## Supplementary Information

The density $\rho_{\text {, }}$ the molar mass in weight $M_{w}$ and in number $M_{n}$, the apparent crystallinity ratio $\chi_{c}$, and the Newtonian viscosity $\eta^{*} 0$ at $240^{\circ} \mathrm{C}$ are reported in Table SI 1. In more details, the Gas Permeation Chromatography tests for PE and PEgMA were performed at $135^{\circ} \mathrm{C}$ in $1,2,4$-trichlorobenzene (TCB) [1] and for PA6 was done at $35^{\circ} \mathrm{C}$ in 1,1,3,3,3-hexafluoro-2-propanol (HFiP) [2]. The rheological frequency sweep tests were performed using Anton Paar MCR 502 rheometer equipped with a plate/plate geometry at $\gamma=1 \%$, in the linear domain, from $\omega=100$ to 0.01 rad. $\mathrm{s}^{-1}$, under nitrogen flow.

Table SI 1: main characteristics of PE, PEgMA and PA6 polymers

|  | $\rho$ | $M_{w}$ | $\boldsymbol{M}_{n}$ | $\eta^{*} 0$ |
| :---: | :---: | :---: | :---: | :---: |
| Polymer | o <br> $\left[\mathrm{g} . \mathrm{cm}^{-3}\right]$ | $\left[\mathrm{g} . \mathrm{mol}^{-1}\right]$ | $\left[\mathrm{g} . \mathrm{mol}^{-1}\right]$ | $\left(240^{\circ} \mathrm{C}\right)$ <br> $[\mathrm{Pa} . \mathrm{s}]$ |
| PE | 0.92 | 122,000 | 24,000 | $\sim 20,000$ |
| PEgMA | 0.92 | 85,000 | 21,000 | $\sim 2,000$ |
| PA6 | 1.18 | 124,000 | 55,000 | $\sim 5,000$ |

Table SI 2: thickness of multilayer films composed of 5 layers (0LME) 129 (5LME) 1024 (8LME) and 2049 (9LME) layers.

| Fraction | LME | $e$ | $\boldsymbol{n}_{\text {tot PA6 }}$ | $n_{\text {Pa6 }}$ | $\boldsymbol{e}_{\text {th PA6 }}$ | $e_{P A G}[n m]$ | $\begin{aligned} & \sigma_{\text {PAG }} \\ & {[n m]} \end{aligned}$ | $n_{\text {PE }}$ | $e_{\text {PE }}$ |  |  | $\phi_{\text {th PAG }}$ | $\phi_{\text {PAG }}$ |
| :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: | :---: |
|  |  | $[\mu \mathrm{m}]$ |  |  | [nm] |  |  |  | $\boldsymbol{e}_{\text {th PE }}[\mathrm{nm}]$ | [nm] | $\sigma_{\text {PE }}[\mathrm{nm}]$ |  |  |
| 25/50/25 | 0 |  | 1 | 4 | 23,630 | 21,720 | 3,290 | 8 | 38,180 | 31,270 | 1,600 | 23.6 | 25.8 |
| 25/50/25 | 5 |  | 32 | 20 | 738 | 766 | 200 | 14 | 2315 | 2590 | 500 | 23.6 | 23.5 |
| 25/50/25 | 8 | 100 | 256 | 68 | 92 | 119 | 51 | 71 | 298 | 395 | 117 | 23.6 | 23.1 |
| 25/50/25 | 9 |  | 512 | 70 | 46 | 46 | 17 | 69 | 149 | 162 | 38 | 23.6 | 22.1 |

The theoretical thicknesses were obtained with the followed equation:

$$
e_{t h x}=\frac{e_{t o t} \Phi_{x}}{n_{x}} \quad \text { Eq SI } 1
$$

The measured thickness is obtained by averaging the value obtained for $n$ measured layers. For example, the PA6 thickness in 5LME films is obtained by taking the average of 20 PA6 layer thicknesses measurement.

PA6 TM-DSC thermogram during first heating shows a large exothermic variation between 30 and 140 ${ }^{\circ} \mathrm{C}$, corresponding to the main physical amorphous chain relaxation (glass transition) and water evaporation (Figure SI 1), roughly estimated to $\sim 6 \%$ in PA6 film. The reversible flux highlights the glass transition while the non-reversible flow shows the water evaporation, at $\sim 100{ }^{\circ} \mathrm{C}$. The glass transition temperature $T_{8}$ is estimated to $\sim 50^{\circ} \mathrm{C}$, in agreement with literature data [3].

REVERSING HEAT FLOW endo up




NON REVERSING HEAT FLOW endo up





Figure SI 1: PE; PEgMA; PA6 and multilayers film $8 M$ TM-DSC thermograms left reversible heat flow right, nonreversible heat flow

Figure SI 2 shows the SAXS and WAXS diffractograms of 0 (a) and 8 (b) irradiated at $\lambda=0.775 \AA$ with X-ray beam along ND at $T=25^{\circ} \mathrm{C}$. At this temperature, the high concentration and the strong intensity of PE patterns hide those of PA6, as observed by Dencheva et al. [4]. The 2D SAXS and WAXS patterns exhibit the quasi-isotropic orientation of orthorhombic peaks of PE for the 0 LME film (Figure SI 2.a), while they are highly oriented for the 8 LME film (Figure SI 2.b). Based on the intensity $I$ azimuthal $\Psi$ profiles obtained from SAXS patterns at $T=25^{\circ} \mathrm{C}$, the Herman's orientation function [5] was calculated using the following equations:

$$
<\cos ^{2} \Psi>=\frac{\int_{0}^{\pi / 2} I(\Psi) \cos ^{2} \Psi \sin \Psi d \Psi}{\int_{0}^{\pi / 2} I(\Psi) \sin \Psi d \Psi}
$$

and

$$
f_{H}=\frac{3\left\langle\cos ^{2} \Psi>-1\right.}{2}
$$

Eq SI 2
where $f_{H}=0$ for isotropic crystal orientation while $f_{H}=-0.5(1)$ for the perpendicular (parallel) orientation of the normal to the plane with the transverse direction (TD) (the reference direction correspond to $\Psi=0^{\circ}$ ). Using SAXS patterns, the Hermans function $f_{H}=-0.38$ (transverse direction TD as reference $\Psi=0^{\circ}$ ) at $q=0.03$ $\AA^{-1}$ compared to $f_{H}=-0.14$ for 0 LME film.


Figure SI 2: 2D SAXS and WAXS patterns of 0 (a) and 8 (b) LME PE/PEgMA/PA6 multilayer films in normal (ND)

Table SI 3 summarizes the interplanar distances, the long periods and the melting points of PE (PE and PEgMA) and PA6 polymers. Except from the peaks at $\sim 8 \AA$ and $4.4 \AA$ the other diffraction peaks of PE and PA6 will be superimposed.

Table SI 3: Interplanar distances, long periods and melting points of PE and PA6.


Figure SI 3 presents the evolution of WAXS patterns with X-ray along ND (a) and azimuthal integration (b) at different temperatures $T(\lambda=0.775 \AA$ ) for PA6 reference film using 0 LME. PA6 film is isotropic with a broad peak at small angle associated to the periodic organization of the crystalline lamellae and two diffraction lines in the wide-angle domain. A small peak attributed to the do20 distance at $2 \theta \sim 5.5^{\circ}(\mathrm{d} \sim 8 \AA)$ and the main peak dooi/ $\mathrm{d}_{200} / \mathrm{d}_{20-1}$, at $2 \theta \sim 11^{\circ}(\mathrm{d} \sim 4,1 \AA)$ of the monoclinic pseudo hexagonal $\gamma$ phase. During heating, this latter Bragg peak becomes sharper, and its intensity increase. This evolution suggests the presence of only $\gamma$-phase (contrary to $\beta$-phase which easily turn in $\alpha$-phase [6]). We cannot calculate a crystallinity ratio with this very narrow $2 \theta$ range but the evolution of the crystalline peak ratio during heating shows a significant increase ( $+50 \%$ ) between $25^{\circ} \mathrm{C}$ to $175^{\circ} \mathrm{C}$. WAXS deconvolution was performed with Origin software using Gaussian and Pearson VII function for amorphous (which shape was fixed for all temperatures and determined in molten state) and crystalline peaks, respectively. The broad peak at low angles associated to the crystalline lamellae order increases in intensity and shifts to low angle between $70^{\circ} \mathrm{C}$ and $200^{\circ} \mathrm{C}$. This
behavior is due to the expansion of the amorphous phase above the glass transition $\left(\operatorname{Tg} \sim 50^{\circ} \mathrm{C}\right)$ and to the increase of crystallinity. Thus, the PA6 $\gamma$ monoclinic crystalline phase is confirmed in the 8LME multilayer film, in agreement with DSC thermograms (Figure SI 3). The presence of $\gamma$-phase can be attributed to the rapid cooling down of films on chill rolls $\left(T=80^{\circ} \mathrm{C}\right)$. This fast cooling leads to the increase in crystallinity observed during heating.


Figure SI 3: 2D WAXS patterns (a) and 1D diffractograms at low (b) and high (c) scattering vector values $q$ (or $2 \theta$ ) of a reference PA6 film in normal (ND) at different temperatures $T$

Figure SI 4 presents the circular 1D integration at various angle $\Psi=0,30,60$ or $90^{\circ}$ in ND, ED and TD. These integrations (with an angle window of $20^{\circ}$ ) were performed from the 2D images using the Foxtrot software.

These profiles help us to identify the various peaks and their respective orientation.


Figure SI 4:2D SAXS (on the left, $a, c$ and e) and WAXS (in the right, $b, d$ and $f$ ) $1 D$ diffractograms integration at various azimuthal angles at room temperature of 8 LME PE/PEgMA/PA6 multilayer films in normal (ND) (a) and extrusion (ED) (b) and transverse (TD) direction.

Figure SI 5 represent one example of each films tested in uniaxial test in order to give an average value at least ten on each sample were tested.


Figure SI 5: raw data of mechanicals tests performed on PE-PEgMA-PA6 films in extrusion and transverse direction.

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