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Supplemental information

**Rapid depolymerization of poly(ethylene terephthalate)
thin films by a dual-enzyme system
and its impact on material properties**

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SUPPLEMENTAL INFORMATION

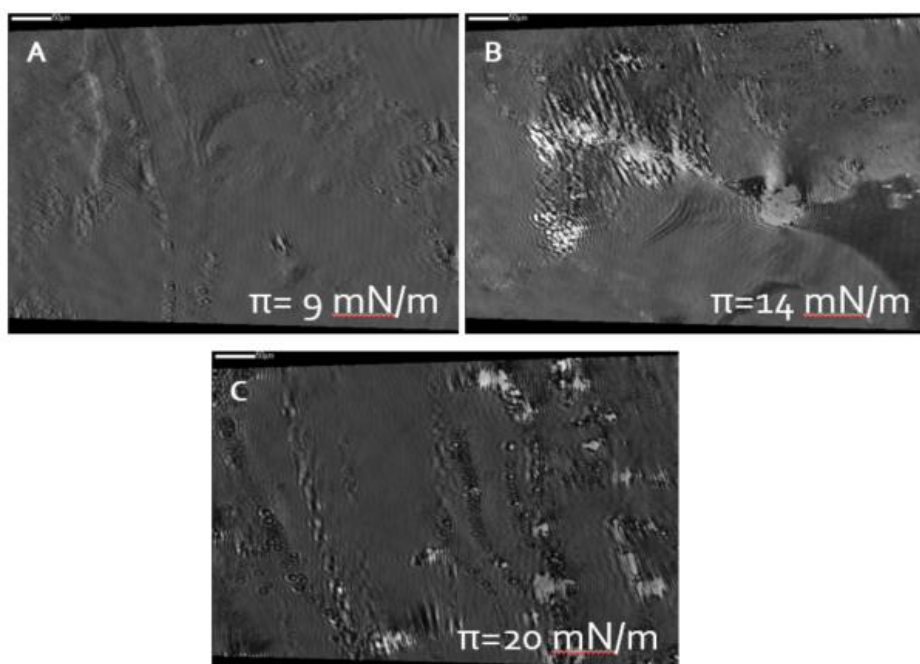


Figure S1. *In situ* Brewster Angle Microscopy images of PET 2D thin films. Images were taken during the Surface pressure - Area (π -A) compression isotherm (Figure 1) at surface pressure >9 mN/m. Scale bar 50 μ m.

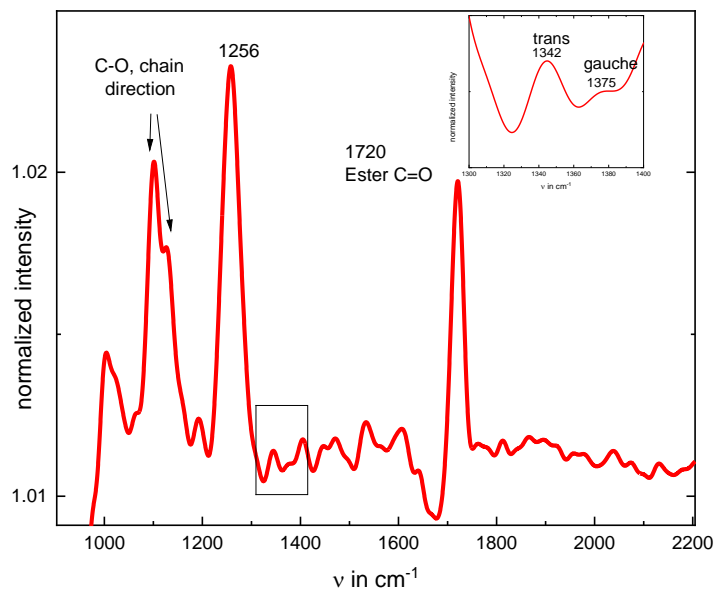


Figure S2. Polarization Modulated Infrared Reflection-Absorbance Spectroscopy (PM-IRRAS) of PET layers at the air-water interface at 21°C. Inset showing area between 1300 and 1400 cm^{-1} . The differential reflectivity spectrum S is calculated from the collected difference (ΔR) and sum spectra (ΣR) of the detected intensities of the p- and s- polarized light as $S = \frac{\Delta R}{\Sigma R} = \frac{R_S - R_p}{R_S + R_p}$. To obtain the spectrum of the film adsorbing to the A-W interface, the sample spectrum is normalized with respect to the spectrum of the bare A-W interface S_0 with $S_{film} = \frac{S - S_0}{S_0}$.

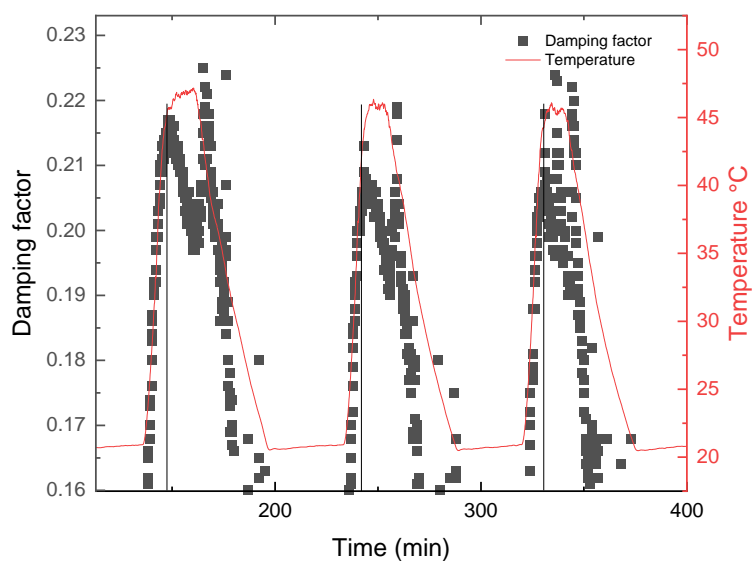


Figure S3. Damping factor vs. Temperature cycles for PET layers. PET films were prepared at surface pressures of 6, 4, and 2 mN/m and heated from 21 °C to 45 °C.

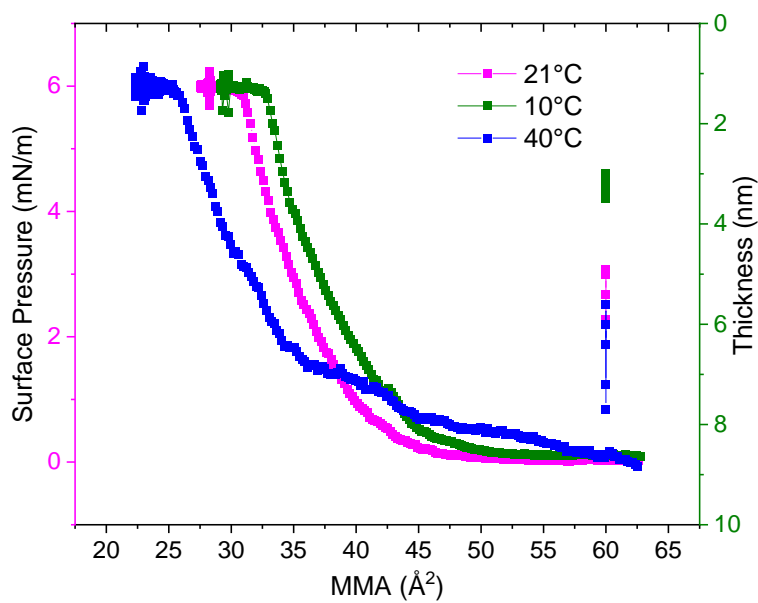


Figure S4. π -A isotherms of PET films. The films were prepared and compressed at 21, 10, and 40 °C. The thickness values measured by ellipsometry (Data of 4 replicates) as described in the main document (material and methods).

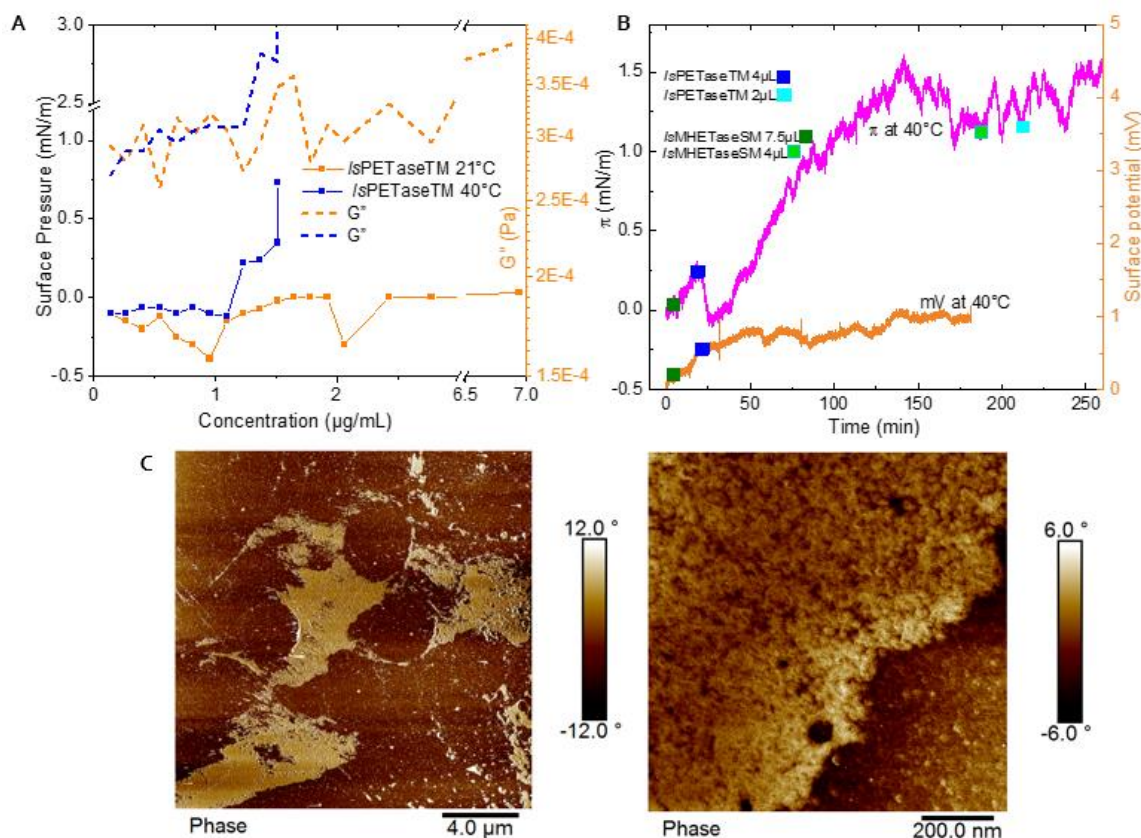


Figure S5. Surface activity of *IsMHETaseSM* and *IsPETaseTM* enzymes. A) *IsPETaseTM* surface pressure at different concentrations at 21 and 40°C in PBS pH 7.4. B) Surface pressure (π) and surface potential (mV) of *IsMHETaseSM* and *IsPETaseTM* while adsorbing at the air-water Interface at concentrations of 0.1 $\mu\text{g/mL}$ and 0.25 $\mu\text{g/mL}$, respectively. C) AFM images of water surface transferred to Si-wafers after adsorption of the enzymes at time 250 min.

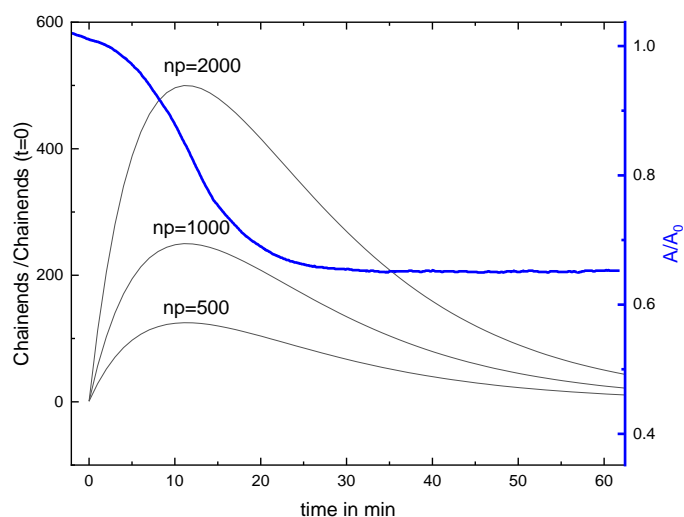


Figure S6. Fitting of the degradation curve in Figure 6B. The curve (second step) was fitted assuming there is a maximum degradability of 36%. Then the rate constant was used to calculate the normalized number of chain ends using an analytical model and Monte Carlo simulations of polymer degradation (F. Hoffmann et al., Accepted). The curve was fitted for different values of average number of monomers per chain (np). Assuming that only monomers are soluble, the average chain length will be close to 1 in the end. When $np = 1000$, from one chain with 2 ends, 1000 fragments with 2000 ends are obtained, meaning a ratio of 1000 in total. However, since fragments are not all formed at the same time, and are also being dissolved, the number of chain ends in the layer maxes out at 250.

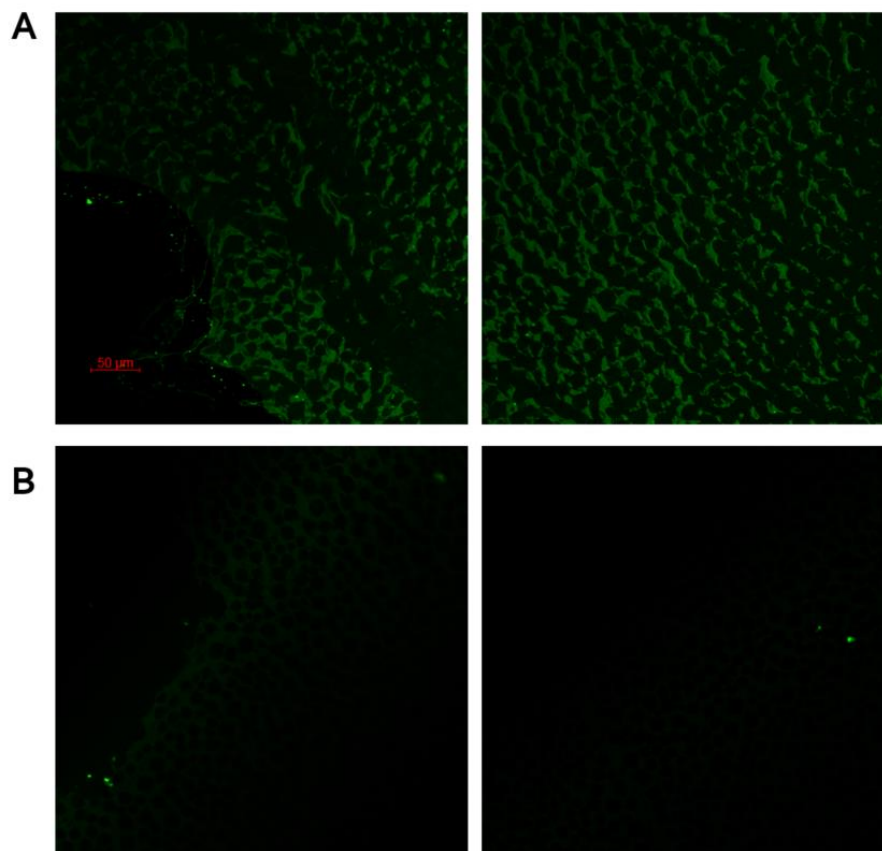


Figure S7. Confocal microscopy images of NHS-labeled films transferred to Si-wafers. A) after degradation by *IsMHETaseSM* and *IsPETaseTM* (From Figure 6) and B) after incubation at 40°C without enzymes (From Figure 4). Left and right pictures show different locations in the sample.

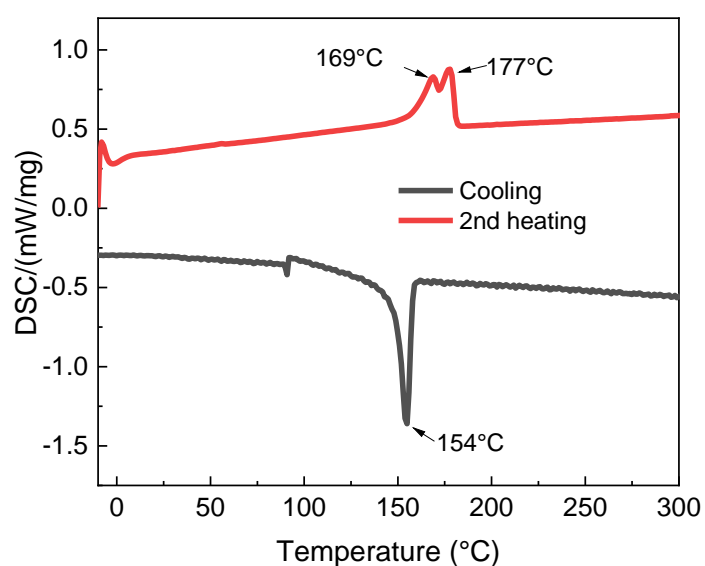


Figure S8. Differential Scanning Calorimetry of bulk PET. The measurement was performed in a Netzsch DSC 204 Phoenix (Selb, Germany) at heating and cooling rates of 10 K·min⁻¹.

Table S1. Contact angle of PET before and after degradation by *IsPETase*™.

Sample	Contact angle °	
	Advancing	Receding
Si-wafer	61.3 (±2.0)	46.3 (±1.2)
Si-wafer + PET 6 mN/m	76.1 (±2.3)	54.5 (±2.5)
Si-wafer + PET after deg.	64.9 (±1.9)	32.7 (±3.4)

Table S2. Reaction rate constants (k^*c_{enz}), and deactivation rate constants (k_{deac}). The constants were calculated in dependence on temperature, from the degradation curves using the kinetic model from Machatschek *et. al.* 2020.^[S1]

Temp	k^*c_{enz}	k_{deac}
°C	1/s	
50	0,022	0,081
40	0,036	0,051
30	0,003	0,0076
<i>with IsMHETase</i>		
40	0,019	0,016
50	0,098	0,099

References

[S1] Machatschek, R., Lendlein, A. (2020). Fundamental insights in PLGA degradation from thin film studies. *J Control Release*. 319, 276-284. 10.1016/j.jconrel.2019.12.044