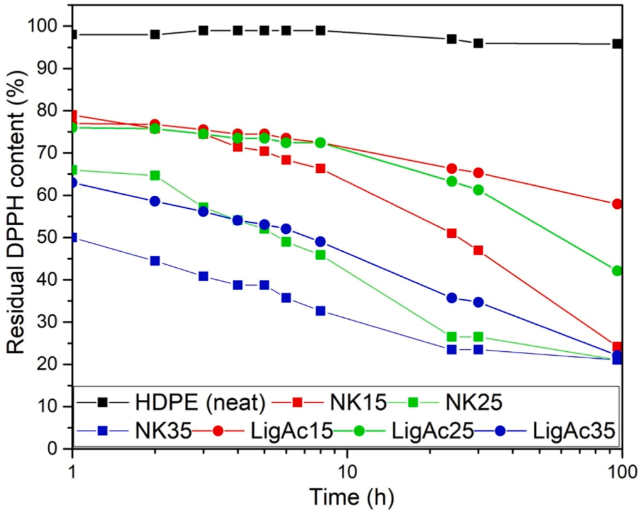


Fig. 6. Antioxidant activity of HDPE-NK and HDPE-LigAc composites at different loadings, measured by monitoring the reduction rate of DPPH radical using UV–vis spectroscopy.

affects the crystallization properties since nanolignin can function as a nucleating agent, promoting polymer crystallization if dispersed homogeneously. This explains the stability of the obtained crystallization temperature. On the other hand, good dispersibility should lead to slightly higher crystallization enthalpy (ΔHc) due to more polymeric chains organizing around dispersed particles. In the case of the presented work, this magnitude decreases with the filler load. This might indicate the presence of some particle agglomerates in the samples subjected to the study or maybe due to the amorphous nature of nanolignin.

3.7. Mechanical properties

Both flexural and tensile coupons were fabricated by injection molding following the procedure described in Section 2. Fig. 8 depicts a) tensile strength, b) elastic modulus, c) flexural strength, and d) flexural modulus of the prepared composites at different filler loadings. The error bars plotted in each case correspond to the standard deviation of the mean value corresponding to a set of 5 coupons evaluated under the exact same conditions. These error bars represent the variability of the represented magnitudes that each point summarizes. The small bars indicate the composite blends are homogeneous and consistent; thus, these bars provide a direct qualitative measure of the good compounding homogeneity systematically obtained by the developed methodology. The analyzed mechanical properties follow a similar trend with both types of fillers, but in all cases, the composites with LigAc presented slightly lower values. Tensile strength decreases as filler content increases (Fig. 8a) due to the progressive agglomeration of fillers with increasing the filler content. The formed agglomerates may function as stress concentrators, and this can result in the premature failure of the specific part under tensile stress. Furthermore, the compatibility between the hydrophilic fillers, such as the lignin, and hydrophobic polymers like HDPE is crucial to the resulting mechanical properties. If compatibility is poor, the weak interfacial interactions limit the stress transfer between fillers and matrix, which can result in reduced tensile strength. Contrary to the tensile strength, the flexural strength increases with higher filler content (see Fig. 8b). More specifically, an increase of up to ~60 % in flexural strength was observed for NK25 and ~30 % for LigAc25. Flexural loads involve a combination of tensile and compressive stresses. The fillers have made a strong contribution in increasing the compression strength of the material in the compression dominated zones. This reinforcing effect counteracts the weakening observed in the tensile properties and results in an increase of the overall flexural strength. A significant increase (~60 %) was also observed in elastic modulus for composites NK35 and LigAc35 (see Fig. 8c). Finally flexural modulus showed a similar trend with increasing filler load, an increase of ~60 % for NK35 and ~50 % for LigAc25 was observed (see Fig. 8d). This is a common observation in polymer nanocomposites and is often attributed to the rigid nature of the fillers, which constrain the mobility of the polymer chains and make the material stiffer. Some studies [35] reported increments of flexural strength when lignin is chemically modified up to +30 % vs unmodified-lignin blends when dispersed in polyethylene at 50 wt % loading of lignin. This value matches the one reported in the present work for the case of LigAc25 but is far from the 60 % enhancement obtained for NK25 %. This result suggests the importance of the particle size of nanolignin to get good dispersibility within a polymeric matrix and the beneficial effect on the compounding properties.

To better understand the influence of the two diverse types of nanolignin on the mechanical properties of the manufactured composites, the ANOVA method has been applied in a straightforward way for each of the magnitudes analyzed. Analysis of the values obtained for tensile strength allowed several important conclusions to be drawn NK particles result in a decrease from 20 to 15.2 MPa as filler content

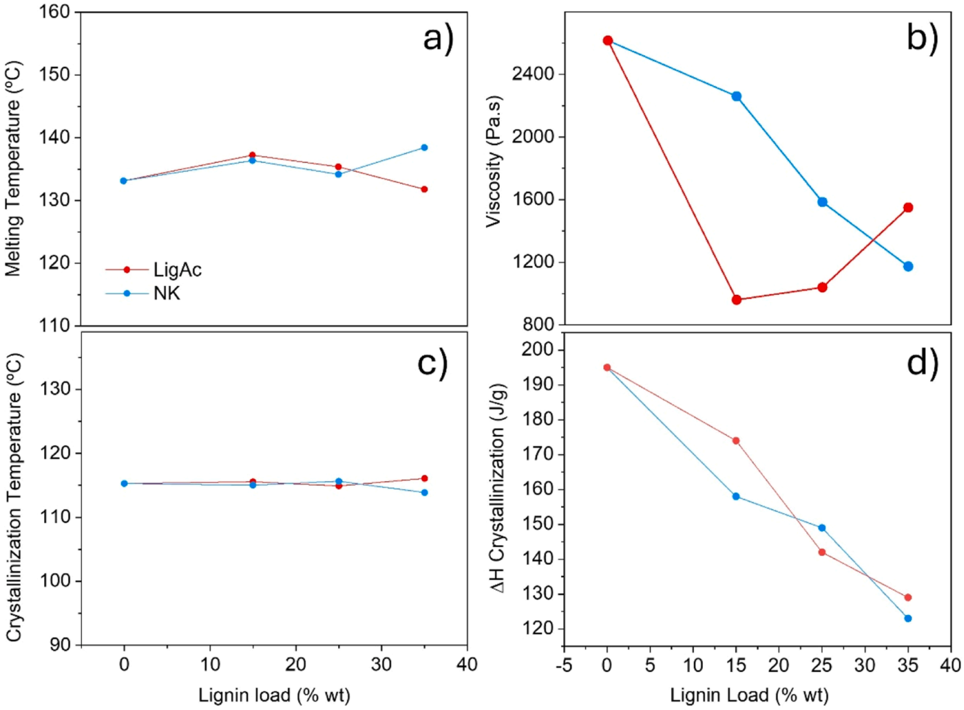


Fig. 7. Evolution obtained for Tm (a) and Tc (b), viscosity (c), and enthalpy of crystallization (d) for the two types of lignin studied: NK (blue lines) and LigAc (red lines).

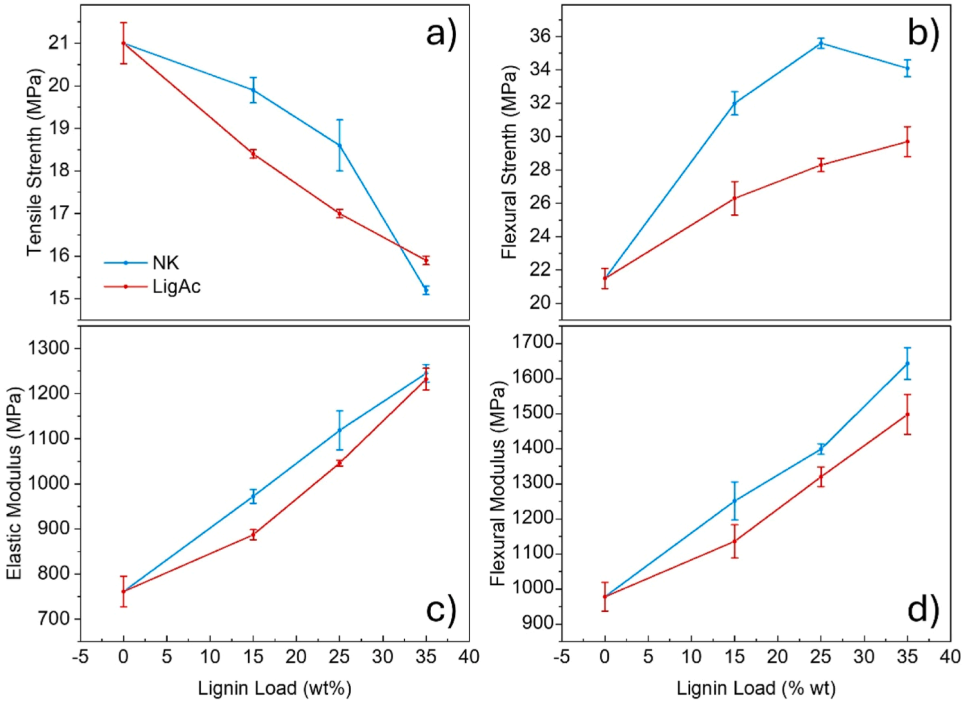


Fig. 8. Evolution with the load of LigAc (red lines) and NK (blue lines) of four significative magnitudes: a) tensile strength, b) elastic modulus, c) flexural strength and d) flexural modulus. Error bar in each case corresponds to the standard deviation of the mean value corresponding to a set of 5 coupons evaluated under the exact conditions.

increases, and LigAc causes a tensile strength to decrease from 20 to 15.9 MPa, a slightly less steep decrease. NK shows a steeper decline in tensile strength with increasing filler content compared to LigAc, so increasing filler content tends to reduce tensile strength for both fillers, due to stress concentration points or poor interfacial bonding at higher loading. LigAc maintains slightly better tensile performance at higher concentrations, indicating it may be more compatible or less disruptive to the matrix at high loadings. On the other hand, the ANOVA analysis on elastic modulus shows that it increases with filler content for both types, indicating that both fillers effectively reinforce the polymer. But NK is more efficient than LigAc at low and intermediate concentrations (15–25 %), producing higher modulus values while at high concentration (35 %), both fillers achieve similar modulus values, suggesting saturation of the reinforcement effect. Regarding flexural strength, NK provides stronger reinforcement at 15–25 % but may show a slight reduction at 35 % while LigAc is more consistent and gradually improves the property across all concentrations. These results might suggest that the inclusion of NK fillers could be a preferable option for maximum enhancement at moderate filler levels and LigAc may be a better option for higher filler loadings or better stability. Finally, some conclusions may be taken from the ANOVA analysis of flexural modulus. NK consistently provides higher performance than LigAc. Differences become more pronounced at higher concentrations, indicating NK is better suited for applications requiring maximum reinforcement. Therefore, the use of NK for high-strength composites should be preferable; nevertheless, LigAc may be considered for moderate reinforcement where other factors (cost, processing…) are important. These mechanical properties are strongly affected by the main mechanism between the interaction of filler particle-polymer: physical dispersion of particles within the polymeric matrix, good dispersion ensures efficient stress transfer from the HDPE matrix to the nanolignin particles since the elastic modulus improves due to the rigid structure of nanolignin.

Considering injection molding, it would be useful in future studies to understand the evolution of key plastic properties such as thermal conductivity (k), specific heat capacity (Cp), and density (ρ) of polymer nanocomposites with the temperature (T) and the nanofiller concentration (ϕ). In the Supplementary Information, the equations proposed are shown. In summary, it is foreseen that density decreases with temperature and increases with filler loading improving dimensional stability and structural integrity under heat. Additionally, the specific heat should increase with temperature (mostly from polymer) but decrease with filler loading which might affect processing conditions such as heating rates and cycle times. Finally, an increment of thermal conductivity with filler loading is expected, moderately influenced by temperature and sensitive to filler connectivity and interface quality. The estimation of these properties will undoubtedly lead to better designing of advanced and multifunctional polymer nanocomposites to be applied in different important industrial areas.

4. Conclusions

HDPE-based composites reinforced with different loadings of two types of nanolignin green fillers (NK and LigAc), ranging from 15 wt % to 35 wt %, were developed. Rheological analysis identified an optimal filler loading of approximately 35 wt %, ensuring good dispersion within the polymer matrix. The inclusion of nanolignin fillers provided several notable benefits. Contact angle measurements indicated increased hydrophilicity with higher filler content, while antioxidant activity also improved proportionally. However, the most significant enhancement was observed in mechanical performance: flexural strength increased by approximately 60 % and 30 % for NK25 and LigAc25 composites, respectively, compared to neat HDPE. Similarly, the elastic modulus increased by about 60 % in the NK35 and LigAc35 samples. Melt extrusion and injection molding parameters were optimized to fabricate specimens for mechanical testing. Results highlight the complex balance between additive fillers and polymer matrix. Emphasizing the importance of optimizing the dispersion of the fillers, selecting appropriate processing conditions which are essential to maximize filler efficiency and achieve an optimal balance between stiffness and strength. Both nanolignin fillers exhibited comparable performance, with no clear advantage of one over the other.

CRediT authorship contribution statement

Olalla. Sanchez-Sobrado: Writing – review & editing, Writing – original draft, Supervision, Project administration, Methodology, Investigation, Formal analysis, Conceptualization. **Alexander F. Tiniakos:** Writing – review & editing, Investigation, Conceptualization. **Rebeca. Abalde:** Writing – review & editing, Methodology, Investigation, Formal analysis, Data curation. **Marisol. Rivas:** Writing – review & editing, Methodology, Investigation. **Alexios. Grigoropoulos:** Writing – review & editing, Supervision, Methodology, Investigation, Conceptualization. **Angeliki. Nikolaou:** Methodology, Investigation, Conceptualization. **Alexandros. Zoikis-Karathanasis:** Writing – review & editing, Formal analysis, Conceptualization. **Ioanna. Deligkiozi:** Writing – review & editing. **Ricardo. Losada:** Writing – review & editing.

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.

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Supplementary materials

Supplementary material associated with this article can be found, in the online version, at doi:10.1016/j.jcomc.2025.100688.

Data availability

The data that has been used is confidential.

References

[1] www.grandviewresearch.com/industry-analysis/lignin-based-biopolymers-market-report2024.
[2] J.K. Fink, Reactive polymers: Fundamentals and applications: a Concise Guide to Industrial Polymers, William Andrew, 2017.
[3] K.B. Nilagiri Balasubramanian, T. Ramesh, Role, effect, and influences of micro and nano-fillers on various properties of polymer matrix composites for microelectronics: a review, Polym. Adv. Technol 29 (2018) 1568–1585, https://doi.org/10.1002/pat.4280.
[4] J. Njuguna, K. Pielichowski, S. Desai, Nanofiller-reinforced polymer nanocomposites, Polym. Adv. Technol. 19 (2008) 947–959, https://doi.org/10.1002/pat.1074.
[5] X. Wang, L. Wang, Q. Su, J. Zheng, Use of unmodified SiO2 as nanofiller to improve mechanical properties of polymer-based nanocomposites, Compo. Sci. Technol. 89 (2013) 52–60, https://doi.org/10.1016/j.compscitech.2013.09.018.
[6] D.M. Marquis, E. Guillaume, C. Chivas-Joly, Properties of nanofillers in polymer, Nanocompo. Polyme. Anal. Method. 59 (2011) 261, https://doi.org/10.5772/21694.
[7] M. Mirkhalaf, H. Yazdani Sarvestani, Q. Yang, M.B. Jakubinek, B.A. Ashrafi, A comparative study of nano-fillers to improve toughness and modulus of polymer-derived ceramics. Sci. Rep. 2021 Mar 26;11(1):6951. doi: 10.1038/s41598-021-82365-3.
[8] B. Fiedler, F.H. Gojny, M.H. Wichmann, M.C. Nolte, K. Schulte, Fundamental aspects of nano-reinforced composites, Compo. Sci. Technol. 66 (16) (2006) 3115–3125, https://doi.org/10.1016/j.compscitech.2005.01.014.
[9] A. Choudhury, A.K. Bhowmick, C. Ong, M. Soddemann, Effect of various nanofillers on thermal stability and degradation kinetics of polymer nanocomposites, J. Nanosci. Nanotechnol 10 (8) (2010 Aug) 5056–5071, https://doi.org/10.1166/jnn.2010.3030.
[10] Q. Fang, K. Lafdi, Effect of nanofiller morphology on the electrical conductivity of polymer nanocomposites, Nano. Expres. 2 (1) (2021) 010019, https://doi.org/10.1088/2632-959X/abe13f.
[11] W. Wildner, D. Drummer, Nanofiller materials for transparent polymer composites: influences on the properties and on the transparency—A review, J. Thermoplast. Compos. Mater. 32 (11) (2019) 1547–1565, https://doi.org/10.1177/0892705718797157.
[12] W. Brostow, H.E.E. Lobland, N. Hnatchuk, J.M. Perez, Improvement of scratch and wear resistance of polymers by fillers including nanofillers, Nanomaterials 7 (3) (2017) 66, https://doi.org/10.3390/nano7030066.
[13] K. Prasad, M. Nikzad, I. Sbarski, Permeability control in polymeric systems: a review, J. Polymer Res. 25 (11) (2018) 232, https://doi.org/10.1007/s10965-018-1636-x.

[14] B. Junchao Ren, R. Tan, C. Huang, J. Chen, M. Huang, L. Wang, H. Lei, F. Wang, Q. Zhang, All-biomass derived nanocomposite films, Nano. Lett. 25 (19) (2025) 7810–7817, https://doi.org/10.1021/acs.nanolett.5c00846.

[15] C.J. Ren, C. Huang, R. Tan, All-biomass nanocomposite films via facile and sustainable design procedure for thermal management and electromagnetic interference shielding, Adv. Sci. (2025) e10372, https://doi.org/10.1002/advs.202510372.

[16] D. Jianlong Chen, X.G.R. Tan, M. Huang, J. Ren, W. Liu, M. Wang, B. Li, Z. Ma, Q. Zhang, Achieving the simultaneous improvement of degradation, thermal, and mechanical properties of polylactic acid composite films by carbon quantum dots, Compos. Part B 299 (2025) 112442. ISSN 1359-8368.

[17] B. Mahesh, T. Khan, K. Devade, B. Krishna, S. Sreekanth, H.K. Eftikhaar, H. Thethi, N. Gupta, Review of composite materials and applications. Mater. Today. (2023). 10.1016/j.matpr.2023.10.026.

[18] T. Mishra, P. Mandal, A.K. Rout, D. Sahoo, A state-of-the-art review on potential applications of natural fiber-reinforced polymer composite filled with inorganic nanoparticle, Compos. Part C 9 (2022) 100298, https://doi.org/10.1016/j.jcomc.2022.100298.

[19] A. Atmakuri, A. Palevicius, A. Vilkauskas, G. Janusas, Review of hybrid fiber-based composites with nano particles—Material properties and applications, Polymers 12 (9) (2020) 2088, https://doi.org/10.3390/polym12092088.

[20] A.Q. Zhang, D. Zhang, H. Xu, W. Lu, X. Ren, H. Cai, H. Lei, E. Huo, Y. Zhao, M. Qian, X. Lin, E.M. Villota, W. Mateo, Biochar filled high-density polyethylene composites with excellent properties: towards maximizing the utilization of agricultural wastes, Ind. Crop. Prod. 146 (2020) 112185, https://doi.org/10.1016/j.indcrop.2020.112185. ISSN 0926-6690.

[21] M.S. Sukiman, F. Erchiqui, T. Kanit, A. Imad, Design and numerical modeling of the thermoforming process of a WPC based formwork structure, Mater. Today Commun. 22 (2020) 100805, https://doi.org/10.1016/j.mtcomm.2019.100805. ISSN 2352-4928.

[22] K. Demagna, A. Bravo, T. Alencar, T. Lotfi, F. Erchiqui, Optimized use of cooling holes to decrease the amount of thermal damage on a plastic gear tooth. Adv. Mech. Eng. 8 (2016). 10.1177/1687814016638824.

[23] F. Erchiqui, F. Godard, A. Koubba, M. Vincent, H. Kaddami, Investigation of relaxation properties and potentiality of the thermoformability of HDPE charged by wood flours, J. Reinforce. Plast. Compos. 28 (2008) 1153–1168, https://doi.org/10.1177/0731684407085431.

[24] F. Erchiqui, F. Godard, A. Gakwaya, A. Koubaa, M. Vincent, H. Kaddami, Engineering investigations on the potentiality of the thermoformability of HDPE charged by wood flours in the thermoforming part, Polymer Eng. Sci 49 (2009) 1594–1602, https://doi.org/10.1002/pen.21394.

[25] K.K. Agbozouhoue, D. Koffi, F. Erchiqui, Bernabé, ZnO ZnO treatment on mechanical behavior of polyethylene/yellow birch Fiber composites when exposed to fungal wood Rot, Polymers 15 (2023) 3664, https://doi.org/10.3390/polym15183664.

[26] M. Tazi, F. Erchiqui, H. Kaddami, Influence of SOFTWOOD-fillers content on the biodegradability and morphological properties of WOOD-polyethylene composites, Polym. Compos (2016) 39, https://doi.org/10.1002/pc.23898, n/a-n/a.

[27] A. Bravo, L. Toubal, K. Demagna, F. Erchiqui, Gear fatigue life and thermomechanical behavior of novel green and bio-composite materials VS high-performance thermoplastics, Polym. Test 66 (2018) 403–414, https://doi.org/10.1016/j.polymertesting.2016.12.031. ISSN 0142-9418.

[28] A. Bravo, D. Koffi, L. Toubal, F. Erchiqui, Life and damage mode modeling applied to plastic gears, Eng. Fail. Anal. 58 (2015) 113–133, https://doi.org/10.1016/j.engfailanal.2015.08.040. Part 1ISSN 1350-6307.

[29] D. Koffi, A. Bravo, L. Toubal, F. Erchiqui, Optimized use of cooling holes to decrease the amount of thermal damage on a plastic gear tooth. Adv. Mech. Eng. 8. (2016). 10.1177/1687814016638824.

[30] D.S. Bajwa, G. Holt, N. Stark, S.G. Bajwa, S. Chanda, M. Quadir, Nano boron oxide and zinc oxide doped lignin containing cellulose nanocrystals improve the thermal, mechanical and flammability properties of high-density poly (ethylene), Polymers 16 (1) (2023) 36, https://doi.org/10.3390/polym16010036.

[31] S.H. Hong, S.H. Hwang, Construction and foamability of lignin-reinforced low-density polyethylene biocomposites, Mater. Today Commun. 28 (2021) 102696, https://doi.org/10.1016/j.mtcomm.2021.102696.

[32] M.R. Ridho, E.A. Agustiany, M. Rahmi, E.W. Madyaratri, M. Ghozali, W.K. Restu, W. Fatriasari, Lignin as green filler in polymer composites: development methods, characteristics, and potential applications, Adv. Mater. Sci. Eng. 2022 (1) (2022) 1363481, https://doi.org/10.1155/2022/1363481.

[33] A. Naseem, S. Tabasum, K. Zia, M. Zuber, M. Ali, A. Noreen, Lignin-derivatives based polymers, blends and composites: a review, Int. J. Biol. Macromol. 93 (2016) 296–313, https://doi.org/10.1016/j.ijbiomac.2016.08.030.

[34] Y. Qian, X. Qiu, X. Zhong, D. Zhang, Y. Deng, D. Yang, S. Zhu, Lignin reverse micelles for UV-absorbing and high mechanical performance thermoplastics, Ind. Eng. Chem. Res. 54 (48) (2015) 12025–12030, https://doi.org/10.1021/acs.iecr.5b03360.

[35] L. Dehne, C. Vila Babarro, B. Saake, K.U. Schwarz, Influence of lignin source and esterification on properties of lignin-polyethylene blends, Ind. Crop. Prod. 86 (2016) 320–328, https://doi.org/10.1016/j.indcrop.2016.04.005, 0926-6690.

[36] M. Kiaei, B. Kord, R. Vaysi, Influence of residual lignin content on physical and mechanical properties of kraft pulp/pp composites, Maderas, Cienc. tecnol., Concepción 16 (4) (2014) 495–503, vnoct.

[37] R.R.N. Sailaja, M.V. Deepthi, Mechanical and thermal properties of compatibilized composites of polyethylene and esterified lignin, Mater. Des. 31 (9) (2010) 4369–4379, https://doi.org/10.1016/j.matdes.2010.03.046.

[38] S. Makri, A. Grigoropoulos, D. Bikiaris, I. Deligkiozi, A. Zoikis-Karathanasis, asignee Creative Nano P.C. Ultrasound-Assisted Process for the Production of Lignin Nanoparticles, p. European Patent Office, EP4471093A1, 2024.