

# Hot-compacted self reinforced polyamide 6 composite laminates



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## ABSTRACT

Polyamide 6 (PA6) based sheets are produced by a typical hot compaction procedure and characterized in terms of thermal properties, mechanical behavior and morphological features. Starting from two commercial clothes mainly constituted by woven PA6 fibers, single polymer composites (SPC<sub>s</sub>) involving 24, 30 and 36 layers of each fabric were prepared and investigated taking compression molded plates based on a commercial neat PA6 grade as the reference material. All materials were prepared under similar processing conditions in terms of pressure profile and duration of the various stages of compaction but using a higher consolidation temperature for the neat PA6 with respect to composite laminated systems. The improvement of both static and dynamic flexural moduli and the simultaneous reduction of the Charpy impact parameters, by increasing of the reinforcement effect, confirmed the typical need to balance stiffness and toughness performances, well established for conventional composite systems, unless optimized production process and/or hybridization approaches are considered. In fact, a good compromise of mechanical performances was detected for the thickest SPC laminates based on a hybrid PA6/polyurethane fabric and ascribed to the occurrence of more efficient failure mechanisms rather than the simple fiber breakage as witnessed by morphological analyses.

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## 1. Introduction

In the last decades, great efforts have been spent toward the development of new lightweight and increasingly high performance materials to be used in replacements of traditional ones such as wood, ceramics and metals for various industrial applications. As well established, outstanding functional and/or structural properties can be achieved by opportunely including in polymer matrices reinforcing fillers and/or fibers even if the interface strength still remains a traditional challenge. In this frame, an emerging group of composites, best known as single-polymer composites (SPC<sub>s</sub>) or all-polymer composites or self-reinforced composites (SRC<sub>s</sub>), overcomes this aspect because both the matrix and the reinforcement are of the same polymer or based on resins belonging to the same family [1–3]. The SPC concept, employed for the first time by Capiati and Porter in 1975 [4] to embed gel spun high modulus polyethylene (PE) fibers in lower melting point PE matrices, has been subsequently extended to many other polymers. The concept is essentially based on the intrinsic peculiarity of semi-

crystalline polymers involving crystallite with varying perfection and thus characterized by different melting temperatures. This feature is enhanced for thermoplastic polymers with the low crystallization temperature (i.e. polyesters) [5,6], by the eventual presence of stereogenic carbons, for resins able to crystallize in different polymorphic modifications [7] or by the contribution of appropriate changes to the molecular architecture such to affect their melting region.

Self-reinforced or single-polymer composite systems offer interesting performances mainly related to the improved adhesion of the fiber-matrix interphase and full recyclability without the need for separation of fibers and matrices with respect to traditional ones.

Currently, many papers are available about the hot compaction of highly oriented thermoplastic fibers [8–10] or woven oriented tapes [11] and specific research interest have been devoted to develop appropriate manufacturing processes [12,13] as well as to study and improve their performances by optimizing processing conditions and fiber orientation [14–16].

Polyamides are characterized by polymorphism [17] and, specifically for PA6, two different crystalline forms  $\alpha$  and  $\gamma$  can be obtained in different conditions of thermal treatment as was

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confirmed by Bhattacharyya et al. [18]. In particular, authors studied laminates obtained by stacking PA6 films prepared in different conditions, in order to favor in each layer the prevailing formation of one of the two mentioned morphologies, followed by compression molding of the defined sequence of stacked films. Obtained samples showed a two-melting signal and improved tensile properties with respect to the constituent PA6 films.

Seyhoglu et al. [19] investigated all-polyamide systems prepared by film stacking and including two different organophosphorus-based flame retardant additives: aluminum diethyl phosphinate (AlPi) and its mixture. Results demonstrated that these additives not affect tensile properties of considered composites and the AlPi is more effective than its mixture in terms of flammability results in all the studied polyamide based composites.

Gong et al. [20] analyzed the effect of processing temperature on tensile properties of SPCs based on recycled PA6 woven fibers highlighting a balance between retaining high orientation of PA6 fibers and forming a continuous fiber-matrix interface by hot compaction.

Dencheva et al. [21] reported on polyamide SPCs prepared by reactive processing. Specifically, they investigated PA6-based composites obtained through activated anionic ring-opening polymerization in terms of morphology, crystalline structure and tensile properties. Authors demonstrated the presence of an oriented transcrystalline layer on the surface of the reinforcing fibers and able to affect the interfacial adhesion strength and, consequently, the mechanical performances of products.

In this paper, the interest was focused on PA6 based samples constituted by 24, 30 and 36 layers, prepared by hot compaction and investigated in terms of thermal and mechanical properties taking compression molded plates of a neat unreinforced PA6 as the reference material.

In details, given that during the hot compaction process the polymer undergoes both a partial melting of the skin of fibers (reinforcement) into an isotropic phase which binds the structure together (matrix) and, simultaneously, a rearrangement or annealing of their internal crystalline structure, calorimetric tests have been performed for each sample to examine any morphological changes by evaluating the enthalpy of melting which, in turns, reflects their total crystallinity.

Flexural and Charpy impact tests were performed at room temperature and within one week from their preparation on adequately sized specimens cut from each composite plates stored under the same humidity conditions.

Finally, morphological observations taken on the surface of the base cloths and on fractured areas of composite specimens allowed to determine the actual fiber content and to gain insights about the involved damage mechanisms.

## 2. Experimental

### 2.1. Materials

Two commercially available polyamide 6 (PA6) based clothes were used as constituents of all investigated composite systems. In particular, a wear resistant 100% PA6 yellow fabric (density: 125 g/m<sup>2</sup>; mean fiber diameter: 22.92  $\mu$ m) coded as Y<sub>tq</sub> and a red fabric (99% polyamide 6 + 1% polyurethane, density: 78 g/m<sup>2</sup>; mean fiber diameter: 13.14  $\mu$ m) coded as R<sub>tq</sub>, the structure of which is indicated in Fig. 1, were considered as base materials.

A neat unreinforced polyamide 6, commercially supplied by Lanxess (Cologne, Germany) under the trade name Durethan B30S with a number average molecular weight of 40,000, a melt flow index of 125.5 g/10 min (260 °C/5 Kg – ISO 1133) and a density of 1.14 g/cm<sup>3</sup> was used as the reference material.

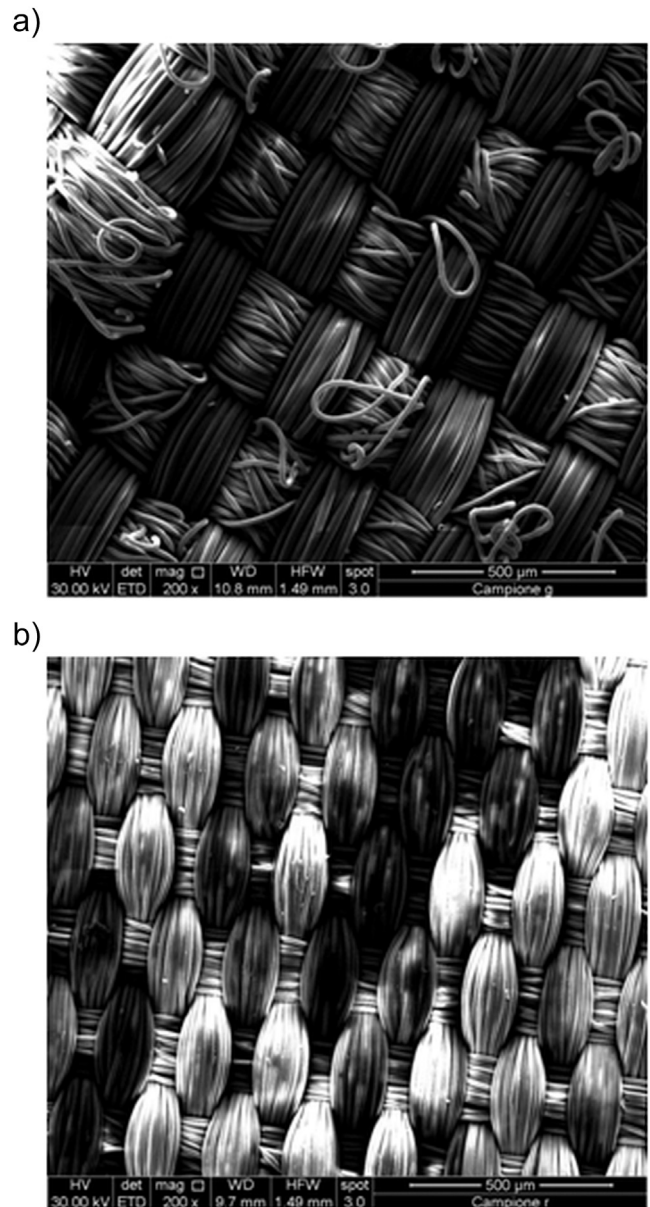


Fig. 1. Structure of PA6 based plain clothes. a) Yellow (Y), b) Red (R).

### 2.2. Sample preparation

Composite plates were prepared by stacking 24, 30 and 36 layers of each above mentioned clothes, predried in a vacuum oven at 80 °C overnight, and compacting the assembly with a hot press Mod LP420B (LabTech Engineering Company Ltd.) under carefully controlled temperature and pressure conditions.

At this regard, previous calorimetric investigations of considered PA6 based fabrics showed in each case the presence of a broad melting region (Fig. 2) with even a splitting in two signals in the case of the yellow cloth but responsible of a shoulder on the low-temperature side of the main melting signal for the red one. Given that, for both cases, the main melting peak is clearly centered at about 225 °C, for the hot compaction step, the temperature was fixed at 220 °C to ensure a selective melting of a thin skin on the surface of the oriented fibers (reinforcing phase) while a pressure of 80 bar was applied in light of previous authors' experiences. Specifically, the compaction procedure involved a preheating of

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