



**NMP3 - CT - 2004 - 500311**

**Sustainpack**

**Innovation and sustainable Development in the Fibre Based Packaging Value Chain**

Instrument: **IP**

**D5.17B. Report on recycling ability of biodegradable matrices in the plastic recycling stream (Months 30).**

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<b>Dissemination Level</b>		
<b>PU</b>	Public	<b>PU</b>
<b>PP</b>	Restricted to other programme participants (including the Commission Services)	
<b>RE</b>	Restricted to a group specified by the consortium (including the Commission Services)	
<b>CO</b>	Confidential, only for members of the consortium (including the Commission Services)	

## SP5 – 3D Composite packaging

### WP5.1. Fibre modification

The recycling of three different renewable biodegradable matrices was studied by their multiple extrusions and the characterisation of the ensuing recycled materials. The matrices studied were starch<sub>materbi</sub>, Polyester Materbi (PEM) and cellulose aceto-butyrate (CAB).

They were processed 1, 5 and 10 times. The extrusion temperature was kept constant at 180°C for starch and CAB and at 100°C for PEM. The duration of each recycling cycle was about 3 minute of mixing and two minutes of crossing the extruder. The recycled materials were the thermoformed to produce specimens suitable for DMA analysis, i.e., samples having about 20, 4-5 and 0,5-0,8 mm of length, width and thickness, respectively.

The resulting recycled matrices were characterised by DSC (Differential Scanning Calorimetry) and DMA (Dynamical Mechanic Analysis). These two apparatus are TA instrument facilities. The rate of temperature increase was kept constant at 10°C/min. The starting temperature was about -100°C, thanks to the use of liquid nitrogen.

### Recycling of PEM

The DSC tracing of these PEM, after 1, 5 and 10 extrusions are shown in Figures 1 to 3. Table 1 summarises the thermal data deduced from these tracings.

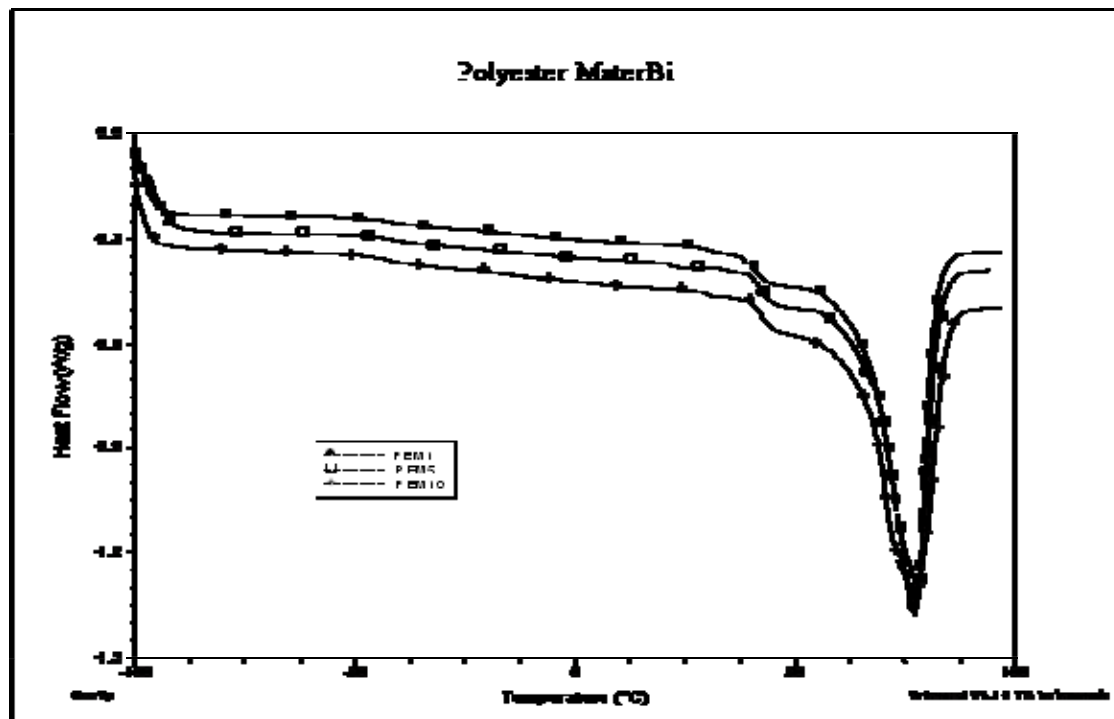


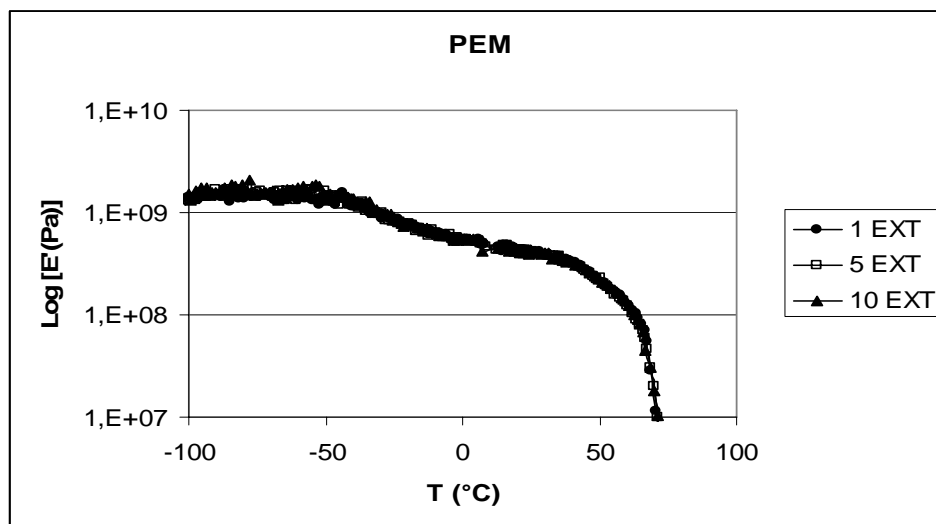
Figure 1. DSC tracing of PEM after 1, 5 and 10 extrusions.

		T <sub>g</sub> (°C)	T <sub>f</sub> (°C)	ΔH <sub>f</sub> (J/g)
PEM	1 extrusion	-40	77	85,7
	5 extrusions	-41	77	81,4
	10 extrusions	-41	76	83,6

**Table 1.** Thermal properties of PEM

The thermal properties of PEM show that it is a semi-crystalline polymer with a glass transition temperature of about -40°C and a melting point of 77°C. These data remained constant when the matrix was submitted to multiple extrusions. The enthalpy of fusion (melting) slowed down slightly, because the polymer did not recover totally its initial crystallinity. It is worth to notice that the PEM recovered very high amount of its initial crystalline morphology, i.e., practically 90%.

These results show clearly that PEM did not suffer from the thermo-mechanical cycles to which it was subjected.



**Figure 2.** DMA tracing of PEM after 1, 5 and 10 extrusions.

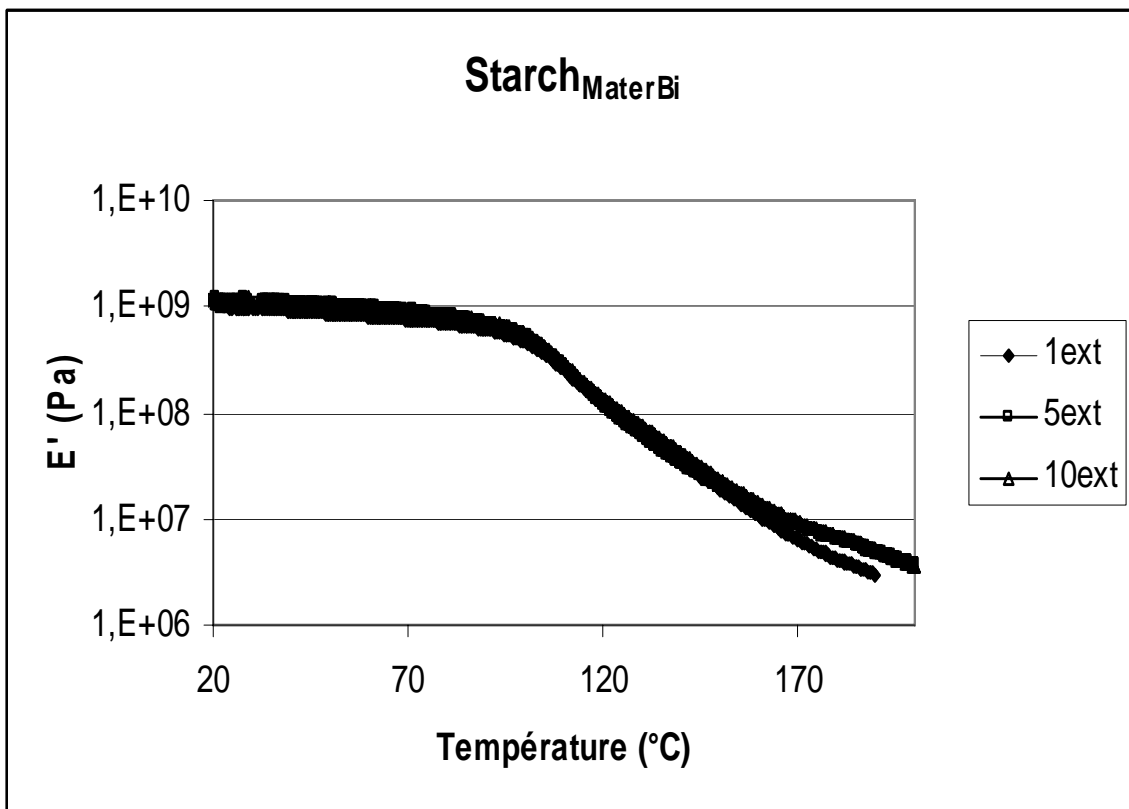
The DMA analysis confirmed the results deduced from DSC, as shown in Figure 2, from which it can be easily seen that the Young modulus ( $E'$ ) of PEM is about  $10^9$  at 25°C. It is also easy to observe that the extrusion of PEM did not alter its mechanical performances, at least when this process is repeated 10 times. Finally, it can be deduced that the melting temperature of PEM is located around 75°C. In fact the Young modulus of PEM slowed down drastically at this temperature. Curiously,  $E'$  remained practically unchanged at the glass transition temperature of PEM (around 40°C, as determined by DSC).

These data predict the suitability of PEM recycling in the plastic stream.

**Recycling of starch**<sub>materbi</sub>

The DSC tracing of starch<sub>materbi</sub> is not exploitable, since this material does not show a clear thermal transition, as in most cases related to polysaccharide polymers. However, the DMA measurements showed that the multiple recycling of this matrix did not affect its mechanical properties, as presented in Figure 3. In fact, the three curves related to DMA tracing of starch<sub>materbi</sub>, after 1, 5 and 10 extrusion cycles, are overlapped very closely. The glass transition temperature of starch<sub>materbi</sub> can be deduced from DMA tracing, which show that this thermal transition is situated around 100°C.

The Young modulus of starch<sub>materbi</sub> is very close to that of PEM, i.e., about  $10^9$  at 25°C. It can also be concluded that the recycling of starch<sub>materbi</sub>, in the plastic stream, can be considered as promising route.



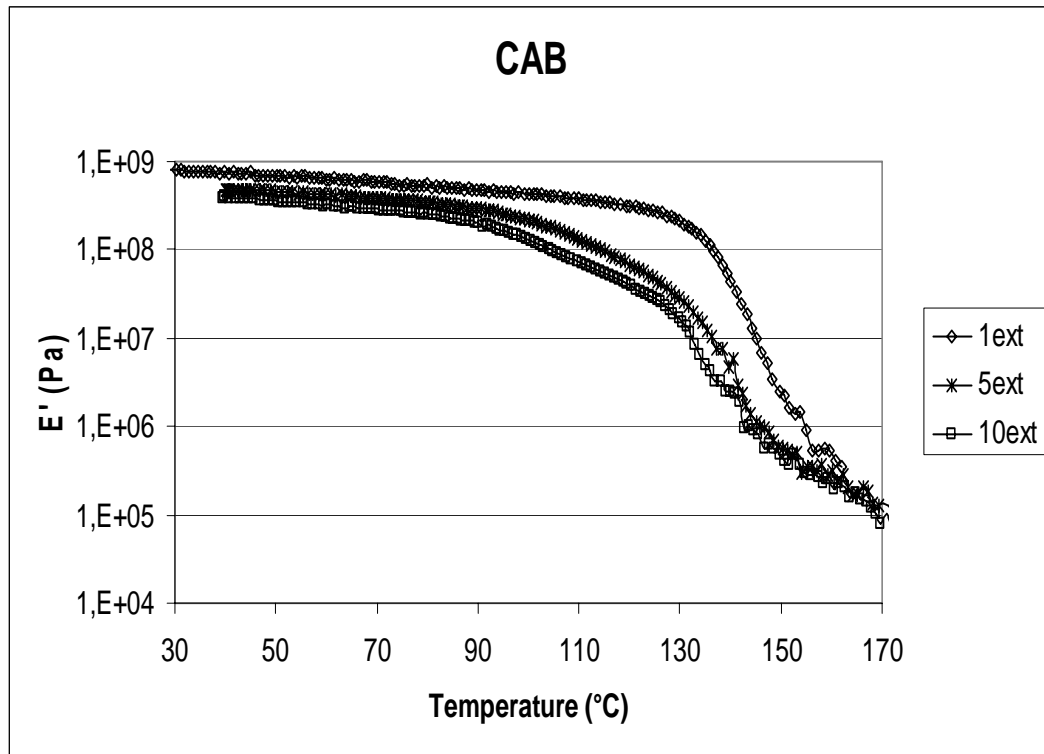
**Figure 3.** DMA tracing of starch after 1, 5 and 10 extrusions.

### Recycling of CAB

As in the case of starch<sub>materbi</sub>, the DSC tracings of CAB were not exploitable. However, the DMA measurements showed that the recycling of this matrix affected significantly its mechanical properties, as presented in Figure 4. In fact, the three curves related to DMA tracing of CAB, after 1, 5 and 10 extrusion cycles, showed two negative features, (i) a decrease in the  $E'$  and (ii) a shift of glass transition temperature to lower values. In fact, CAB displayed an  $E'$  of about  $10^9$  at 25°C and after 15 times of recycling cycles, this value shifted to  $0.5 \cdot 10^9$  at 25°C. The glass

transition temperature of CAB was about 130°C for initial polymer and slowed to about 110 and 90°C for recycled CAB using 5 and 10 extrusion cycles respectively.

From these data, it can also be concluded that the recycling of CAB, in the plastic stream, will cause a lost of its mechanical performances.



**Figure 4.** DMA tracing of CAB after 1, 5 and 10 extrusions.

## CONCLUSION

This work showed very clearly that the recycling of PEM and starch<sub>materbi</sub> can be processed for 15 times without affecting negatively their thermal and mechanical. Thus, these properties remained practically unchanged. Form CAB matrix, the repeated processing yielded a significant lost of its thermal mechanical performances.