ECO-RESPONSIBLE PARTS FOR AUTOMOTIVE INTERIORS MADE FROM NANOFIBRILLATED CELLULOSE (NFC) AND POLYPROPYLENE

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Abstract

The emergency due to current environmental changes mandates for immediate solutions to decrease fuel consumption and gas emissions of vehicles to further reduce their environmental impact. Transport sector is responsible for 25% of the total GHGs emitted, where the fingerprint of car weight is critical. Weight-lightening of car parts can be done, among other solutions, by reducing the use of petroleum-based materials while increasing the bio-sourced materials. Plastic-based composites represent 15% of materials used in vehicles. Their performance is often boosted by the addition of synthetic fibers or mineral fillers, both being high density materials. It is possible to lighten those composites by using eco-friendly bio-sourced materials without losing the mechanical and thermal performance of final parts. This work introduces biomaterial solutions, integrating renewable/sustainable and recycled materials, while having great potentials to compete with heavy compounds conventionally used for automotive interior parts.

Our paper presents the benefits of using Nanofibrillated Cellulose (NFC) fibrils to replace synthetic fibers and heavy mineral fillers for the manufacturing of more environmentally friendly biocomposite parts. NFC fibrils, nano-scale building blocks of micro and macro fibrils within wood fibers, consists in 100% cellulose macromolecules, a natural and renewable biopolymer. What is more, NFC fibrils have a lower density compared to usual fillers. We present here two efficient ways of using NFC fibrils in the fabrication of eco-parts. First, we present the fabrication of eco-nonwovens from NFC - NBSK mixtures in combinations to recycled polypropylene (rPP) fibers and recycled carbon fibers (rCF), followed by consolidation by compression molding and stamp-forming to fabricate eco-substrates. Secondly, biocomposites based on rPP and NFC - NBSK mixtures were obtained using the compounding process followed by injection molding to fabricate the same eco-substrates. The prototypes obtained from both processes presented mechanical performance (tensile, flexural, Izod impact), heat deflection temperature, humidity sensitivity, odor rating, and fire performance having excellent fit in automotive standards. Thus, NFC containing biocomposites can be considered as eco-choices to fabricate responsible, bio-sourced, lower-weight materials for utilization in headliners, dashboards, door panels, bolsters or other parts.

Background

In an effort to increase the sustainability of car parts, North American automotive industry is seeking cost-competitive alternative materials based on renewable and recycled resources, offering lower weight and at least equivalent performance compared to current parts. Laminated

structures are largely used in the construction of automotive interior parts, such as instrument panels, door bolsters, headliners, central consoles, etc.. They are manufactured by stacking, typically, three different layers: a bottom-layer consisting on an injection-molded composite, an inner layer of polymeric foam for haptics, and a top layer which is a polymer film for esthetics. Among these layers, the heaviest bottom one, made by injection-molding from a thermoplastic composite containing minerals or/and glass fibers, is the one to be replaced by a more eco-responsible one such a consolidated eco-nonwoven or/and cellulosic biocomposites.

Different processes are used in the manufacturing of nonwovens, such as dry- and wet-processes [1, 2]. The dry-process consists in cutting the staple fibers (synthetic or natural) at a length of 50 to 100 mm, followed by opening, cleaning, blending, web formation and folding, bonding by thermal, mechanical or chemical technics, and web finishing. Thermal bonding is the most used bonding method and is done by calendaring or compression molding. At the hot-pressing temperature, the polymer fibers will melt and coalesce creating points of contact amid the remained un-melted fibers. For example, nonwovens were obtained by dry-process using PP fibers and hemp fibers followed by thermal consolidation by compression at 190 °C. Optimal mechanical properties were obtained for nonwovens containing 40 - 50 wt.% of hemp fibers [3]. For kenaf / PP consolidated nonwovens designated for automotive interior parts, the nonwovens were produced by dry-process using 50 wt.% PP fibers and 50 wt.% kenaf followed by thermal bonding. Optimal mechanical properties were obtained in this case for very short consolidation times, i.e. 60 s at 230 °C [4]. Good acoustical properties were obtained by controlling the porosity by varying the compaction rate during the consolidation step. A very large porosity of 60 vol.% was required to reach acoustic properties, but, at such high porosity, the mechanical properties dropped drastically [5]. Different types of cellulosic fibers were tested in mixtures with polymer fibers to obtain nonwovens including jute, hemp, agave, coconut fibers, banana fibers [6-8]. Very limited number of studies exists on using the dry-process for the manufacture of nonwovens using mixtures of cellulosics fibers from pulp and paper industry, polymer fibers and carbon fibers [9].

Nonwoven wet-manufacturing process consists in similar steps as the dry-process but with some differences. The fibers mixing takes place in water to further form the nonwoven web by water extraction. Another difference is the length of raw fiber materials, i.e. they are much shorter that the ones used in dry-process to avoid fibers agglomerations and intermingling during water-mixing step. Wood fibers from pulp and paper industry, such as Northern Bleached Softwood Kraft (NBSK) have shown a promising potential for the fabrication of nonwovens by wet-process due to their short length, i.e. of 0.5 - 4 mm [10]. Moreover, the presence of 0.1 - 20 wt.% nanofibrillar cellulose in the aqueous mixture would generate cohesion between remaining synthetic fibers [11, 12]. Nonwovens were also obtained in an aqueous slurry containing carbon fibers and PP fibers, viscosity modifiers, surfactants, and flocculants [13, 14]. With this formulation, mats were fabricated containing up to 10 wt.% of carbon fibers (length of 3 - 25 mm) and up to 90 wt.% of binder fibers (PP or other thermoplastic fibers of 2 - 12 mm in length). Wet-laid process for flax fibers (as base) and polypropylene fibers (as binders) was applied to obtain nonwovens. These nonwovens have been processed by stacking eight individual flax-based sheets (70 wt.%) and PP-based sheets (30 wt.%) while applying moderate temperature and pressure. As the amount of binder PP fibers is relatively low, it is possible to obtain eco-friendly composites but they revealed high dependency of tensile and flexural strength on the total amount of PP fibers [15-17]. Only one invention exists on the use of cellulosic fibers (kenaf, among others), carbon fibers, and polymer fibers for the fabrication of nonwovens by wet-process [18] and a second one defines the compression molding parameters to be applied for their thermal bonding [19]. Thus, publications exist about the fabrication of thermoplastic / cellulosic fibers nonwovens based on two types of different fibers, but, as per our knowledge, limited studies were reported on using three or more different fibers.

A number of new techniques have been developed lately for producing high-tech, nano-structured celluloses. As a function of the manufacturing process used, they are ranging from the smallest cellulose nanocrystals (~10 nm x ~200 nm), to highly-fibrillated, high aspect ratio nanofibrillated cellulose NFC (~100 nm x 1000 nm), to more heterogeneous micro/nano fibrillated cellulose encompassing a range of different filaments dimensions. Performance BioFilaments Inc. (Vancouver, Canada), is actively manufacturing micro/nanofibrillated cellulose using a high-yield, low-capital technology. They produce a range of NFC products using a high-consistency refining step. Pulp fibers are fed into the refiner, where high shear forces pull apart the fibers, resulting in peeling and delaminating of the wood fibers walls. The obtained micro/nanofibrillated cellulose fibrils have exceptionally high aspect ratio and flexibility and can be used as reinforcing agents in thermoset and thermoplastic composites, for enhancing internal curing and structural properties in concrete [20, 21], in filtration media, and as rheology modifiers for drilling and industrial fluids [22, 23]. In this work, nanofibrillated and micro/nano-fibrillated celluloses were considered in nonwoven and biocomposites fabrication to emphasize their uniqueness and performance.

This paper presents two ways of manufacturing of environmentally friendly biocomposites layers containing eco-responsible materials such as: nanofibrillated cellulose (NFC), NFC / Northern Bleached Softwood Kraft (NFC - NBSK mixtures of 20 - 80), recycled carbon fibers (rCF), and recycled polypropylene (rPP, fibers and pellets). One way of manufacture of such eco-layers is by nonwovens formation and compression molding consolidation and the other one is by compounding followed by injection molding. We report here the performance of obtained eco-layers and prototypes at laboratory and high-scale levels. These eco-layers prove to have high potential to replace conventional PP- or ABS- composites layers, containing talc, glass fibers, or other synthetic fillers and reinforcements.

Materials and Methods

Nonwovens

NFC fibrils (8 wt.% suspension in water) and mixtures of NFC - NBSK (20 wt.% - 80 wt.%; 20 - 80) were supplied by Performance BioFilaments Inc. (Vancouver, BC, Canada). NFC fibrils have a width distribution range of 80 nm - 300 nm, a length distribution range of 100 µm - 2 mm and an aspect ratio up to 1200. NFC fibrils have a relative small mass while their surface area is large, i.e. up to 80 m^2/g. NFC - NBSK were supplied mixed in a proportion of 20 wt.% : 80 wt.%. The rCF, 13 mm in length and 5.1 µm in diameter, extracted from end-of-life carbon fiber composites, were bought from Carbon Conversions (Lake City, NC, USA). Recycled PP fibers of 5 Denier, 50 mm in length, and 30-40 µm in diameter, were bought from Drake Extrusion (Ridgeway, VA, USA). Hemp fibers from Plains Industrial Hemp Processing Ltd. (Gilbert Plains, Manitoba, Canada) and kenaf fibers from Sunstrand (Louisville, KY, USA) were used in selected formulations. The rPP fibers, hemp or kenaf fibers were cut at 6 mm length before using them in the wet-process. Otherwise, all the fibers were used as received in wet- and/or dry-process. Morphologies from scanning electron microscopy (SEM) and optical microscopy (OM) of main fibers used in this work are presented in the Figure 1. The combinations, at high level, of fibers in the nonwovens obtained from dry-process are presented in the Table I and obtained from the wet-process in the Table II. More details on the formulations will be shown further in the Tables IV, V and VI.

Dry- and wet-processes were applied first at NRC's laboratory scale for the fabrication of nonwovens of 1200 g/m^2. The difference between the two processes consisted in the step of fibers mixing, i.e. in air vs. in water respectively. The mixing of the 3 types of fibers for dry-process was done using pressurized air in a closed chamber. The mixing of the 3 types of fibers in water was done using an Adirondack sheet mold device, Formax 12" x 12" (Adirondack Machine Corporation, Hudson Falls, NY, USA). The formulations of nonwovens was kept the same at 50

wt.% rPP / 15 wt.% rCF / 35 wt.% cellulosics, where only the cellulosics combinations and types were varied. Figure 2 presents, as example, the physical aspect of a nonwoven obtained in wet-process at laboratory scale. Thermal bonding was done by compression molding using a Wabash compression press (Wabash, IN, USA). The nonwovens consolidation down to 2 mm thickness was done at 230 °C in 2 steps: (1) at 0 tons for 4 min for heating and equilibration and (2) applying up to 6 tons for 1 minute for consolidation. Prototyping of nonwovens at large scale was done only by dry-process, more precisely by air-laying. The air-laid nonwovens were consolidated to produce parts at our laboratory but also with the help of our industrial partners. Selected formulations containing hemp, kenaf fibers were also obtained for comparison purposes. CNC was used to cut down specimens from lab and industrial prototypes for performance evaluation.

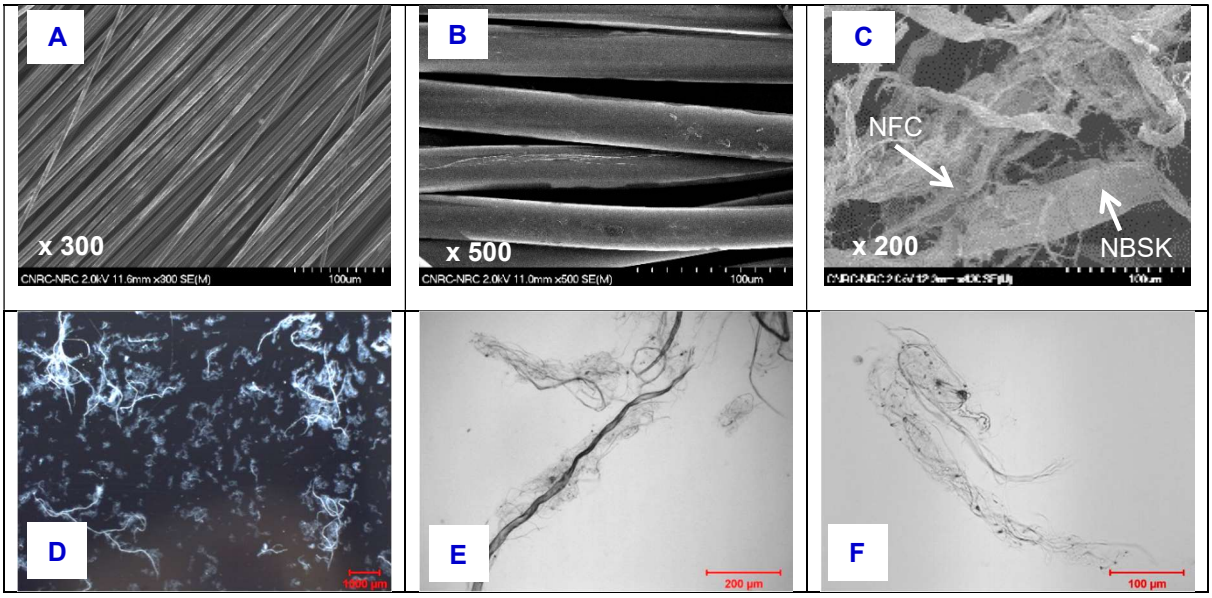

Figure 1: SEM and OM micrographs representing: a) rCF fibers (x300), b) rPP fibers (x500), c) NFC - NBSK 20 - 80 dry mixtures (x200), d) & e) NFC - NBSK 20 - 80 wet mixtures and f) NFC nanofibrils alone

Table I: Types of formulations of the nonwovens obtained by dry-process at lab scale

1200 g/m² nonwovens from Dry-process			
rPP (wt.%)	rCF (wt.%)	*Kenaf or Hemp (wt.%)	NFC - NBSK (wt.%)
50	15	35	-
50	15	20	15
50	15	-	35

Table II: Types of formulations of the nonwovens obtained by wet-process at lab scale

1200 g/m² nonwovens from Wet-process				
rPP (wt.%)	rCF (wt.%)	*Kenaf or Hemp (wt.%)	NFC - NBSK (wt.%)	NFC suspension (wt.%)
50	15	35, 30, 25	-	0, 5, 10
50	15	20	15	-
50	15	-	35	0
50	15	-	25	15

Figure 2: Physical aspect of one nonwoven containing 35 wt.% NFC - NBSK 20 – 80 obtained in wet-process at laboratory scale

Biocomposites

rPP, PP Repro C028-02, was bought from Group Lavergne (Qc., Canada). The extrusion line used to compound the biocomposites was a Coperion 34 mm co-rotating twin-screw extruder (Houston, TX, USA), having 12 mixing zones, and L/D ratio of 40. The screw configuration was specially designed by our team to assure an adequate dynamic in the barrel with the purpose to disperse and distribute uniformly the NFC - NBSK 20 - 80 mixtures in the polymer melt. A die strand, with 6 holes of 2 mm diameter each, was used at the exit of the extrusion line. The compounded formulations of rPP biocomposites are presented in the Table III. One formulation with 20 wt.% hemp was also compounded for comparison purposes. Performance values of commercial grades PP / 20% and 40% talc, Accutech HP0344T20L and Accutech HP0344T40L (from RTP Co., Winona, MN, USA) were taken from their TDS sheets and used as references.

Biocomposites pellets were first dried and then injection molded using a 34 ton Boy press (BOY Machines, Exton, PA, USA). Standard specimens were molded according to ASTM D638, ASTM D790, ASTM D256, and ASTM D648 for tensile, flexural, Izod impact, and heat deflection temperature evaluation, respectively. Prototyping of biocomposites parts took place at large scale with the help of our industrial partners. For performance evaluation, CNC was used to cut down specimens from the obtained parts.

Table III: Formulations in terms of feeding rates (for a total flow rate of 20 kg/h) of compounded and injected biocomposites based on rPP and NFC – NBSK 20 wt.% - 80 wt.% mixtures

Name	Recycled PP (kg/h)	Coupling Agent (kg/h)	NFC - NBSK (kg/h)
10% NFC - NBSK	18,00	0,00	2,00
20% NFC - NBSK	16,00	0,00	4,00
30% NFC - NBSK	14,00	0,00	6,00
10% NFC - NBSK + CA	17,00	1,00	2,00
20% NFC - NBSK + CA	15,00	1,00	4,00
30% NFC - NBSK + CA	13,00	1,00	6,00

Performance Evaluation

Consolidated nonwovens, biocomposites, and prototypes were characterized using following standard methods: tensile (ASTM D638), flexural (ASTM D790), Izod notched impact (ASTM D256), heat deflection temperature HDT (ASTM D648), coefficient of thermal diffusivity (Flash Method), odor rate (LP-463KC-09-01), water absorption (ASTM D5529) and fire resistance (FMVSS 302 - Federal Motor Vehicle Safety Standard No. 302, Flammability of Interior Materials).

Results and Discussions

Nonwovens fabrication and characterization at laboratory scale

In this first part of our study, 8 wt.% suspensions of pure NFC fibrils and NBSK - NFC 80 wt.% - 20 wt.% (80 - 20) dry mixtures were considered in nonwoven fabrication by wet-process with the purpose to determine their effect on the performance of consolidated nonwovens. Table IV presents details about the nonwoven formulations that were obtained for this purpose. All the nonwovens contained 50 wt.% rPP fibers, 15 wt.% rCF, and 35 wt.% of cellulosic fibers (i.e. hemp, or NFC - NBSK 20 - 80 mixture, both in combinations with pure NFC fibrils). Nonwovens containing 35 wt.% hemp fibers were obtained also by replacing 5 and 10 wt.% of hemp by pure NFC fibrils and by replacing 15 wt.% of hemp by the dry mixture NFC - NBSK 20 - 80. Nonwovens containing 35 wt.% NFC - NBSK were also obtained by replacing 15 wt.% from dry NFC - NBSK 20 - 80 mixture by pure NFC suspensions. All nonwovens were consolidated by compression molding down to 2 +/- 0.03 mm thickness and the specimens for characterization were cut down by CNC following tensile, Izod impact and HDT standards dimensions. The specimens were conditioned 40 hours, at 25 °C and 50% humidity before performance evaluation. Tensile properties, available from references [18] and [19] were also included in Table IV for comparison purposes, being nonwovens that address similar needs for interior automotive parts.

Table IV: Formulations and performance of nonwovens fabricated by wet-process at laboratory scale containing NFC fibrils in a form of NFC 8 wt.% suspensions and NFC - NBSK 20 - 80 dry mixtures

Formulations - 1200 g/m² 50 wt.% rPP / 15 wt.%rCF +…	TS (MPa)	TM (MPa)	IS (kJ/m²)	HDT (°C)
+ 35 wt.% cellulosic fibers, kenaf Ref. [18, 19] - Reference	16 - 29	2050 - 2300	-	123 - 153
+ 35 wt.% Hemp	25,5 (13)	5277,8 (1506)	17,5 (6)	135 - 145
+ 30 wt.% Hemp / 5 wt.% NFC (8% suspension)	26,8 (9)	5243,1 (500)	**19,1 (6)**	
+ 25 wt.% Hemp / 10 wt.% NFC (8% suspension)	19,2 (2)	2868,5 (685)	**28,1 (9)**	
+ 20 wt.% Hemp / 15 wt.% NFC - NBSK 20 - 80	23,7 (4)	2439,0 (552)	**26,2 (5)**	
+ 35 wt.% NFC - NBSK 20 - 80	24,4 (4)	2362,6 (125)	**24,9 (7)**	
+ 20 wt.% NFC - NBSK 20 - 80 / 15 wt.% NFC (8% suspension)	18,4 (1)	2183,0 (468)	**28,0 (5)**	

Replacing 5 wt.% of hemp by equivalent NFC content from 8 wt.% water suspension slightly increased the mechanical performance of the consolidated nonwoven while, when 10 wt.% of pure NFC suspension was used to replace 10 wt.% of hemp, a clear boost of 61% of the impact strength (IS), from 17,5 to 28,1 kJ/m^2, was obtained. An increase of 35% in IS, from 17,5 to 26,2 kJ/m^2, was also observed when 15 wt.% of hemp was replaced by the dry NFC - NBSK 20 - 80 dry mixture (i.e. at only 3 wt.% pure NFC). Improvement in IS was as well observed for the

nonwoven containing 35 wt.% NFC - NBSK dry mixture (IS of 26,2 kJ/m^2) when 10 wt.% from this dry mixture was replaced by 15 wt.% pure NFC suspension (IS of 28 kJ/m^2). It is obvious that the NFC fibrils have an important role in strengthening the consolidated nonwovens. The tensile strength (TS) of NFC containing nonwovens remained in the same range or slightly decreased compared to the 35 wt.% hemp nonwoven. The tensile modulus (TM) was gradually decreased by increasing NFC content but remained similar and even superior to the reference one. All HDT values were evaluated between 135 to 145 °C for all NFC containing nonwovens, all being superior to 90 °C, the standard recommended by automotive industry being the maximum temperature resistance for car interior parts based on polypropylene. Therefore, adding NFC fibrils to a mixture of fibers in a nonwoven wet-process generates increased cohesion between the components fibers. Increasing the NFC content increases the strength of a wet-laid nonwoven.

Selected formulations of nonwoven were fabricated in a 2nd round of wet- and, as well as in a dry-process at laboratory scale and they are disclosed in the Table V. Nonwovens containing 50 wt.% rPP / 15 wt.% rCF / 35 wt.% NFC - NBSK 20 - 80 were obtained by both processes. Moreover, variants of the nonwovens were obtained in dry-process contained also Pumice and PP film as double-side coverings. The formulation of the nonwoven obtained in the wet-process included only a double-side covering of PP film (Pumice couldn't be used in the wet-process due to its innate high water absorption). Pumice, SafSil® CT-550 grade (CR Minerals, Southlake, TX, USA) is a powdered volcanic rock that has a highly vesicular internal structure and is recommended to absorb odors and to increase the mechanical properties of plastics. It had particles of size of 5 µm and only 5 g were added in the mentioned formulations. PP film, BHP2V Performance Coex CPP grade (COPOL International Film Ltd., North Sydney, NS, Canada) it is 3-layer film heaving a sealant exterior layer. Nonwovens were covered on the two sides by a total of 5-7 g of film before their consolidation. The sealant layer was kept at the exterior of the nonwovens. Pumice and PP film solutions were selected with the purpose to observe their effect on the odor reduction (generated by cellulosics) and in reducing the humidity absorption of consolidated nonwovens.

Table V: Optimized formulations of nonwovens obtained from dry-process and wet-process at laboratory scale and their mechanical performance after consolidation

Formulations 1200 g/m^2	TS (MPa)	TM (MPa)	Thickness (mm)	IS (kJ/m^2)
50 wt.%PP / 15 wt.% rCF /35 wt.% cellulosic fibers [18, 19]	16 - 29	2050-2300	-	14 - 24
Consolidated nonwovens from dry-process (addition of Pumice & PP film)				
50 wt.%PP / 15 wt.%rCF / 35 wt.% NFC - NBSK	23,2 (7)	3226 (963)	2,1 (0,04)	24,5 (3,6)
50 wt.%PP / 15 wt.%rCF / 35 wt.% NFC - NBSK + Pumice	35,8 (6)	4794 (1397)	2,0 (0,05)	33,5 (3,7)
50 wt.%PP / 15 wt.%rCF / 35 wt.% NFC - NBSK + PP Film	39,6 (9)	3322 (978)	2,0 (0,03)	37,4 (8,0)
Consolidated nonwovens from wet-process (addition of PP film)				
50 wt.%PP / 15 wt.%rCF / 35 wt.% NFC - NBSK	16,5 (2,8)	2766 (390)	1,8 (0,1)	22,1 (5,1)
50 wt.%PP / 15 wt.%rCF / 35 wt.% NFC - NBSK + PP Film	20,6 (7)	2815 (250)	1,8 (0,1)	27,5 (11)

As can be seen in the Table V, the nonwovens obtained in the dry-process presented

important increments in TS, i.e. from 23,2 MPa up to 35,8 MPa for Pumice formulation and up to 39,6 MPa for PP film formulation while the TM remained at relatively same level (standard deviation was taken into account). The IS of nonwovens obtained from dry-process disclosed an important increment, from 24,5 kJ/m^2 up to 33,5 kJ/m^2 for Pumice formulation and up to 37,4 kJ/m^2 for PP film formulation. Similar effect was obtained for PP film nonwoven formulation resulted from the wet-process. All these mechanical properties are superior to those used as references. Mechanical performance evaluation demonstrated exceptional properties for our consolidated nonwovens containing 35 wt.% NFC - NBSK 20 - 80 mixtures. The effects of the presence of Pumice and PP film on consolidated nonwovens on the humidity uptake and odor rating were evaluated further.

ASTM D5229, “Standard Test Method for Moisture Absorption Properties and Equilibrium Conditioning of Polymer Matrix Composite Materials” was used to determine the moisture absorption behavior of the nonwovens. The test specimens were cut in squares of 6 x 6 cm^2 from the consolidated nonwovens and 3 specimens were tested for each formulation. They were dried in an oven for 48 h at 50 ºC and immediately weighed to the nearest 0.0001 g. The specimens were conditioned for 180 hours in an oven at 40 ºC and 90 % RH. The weightings were repeated each hour for the first day of testing and once a day for the remaining time (at the same hour). Excepting the nonwovens containing 35 wt.% NFC - NBSK mixtures, nonwovens containing 35 wt.% kenaf and 35 wt.% hemp, fabricated in the same conditions in our laboratory, were also tested. The results are presented in the Figure 3. First, it is obvious that kenaf and hemp containing nonwovens presented higher humidity uptakes than NFC - NBSK 20 - 80 containing nonwovens (i.e., at equilibrium, around 8.5 wt.% and 7 wt.% respectively, compared at 5.5 - 6 wt.%). At the addition of Pumice and PP film, the humidity uptake of NFC - NBSK containing nonwovens was further decreased. Therefore, our optimization using Pumice and PP sealant films in the consolidated nonwoven, not only improved the mechanical performance but also decreased their humidity uptake. More optimization is to be done to cut more this humidity uptake.

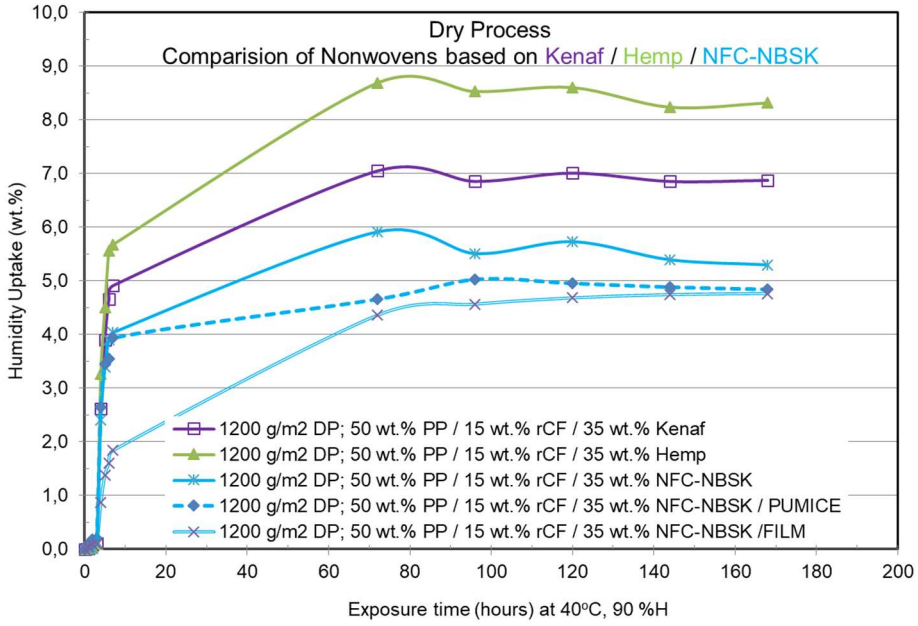

Figure 3: Humidity uptake evaluation following ASTM D5229 for 1200 g/m^2 nonwovens fabricated in dry-process at laboratory scale

The odor evaluation was done on consolidated nonwovens obtained in dry- and wet-process at laboratory scale. The odor testing was done using the laboratory procedure LP-463KC-09-01. The standard test temperatures were room temperature and 85°C. Test specimens from consolidated nonwovens were cut to have a surface area of 250 cm^2. An odor panel of 6 individuals was selected to evaluate the odor of specimens. Prior to testing at 23 ºC, the specimens were conditioned 24 hours at 23°C and 50% RH. Prior to testing at 85 ºC the test specimens were conditioned for 1 hour at 85°C and 50% RH. Test jars (including lids and rings) were cleaned to avoid contamination and were kept in an oven at 85°C for 1 hour. This procedure defines the maximum acceptable odor rating of 6, from a maximum of 10. Table VI presents the results from odor rating evaluation. A first observation is that the consolidated nonwovens containing 30 wt.% hemp always presented an odor rating higher than the 35 wt.% NFC - NBSK consolidated nonwovens, no matter what were the fabrication processes or rating temperatures. The odor ratings from Table VI are also demonstrating that the use of Pumice or PP film presented a positive effect on reducing the odor ratings.

Table VI: Results of odor evaluation of optimized nonwovens obtained by wet- and dry-process at laboratory scale

Nonwovens from wet-process	23 ºC	85 ºC
Empty Jar	1,1	2,1
1200 g/m² – 50 wt.% rPP / 15 wt.% rCF / 35 wt.% Hemp	4,8	5,8
1200 g/m² – 50 wt.% rPP / 15 wt.% rCF / 35 wt.% Hemp + Film	3,1	4,6
1200 g/m² – 50 wt.% rPP / 15 wt.% rCF / 35 wt.% NFC-NBSK	4,4	5,3
1200 g/m2 – 50 wt.% rPP / 15 wt.% rCF / 35 wt.% NFC-NBSK + Film	3,0	4,3
Nonwovens from dry-process	**23 ºC**	**85 ºC**
1200 g/m² – 50 wt.% rPP / 15 wt.% rCF / 35 wt.% Hemp	4,8	5,8
1200 g/m² – 50 wt.% rPP / 15 wt.% rCF / 35 wt.% Hemp + Film	3,3	5,3
1200 g/m² – 50 wt.% rPP / 15 wt.% rCF / 35 wt.% Hemp + Pumice	2,9	4,9
1200 g/m² – 50 wt.% rPP / 15 wt.% rCF / 35 wt.% NFC-NBSK + Film	2,2	4,7
1200 g/m² – 50 wt.% rPP / 15 wt.% rCF / 35 wt.% NFC-NBSK + Pumice	2,9	4,4

The main conclusions for this first part of our study on using NFC pure fibrils and NFC - NBSK 20 - 80 dry mixtures in the formulations of consolidated nonwovens fabricated by dry-process or wet-process are:

- NFC fibrils, from 8% suspensions, or from NFC - NBSK 20 - 80 dry mixtures can be easily used in the nonwoven fabrication processes.
- NFC fibrils have a very important role on increasing the performance of consolidated nonwovens; they were at least similar or superior compared to hemp or kenaf fibers (for equivalent contents in nonwoven formulations).
- Impact properties of consolidated nonwovens are highly increased in the presence of NFC fibrils.
- NFC fibrils possibly create cohesion between the components fibers in a nonwoven.
- The use of NFC fibrils, from 8% suspensions or from NFC - NBSK dry mixtures, in the nonwovens helps to reduce the overall odor rating of the final part, compared to hemp

of kenaf containing nonwovens.

- Use of NFC fibrils, from 8% suspensions or from NFC - NBSK dry mixtures, in the nonwovens helps to reduce the overall humidity or water uptake of the final part, compared to hemp or kenaf containing nonwovens.

Biocomposites fabrication and characterization at laboratory scale

Biocomposites having recycled polypropylene rPP as matrix and containing 10, 20, and 30 wt.% of NFC - NBSK 20 - 80 dry mixtures were fabricated by compounding without and with coupling agent. Table VII contains the description of the compounded formulations, their evaluated performance and, as well, the performance of rPP alone and of two commercial references for comparison purposes.

Table VII: Performance of biocomposites rPP / NFC - NBSK 20 - 80 obtained in compounding and injection molding

Name	TS (MPa)	TM (MPa)	ε at yield (%)	FS (MPa)	FM (MPa)	IS (kJ/m²)	HDT (°C)
Ref. rPP REPRO C028-02	20,0	1245	NA	33,2	922	4,7	70,0
Ref. PP / 20% talc Accutech HP0344T20L	35,0	1800	6,0	45,0	2400	4,0	63,0
Ref. PP / 40% talc Accutech HP0344T40L	30,0	2600	4,0	45,0	3600	3,0	70,0
rPP / 10 wt.% NBSK - CF 20 - 80	21,3	1728	6,88 (0,01)	35,8	1274	4,1	82,7
rPP / 20 wt.% NBSK - CF 20 - 80	22,7	2181	8,44 (0,2)	39,8	1720	4,5	83,4
rPP / 30 wt.% NBSK - CF 20 - 80	23,5	2808	9,96 (0,5)	42,2	2337	4,6	93,1
rPP / 10 wt.% NBSK - CF 20 - 80 / + CA	**27,8**	**1742**	**7,51 (0,2)**	**50,0**	**1585**	**4,6**	**72,6**
rPP / 20 wt.% NBSK - CF 20 - 80 / + CA	**31,4**	**2205**	**9,13 (0,1)**	**50,2**	**1705**	**5,2**	**84,8**
rPP / 30 wt.% NBSK - CF 20 - 80 / + CA	**36,8**	**2940**	**11,45 (0.2)**	**66,3**	**2591**	**5,2**	**103,1**

The first observation is that the performance of rPP increased with the content of NBSK - CF 20 - 80 dry mixture. More, the addition of the coupling agent (CA) boosted the biocomposite performance, testifying its efficacy. Important increments can be observed: from 20 MPa up to 36,8 MPa for TS, from 1245 MPa up to 2940 MPa for TM, from 33,2 MPa up to 66,3 for FS (a 3 fold increment), and for HDT and increment from 70 ºC up to 103 ºC. The elongation at yield of our biocomposites, i.e. the maximum reversible deformation that a material can sustain prior to irreversible deformation and fracture, was more important when compared at commercial compounds. This would be explained by the flexibility of cellulosic macro/micro fibrillated cellulose compared at the rigidity of glass fiber from the commercial compounds.

All these biocomposites presented nice performance and, therefore, the formulation rPP / 20 wt.% NBSK - CF 20 - 80 / + CA was selected to produce parts at industrial level. 100 kg of this biocomposite was extruded in our laboratory for large-scale fabrication of injection-molded prototypes.

Prototyping at large-scale and performance evaluation of prototypes

Nonwovens were fabricated at industrial level using a dry-process, more precisely an air laid web processing. Nonwovens of 1200 g/m^2 having a formulation of 50 wt.% rPP / 15 wt.% rCF / 35 wt.% NFC - NBSK 20 - 80 dry mixture were fabricated at Rando Machine Corporation (Macedon, NY, USA) using a 40 inch Rando SBD feeder and webber and an edge slitter. Figure 4 presents the physical aspects of the obtained nonwovens.

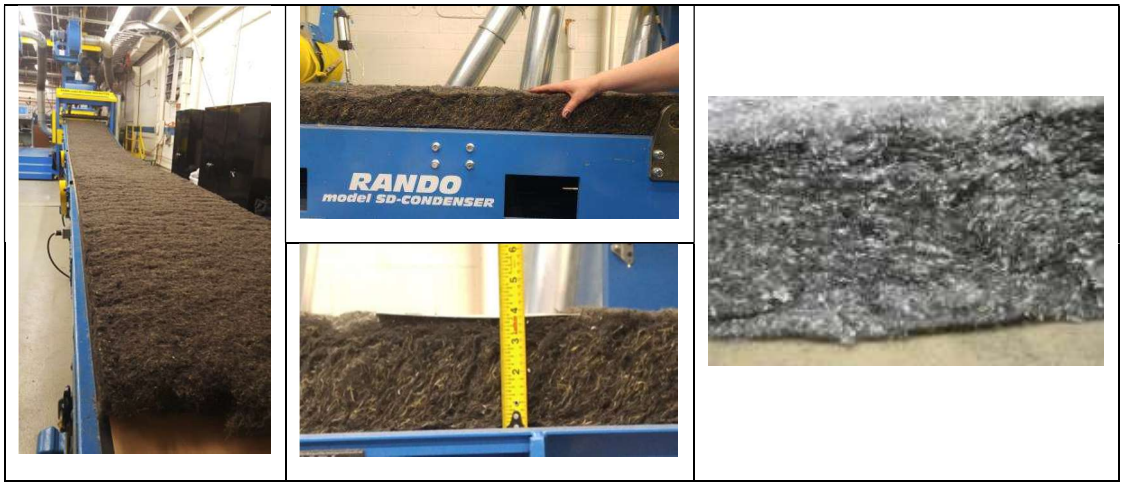

Figure 4: Nonwovens fabricated at industrial scale by Air Laid Web Processing

Two different processes were used to fabricate prototypes using the 1200 g/m^2 nonwovens containing 50 wt.% rPP / 15 wt.% rCF / 35 wt.% NFC - NBSK 20 - 80 dry mixtures. The 1st one was a NRC's stamping process using a 1250 ton capacity press. A generic side-door impact beam was fabricated. Images of the process and of final stamped part are presented in the Figure 5.

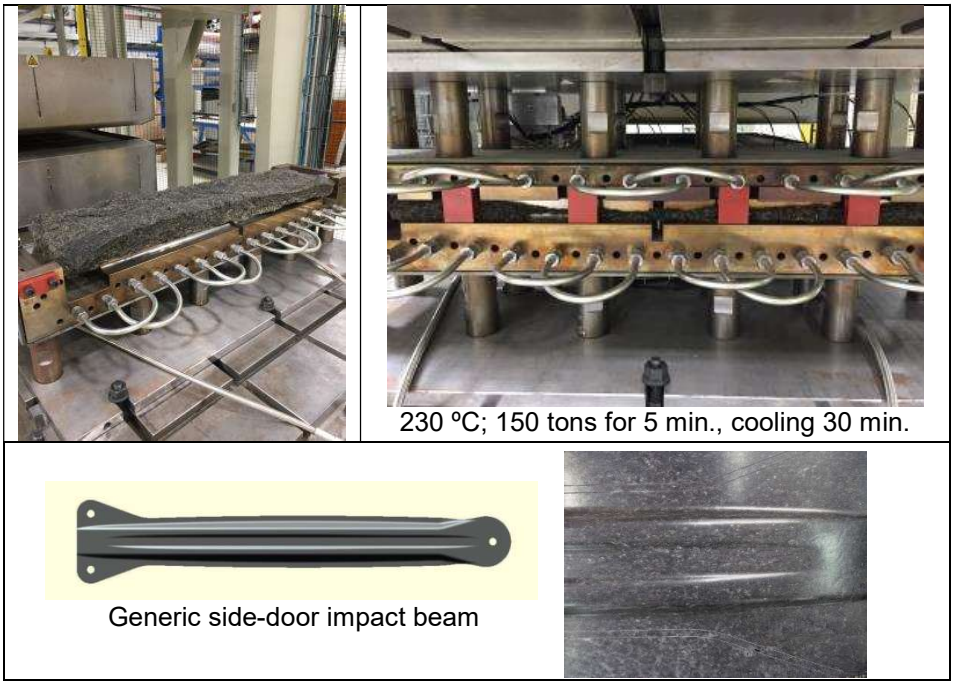

Figure 5: NRC's stamping process using a 1250 ton capacity press and the physical aspect of the final part

The 2nd process for nonwovens consolidation and prototyping was a two-steps process, i.e. using a press for pre-consolidation followed by a vertical lamination press to shape and laminate the final parts. Similar prototypes as for the nonwovens were fabricated from the biocomposites pellets by injection molding. Images of obtained laminated parts containing 50 wt.% rPP / 15 wt.% rCF / 35 wt.% NFC - NBSK 20 - 80 nonwovens and of injection molded parts based ob rPP / NFC - NBSK 20 – 80 biocomposites are shown in the Figure 6.

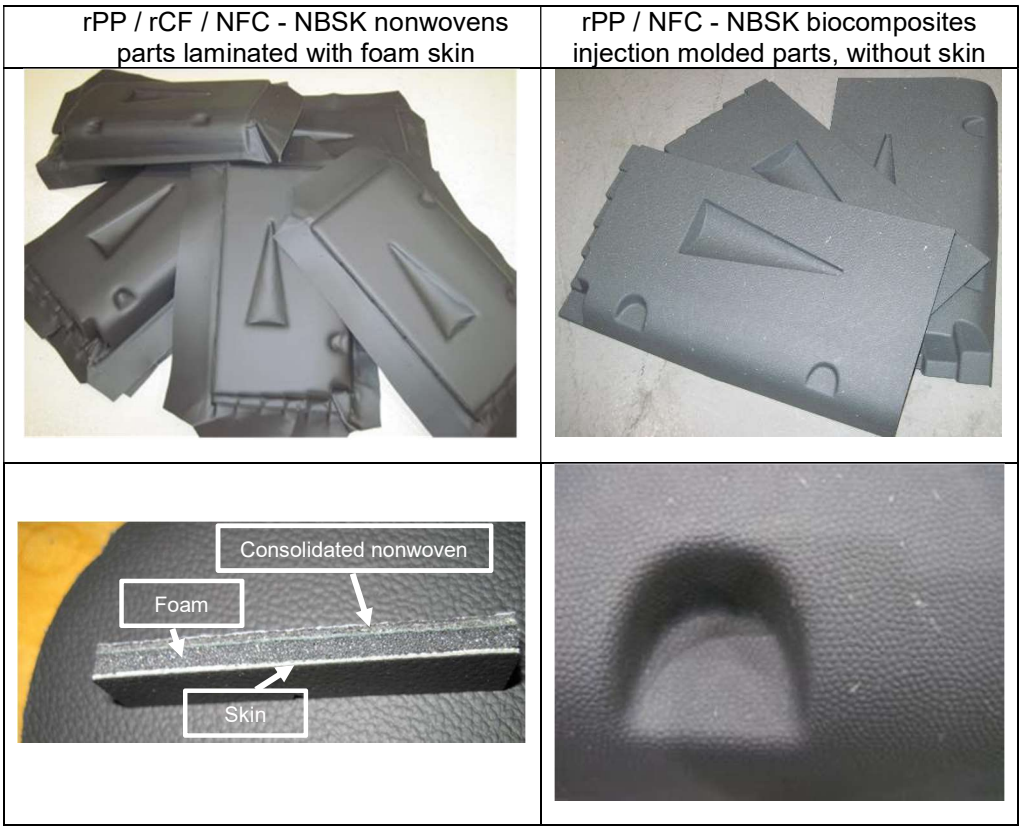

Figure 6: Prototypes obtained at large scale containing NFC - NBSK 20 - 80 dry mixtures: right - laminated parts containing 35 wt.% NFC - NBSK 20 – 80 dry mixtures nonwovens and left - injection molded parts made from rPP / 20 wt.% NBSK - CF 20 - 80 / + CA biocomposites

Table VIII: Mechanical performance of final parts, without skin, containing NFC - NBSK 20 - 80 dry mixtures

Name	TS (MPa)	TM (MPa)	IS (kJ/m²)	FS (MPa)	FM (MPa)
50 wt.% PP / 15 wt.% rCF / 35 wt.% cellulosic fibers [18, 19]	16 - 29	2050-2300	14 - 24	-	-
Prototypes, without skin, obtained from the 1200 g/m² nonwovens					
50 wt.% rPP / 15 wt.% rCF / 35 wt.% Hemp	25,8 (14)	5644 (619)	23,1	35,9 (5)	3022 (551)
50 wt.% rPP / 15 wt.% rCF / 35 wt.% NFC - NBSK	16,3 (3)	4817 (673)	36,8	42,7 (5)	2892 (379)
Prototypes, without skin, obtained from the biocomposites					
rPP / 20 wt.% Hemp / + CA	16,5 (2)	1689 (63)	4,6	43,8 (2)	1566 (133)
rPP / 20 wt.% NFC - NBSK / + CA	27,2 (2)	1844 (57)	5,5	58,2 (2)	1307 (53)

Specimens were cut-down at our laboratory from all prototypes for characterization purposes, to evaluate the tensile, Izod impact, flexural, thermal diffusivity coefficients and fire performance. Mechanical performance of final parts, without skin, containing NFC - NBSK 20 - 80 cellulosic mixtures are shown in the Table VIII. This table discloses similar or higher mechanical performance of the NFC-based prototypes obtained from 1200 g/m^2 nonwovens and of prototypes obtained from rPP biocomposites when compared to the reference.

Coefficients of thermal diffusivity were evaluated using Flash method at ambient temperature. Thermal diffusivity (α) of a medium is a thermo-physical property that determines the speed of heat propagation by conduction during changes of temperature. It is a function of sample thickness and the time of reaching a half of the maximal sample temperature increase. The measured values of thermal diffusivity coefficients for specimens cut from the NFC – NBSK 20 - 80 containing parts, fabricated at laboratory and large scale, along with some examples and references, are presented in the Table IX. It can be observed that very low values of thermal diffusivity were obtained for all NFC - NBSK containing parts and this indicates that our prototypes would be thermally insulated materials.

Table IX: Coefficients of thermal diffusivity evaluated using Flash method at ambient temperature

Name	$t_{1/2}$ (sec)	Thickness (mm)	Thermal diffusivity (m^2/sec)	Thermal diffusivity (mm^2/sec)
Consolidated 1200 g/m^2 nonwovens at laboratory scale (no skin)				
50 wt.% rPP / 15 wt.% rCF / 35 wt.% Kenaf	2.88	2.07	2.07E-07	0.207
50 wt.% rPP / 15 wt.% rCF / 35 wt.% Hemp	2.80	1.81	1.62E-07	0.162
50 wt.% rPP / 15 wt.% rCF / 35 wt.% NFC - NBSK	2.49	1.84	1.89E-07	0.189
Nonwoven based parts with skin fabricated at large scale				
50 wt.% rPP / 15 wt.% rCF / 35 wt.% Hemp	1.89	1.51	1.67E-07	0.167
50 wt.% rPP / 15 wt.% rCF / 35 wt.% NFC - NBSK	2.04	1.75	2.08E-07	0.208
Biocomposites parts without skin fabricated at large scale				
rPP / 20 wt.% Hemp / + CA	13.38	2.95	9.03E-08	0.090
rPP / 20 wt.% NFC - NBSK / + CA	11.72	2.91	1.00E-07	0.100
Examples of thermally insulating materials				
Wood (Yellow Pine)			8.20E-08	0.082
PP (Polypropylene) at 25 °C			9.60E-08	0.096
Examples of very thermally conductive material				
Air at 25 °C			1.90E-05	19.000
Pyrolytic graphite, parallel to layers			1.22E-03	1220

Fire testing, following the Standard document No. 302 from Transport Canada, was applied

to determine the fire resistance of parts fabricated at large scale. Each tested specimen had the dimensions of a rectangle of 102 mm wide by 356 mm long and thickness of maximum 4 mm. The specimens were mounted so that both sides and one end are held by the U-shaped frame situated in a metal cabinet (381 mm long, 203 mm deep, and 356 mm high) for protecting the test specimens from drafts. Each specimen was conditioned for 24 hours at a temperature of 21 ºC and a relative humidity of 50 % before testing. Specimen were exposed to the flame for 15 seconds and the initiate timing for testing started when the flame from the burning specimen reaches a point 38 mm from its open end. This method stipulates that a material shall not burn, nor transmit a flame front across its surface, at a rate of more than 102 mm per minute. Figure 7 shown physical aspects of the specimen cut off from a part with skin made in 50 wt.% rPP / 15 wt.% rCF / 35 wt.% NFC - NBSK 20 - 80 consolidated nonwoven. The results from fire resistance testing for all tested parts are presented in the Table X.

Part before test	Part after test Face with skin	Part after test Lateral
		

Figure 7: Physical aspects of the specimen cut off from a part with skin made in 50 wt.% rPP / 15 wt.% rCF / 35 wt.% NFC - NBSK 20 - 80 consolidated nonwoven: before and after fire testing

The results of fire performance are shown in the Table X. All the nonwovens pass the test. The biocomposites (without skin) did not pass, as expected, although the presence of skin would help.

Table X: Numerical values for fire performance of prototypes

Sample name	Burning Distance (mm)	Burning Time (seconds)	Burning Rate (mm/min.)	Suc-cess	Comments
Nonwoven containing parts, without and with skin, fabricated at large scale					
50 wt.% rPP / 15 wt.% rCF / 35 wt.% Hemp	75	228	19.74	Yes	Preserve integrity when burnt
50 wt.% rPP / 15 wt.% rCF / 35 wt.% Hemp + Skin	10	219	2.74	Yes	Self-extinguish
50 wt.% rPP / 15 wt.% rCF / 35 wt.% NFC-NBSK	75	191.5	23.50	Yes	Preserve integrity when burnt
50 wt.% rPP / 15 wt.% rCF / 35 wt.% NFC-NBSK, Skin	19.5	340	3.44	Yes	Self-extinguish
Biocomposites containing parts, without skin, fabricated at large scale					
rPP / 20 wt.% Hemp / + CA	75	179	25.14	No	X Fail
rPP / 20 wt.% NFC-NBSK / + CA	75	155.5	28.94	No	X Fail

Summary

NFC fibrils and NFC - NBSK 20 - 80 dry mixtures were successfully used in the fabrication of nonwovens containing 50 wt.% rPP / 15 wt.% rCF / 35 wt.% NFC - NBSK by wet- and dry-process. NFC-based consolidated nonwovens demonstrated excellent mechanical and thermal performance, higher than those obtained using hemp or kenaf, fact that gives them the opportunity to be used in automotive interior applications. The developed eco-substrates presented also low thermal diffusivities, between 0.09 mm^2 / sec (the one of wood) and 0.5 mm^2 / sec (the one of pure PP) and they passed the odor rating evaluation. NFC - NBSK 20 - 80 dry mixtures were also successfully used in extrusion and injection molding to produce biocomposites, therefore in the fabrication of a second type of eco-layer. Their mechanical and thermal performance were similar or higher when compared to commercial composite grades while being obtained based on a recycled polymer matrix and a bio-sourced reinforcement. The biocomposites passed also the test of odor evaluation and also proved, as the nonwoven eco-substrates, to be thermally insulating materials. Therefore, the cellulosic micro/nanofibrillated cellulose NBSK - NFC, recycled carbon fibers from end-of-life composite parts, and recycled PP were successfully used in the fabrication of consolidated nonwovens and biocomposites eco-layers for uses in automotive interior applications. The demonstrators manufactured using these eco-layers revealed performances at least equal compared to current materials used in automotive parts while being designed to have lower environmental impact.

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