Supplementary Material

for

Mechanical and ballistic performance of multilayer aramid laminates with nanofiller-reinforced HDPE/LDPE films

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METHODS

The production of the nanostructured polymeric films and multilayer composite plates consisted of four sequential stages: (i) production of cellulose nanofibers, (ii) preparation of the LDPE/CNF masterbatch, (iii) extrusion of nanostructured HDPE/LDPE films, and (iv) manufacture of multilayer composite plates by hot compression molding.

2.2.1 Production of cellulose nanofiber (CNF)

Initially, the bleached cellulose was dispersed in distilled water at a concentration of roughly 3 wt%. The suspension was subjected to grinding/defibrillation in a high-shear stone mill (MASUKO MKCA 6-2), with the gap between the grinding stones adjusted to 0.00–0.05 mm. This process aimed to generate nanoscale fibers through friction and contact. Nanofibrillation occurred through repeated mechanical shearing and friction between the stones. The cellulose suspension was continuously recirculated through the grinding chamber for 5 h at 2500 rpm using an integrated recirculation pump. This procedure was adapted from previous studies [1, 2]. The resulting aqueous CNF suspension was subsequently used for the preparation of the LDPE/CNF masterbatch.

2.2.2 Production of the LDPE/CNF masterbatch

After mechanical fibrillation, the CNF suspension was concentrated by centrifugation to approximately 5 wt.% solids. The concentrated suspension was then incorporated into molten LDPE using a Drais 600C high-shear homogenizer operating at 7500 rpm. The homogenization chamber temperature was maintained between 130 and 180 °C. Initially, 20 g of LDPE were melted for approximately 30 s, followed by the gradual addition of 60 g of concentrated CNF suspension. The mixture was homogenized for 450 s, during which the combined effects of high shear and elevated temperature promoted the preliminary dispersion of the cellulose nanofibers within the molten polymer, producing a LDPE/CNF masterbatch.

After homogenization, the material was cooled, ground in a knife mill (Primotécnica, model 1001), and used as the masterbatch for subsequent melt compounding. The CNF concentration in the masterbatch was determined by a mass balance considering the dry mass of cellulose incorporated into the molten LDPE after complete water removal during processing, according to Equation (1):

$$CNF\ (\%) = \frac{m_{CNF,dry}}{m_{LDPE} + m_{CNF,dry}} x\ 100$$

where $m_{CNF,dry}$ is the dry mass of cellulose present in the concentrated suspension and m_{LDPE} is the mass of LDPE used during homogenization.

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Based on this calculation, the resulting masterbatch contained approximately 14 wt.% CNF. The masterbatch pellets were subsequently diluted with HDPE during twin-screw extrusion to obtain the desired final CNF concentrations.

2.2.3 Production of nanostructured HDPE/LDPE films

For the development of nanostructured polymer films with the addition of cellulose (HDPE/LDPE/CNF), the masterbatch was added to HDPE at final CNF concentrations of: 0, 0.5, 1.0, and 1.5% (wt.%). Table 1 shows the identification and composition of the samples.

Prior to extrusion, HDPE pellets, LDPE pellets, and the LDPE/CNF masterbatch were dried at 80 °C for 12 h to remove residual moisture. The materials were then melt compounded in a co-rotating twin-screw extruder (MH Equipamentos, model COR-20-46-LAB). Twin-screw extrusion was selected because its distributive and dispersive mixing mechanisms promote homogeneous nanofiller distribution throughout the polymer matrix while minimizing particle agglomeration. The extruder featured temperature zones programmed at 100, 150, 160, 165, 170, 175, 175, 180, 180, 180, 180, and 170°C, in sequence, while operating at a screw speed of 200 rpm. The extruded strands were pelletized and dried again at 80 °C for 12 h before film production. The pellets were processed in a blown-film single-screw extruder (SEIBT Máquinas, model ES-35), operating at barrel temperatures of 170, 180 and 190 °C and screw speed of 56 rpm, producing continuous nanostructured HDPE/LDPE films.

Table 1. Nomenclature and composition of the polymeric films and multilayer plates.

	Polymeric films						Multilayer plates	
Sample	CNF % (m/m)	CLAY % (m/m)	GNP % (m/m)	CA % (m/m)	LDPE % (m/m)	HDPE % (m/m)	No. layers of aramid fabric	N° layers of film
HDPE/LDPE	-	-	-	-	14.0	86.0	-	-
HDPE/LDPE/0.5CNF	0.5	-	-	-	13.5	86.0	-	-
HDPE/LDPE/1.0CNF	1.0	-	-	-	13.0	86.0	-	-
HDPE/LDPE/1.5CNF	1.5	-	-	-	12.5	86.0	-	-
HDPE/LDPE/AR	-	-	-	-	14.0	86.0	20	21 (interspersed 0/90°)
HDPE/LDPE/CA/AR	-	-	-	2.0	14.0	84.0		
HDPE/LDPE/0.5CNF/AR	0.5	-	-	-	13.5	86.0		
HDPE/LDPE/1.0CNF/AR	1.0	-	-	-	13.0	86.0		
HDPE/LDPE/1.0CNF/CA/AR	1.0	-	-	2.0	13.0	84.0		
HDPE/LDPE/1.5CNF/AR	1.5	-	-	-	12.5	86.0		
HDPE/LDPE/1.0CLAY/CA/AR	-	1.0	-	2.0	13.0	84.0		
HDPE/LDPE/1.0GNP/AR	-	-	1.0	-	13.0	86.0		

Based on the mechanical and SHPB characterization of the HDPE/LDPE/CNF composites, the formulation containing 1.0 wt.% CNF was selected for comparison with the other nanofillers. Polymer films containing 1.0 wt.% organoclay (CLAY) and 1.0 wt.% graphene nanoplatelets (GNP) were produced following the same melt-compounding and blown-film extrusion procedure. Additionally, formulations containing 2 wt.% coupling agent (CA) were prepared to evaluate its influence on the mechanical and ballistic performance of the multilayer composites.

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2.2.4 Production of multilayer plates with aramid fabric

To develop the plates, the aramid fabric and the nanostructured polymeric films (Table 1) were previously cut to standard dimensions of 200 x 200 mm. 20 layers of aramid fabric were arranged for each plate made, a sufficient number to guarantee minimum ballistic protection [3, 4, 5].

Twenty layers of woven aramid fabric were interleaved with twenty-one polymeric films, resulting in a multilayer laminate. The woven aramid fabric maintained its original bidirectional (0°/90°) architecture throughout the laminate. The polymeric films, however, were alternately rotated by 90° between consecutive layers during stacking in order to minimize preferential orientation and promote a more uniform laminate architecture. Hot compression molding was carried out in a manual press with heating on the top and bottom of the sheets, with a fixed temperature of 150°C and a load of 7 t (≈ 1.7 MPa over the 200 × 200 mm area), for 30 min. Afterwards, the samples were cooled by cold compression in a press with water recirculation at around 20°C, for 10 min, at a compression pressure of 7 tons. The molding processing parameters were based on the methodology proposed by Tessari et al. [4]. The samples were identified according to Table 1, with AR in the nomenclature referring to aramid fabric.

2.3 Characterization

2.3.1 Hopkinson bar dynamic compression (SHPB)

The SHPB characterization of multilayer plates was carried out in the Materials Laboratory of the Army Technological Center (CTEx – Brazil). The samples were subjected to water jet cutting in specific dimensions of 14, 16 and 18 mm in diameter. (Supplementary Fig. S1); the 14 mm diameter provided the most adequate dynamic response and was adopted for the CNF series. For the nanofiller series (Section 3.2), specimens of 12 mm in diameter were used. After coming into contact with water, the samples were immediately dried in an oven at 80 °C for 12 h.

The SHPB test was carried out on equipment manufactured by the company Rel Inc. equipped with the PicoScope Series 5000® oscilloscope, from Pico Technology, and a 2300 System® signal amplification system, from Vishay Micro-Measurements. The incident bar and transmission bar were 2 m long. A set of bars with 9.53 mm in diameter and another with 19.05 mm were used. The striker bar was 203.2 mm long. The striker and the incident and transmission bars were made of 7075-T6 aluminum alloy. The striker bar was accelerated with compressed air at 20 psi (≈ 138 kPa) for the CNF series and at 15 psi (≈ 103 kPa) for the nanofiller series.

To evaluate the results, the incident pulse data (first peak of the time (s) vs volts (V) curve coming from the incident bar strain gauge), reflected pulse (second peak of the time (s) vs volts (V) curve coming from of the incident bar strain gauge) and transmitted pulse (first peak of the time (s) vs volts (V) curve coming from the transmitted bar strain gauge) were treated with Sure Pulse software.

2.3.2 Ballistic level test and subsequent deformation

The test to validate the ballistic level and subsequent deformation was carried out in accordance with NIJ Standard 0101.06 [6] and ABNT NBR 15000-2 [7]. The test was carried out at the Firing Line IV of the Army Assessment Center (CAEx – Brazil), using a test barrel to fire the projectile, a set of optical barriers, 1 meter apart, connected to a chronograph, which has the function of measuring the impact speed of the projectile on the sample; and a support for fixing the specimen (ballistic plate). A shot was considered valid when the incidence angle was less than 5°, the impact point was at least 50 mm from the edge of the panel or from another impact, and the impact velocity was within the tolerance of the protection level (426 ± 15 m/s, i.e., 411–441 m/s, for Level III-A according to NBR 15000-2). The impact velocity cannot be set individually for each shot: it is determined by the propellant (powder) charge and the projectile mass of the ammunition lot, and therefore shows a natural shot-to-shot dispersion. The velocity of each projectile was measured by the two optical barriers placed 1 m apart and connected to the chronograph, i.e., as the average velocity over this 1 m path ($v = \Delta x/\Delta t$), and the mean and standard deviation of the valid shots are reported for each formulation. Shots that exceeded the upper

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velocity tolerance without perforating the plate were also accepted, since a higher impact velocity implies a higher projectile kinetic energy ($E = mv^2/2$) and therefore a more severe condition than that required by the standard. The tests followed NIJ 0101.06 within the limitations of the test facility and were performed with 9 mm Luger FMJ ammunition (CBC; projectile mass of 8.0 ± 0.1 g; nominal impact energy of 726 J at 426 m/s) fired from a distance of 5–5.5 m. Two plates were tested per formulation, each receiving two to four valid impacts (four for HDPE/LDPE/AR and two for the other formulations), since the plate dimensions (200 × 200 mm) limited the number of impact points that respected the minimum distance of 50 mm; for comparison, NBR 15000-2 requires five impacts per panel for a formal Level III-A classification.

According to NIJ 0101.06, ballistic performance was classified as:

- NP (No Penetration): the projectile was completely stopped by the multilayer plate without perforation.
- PP (Partial Penetration): the projectile partially penetrated the armor system but did not completely perforate the last layer.
- CP (Complete Penetration): complete perforation of the armor system by the projectile.

The backface deformation (DP, depth of the indentation produced in the backing clay) was measured in the same impact tests described above, under Level III-A conditions, with the plates placed against a box containing 90 kg of Plastilina clay, a backing material widely employed to simulate the mechanical response of the human torso and to quantify the deformation transmitted by the projectile impact–[26]. The maximum acceptable backface deformation is 44 mm [24]. DP was measured for each valid impact and averaged per plate and then per formulation; for some formulations, only one valid DP measurement could be obtained.

The maximum backface deformation was measured using a FARO Freestyle 3D laser scanner (FARO Technologies Inc., USA). The scanner captures the three-dimensional geometry of the deformed clay surface by generating a point cloud through optical scanning. The acquired data were processed using SCENE software (FARO Technologies Inc.), which reconstructs the scanned surface and enables accurate determination of the maximum indentation depth. In the present study, only the maximum backface deformation (DP) was used as the quantitative parameter for evaluating the ballistic performance of the multilayer laminates, in accordance with the requirements of NBR 15000-2.

2.3.3 Tensile strength

The tensile strength test was carried out on an EMIC DL universal testing machine, with a capacity of 200 kN. The specimens were cut with a water jet to dimensions of 150 x 25 x ~7.8 mm. The test, based on the ASTM D3039-14 standard with adaptation of the ASTM A370 standard for steel, was carried out at a speed of 5 mm/min. Five specimens were tested per formulation in the tensile, flexural, and Charpy tests.

2.3.4 Flexural strength

The flexural strength test was carried out using a universal mechanical testing machine EMIC DL-2000, with a 200 kgf load cell. The specimens were cut with a water jet to dimensions of 127 x 12.7 x ~7.8 mm. The test, based on ASTM D790, was carried out at a speed of 3.7 mm/min.

2.3.5 Impact resistance

Unnotched Charpy impact tests were carried out with Veb Werkstoffprüfmaschinen Leipzig equipment (Germany), using the maximum pendulum energy of 294 J. The test specimens were cut with a water jet in dimensions of 63.5 x 12.7 x ~7.8mm. The test was based on ASTM D256-10 and ASTM E23-12C (adaptation).

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FIGURES

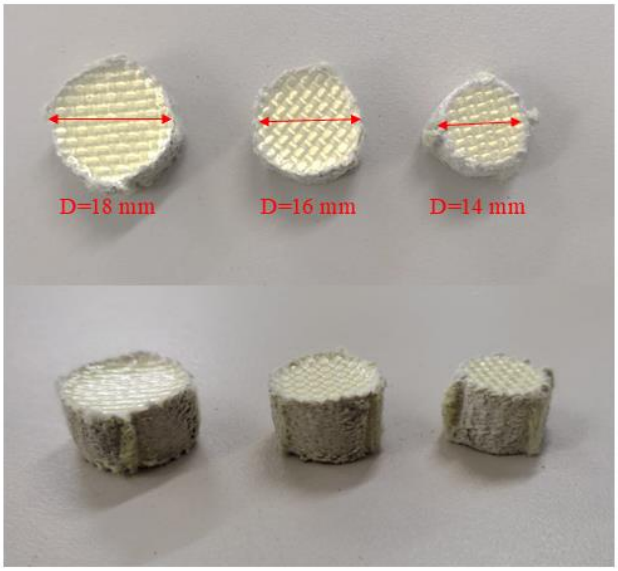
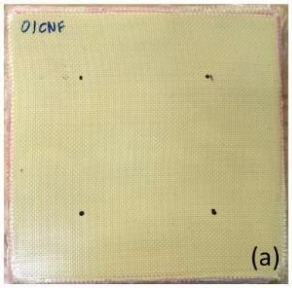
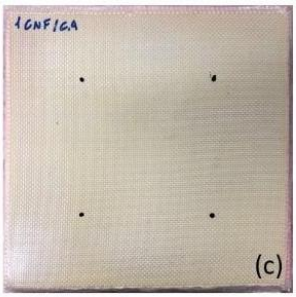
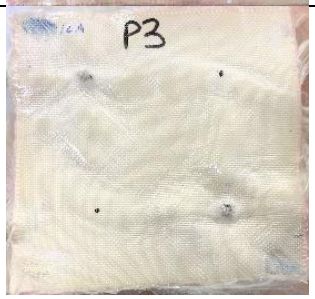


Fig. S1. SHPB specimens of the multilayer plates cut by water jet with diameters of 18, 16, and 14 mm (left to right). The 14 mm diameter was adopted for the CNF series; specimens of 12 mm were used for the nanofiller series.

Sample	Before ballistic impact	After ballistic impact
HDPE/LDPE/AR		
HDPE/LDPE/1.0CNF/CA/AR		

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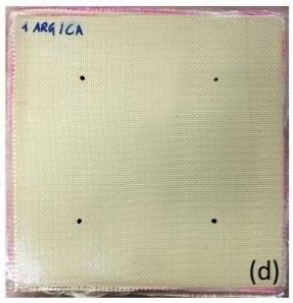
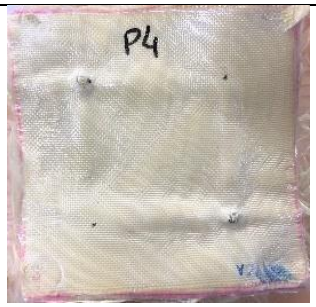
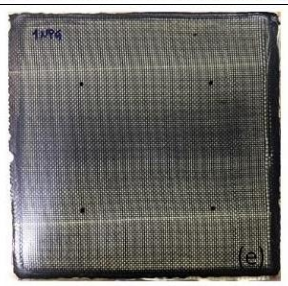
HDPE/LDPE/1.0CLAY/CA/AR		
HDPE/LDPE/1.0GNP/AR		

Fig. S2. Front face of the multilayer plates made with nanostructured HDPE/LDPE films (CNF, CLAY, and GNP), with and without coupling agent, and aramid fabric, before and after the ballistic impact. No images are available for the HDPE/LDPE/1.0CNF/AR plate.

TABLES

Table S1. One-way ANOVA (α = 0.05) of the backface deformation (DP) of the multilayer plates with HDPE/LDPE/CNF films (two plates per formulation, four valid measurements per formulation).

Formulation	**n**	**Mean DP (mm)**	**Variance (mm²)**
HDPE/LDPE/AR	4	25.50	3.53
HDPE/LDPE/0.5CNF/AR	4	25.50	5.19
HDPE/LDPE/1.0CNF/AR	4	26.15	1.53
HDPE/LDPE/1.5CNF/AR	4	25.90	0.76

Source of variation	**SS**	**df**	**MS**	**F**	**p-value**	**F critical**
Between groups	1.23	3	0.41	0.15	0.93	3.49
Within groups	33.03	12	2.75			
Total	34.26	15				

Since $p > 0.05$ and $F < F$ critical, the mean DP values of the four formulations are not significantly different.

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