InComEss

Innovative polymer-based composite systems
for high-efficient energy scavenging and storage

Training Material on the scale-up of thermoplastic-based thermoelectric composites

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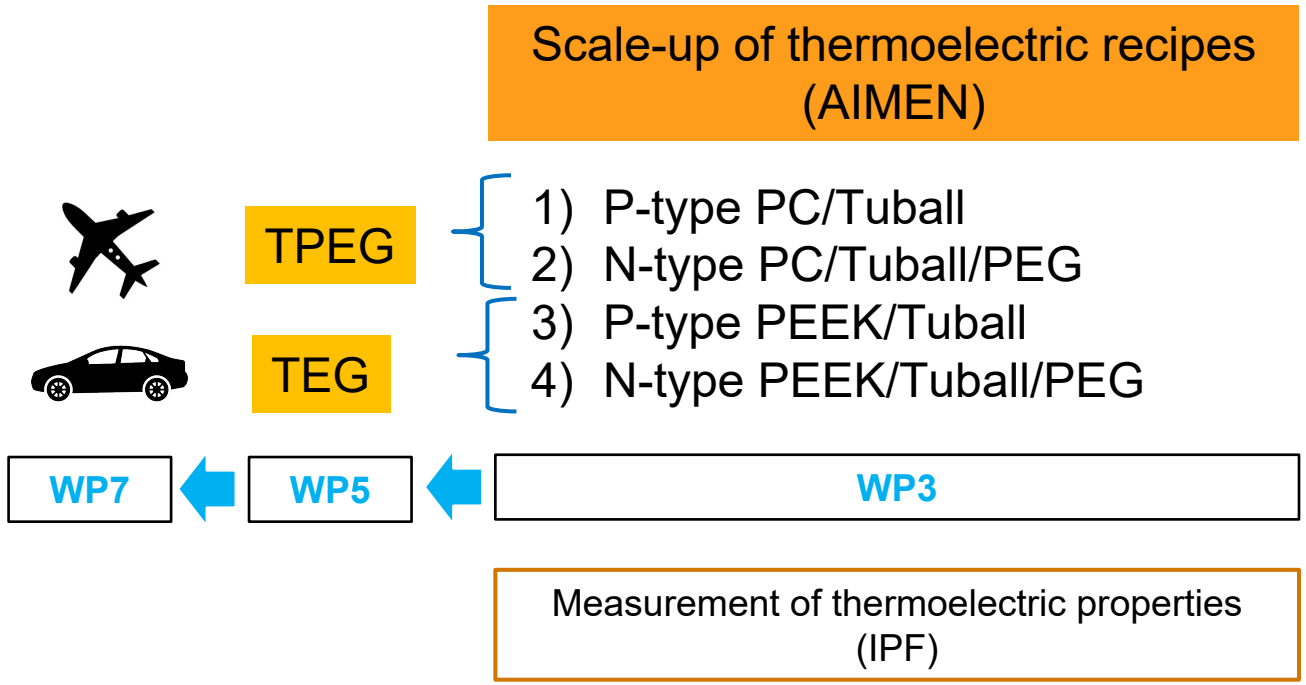
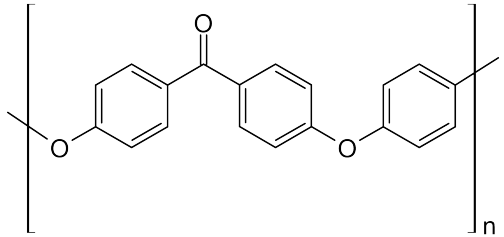
ipf Leibniz-Institut für Polymerforschung Dresden

Scale-up of materials for fabrication of energy harvesting components

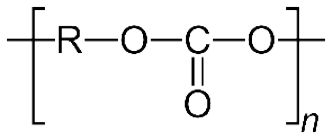
- **Production of p/n-type TE thermoplastic composites masterbatches** for the **construction of TEG** (thermoelectric generator) **and TPEG** small scale **prototypes** (WP5) **and** the **demo prototypes** (WP7) within InComEss project.
- **AIMEN's role: to optimize the recipes developed by IPF** for continuous production of CNT-thermoplastic filaments at large-scale.



Scale-up of materials for fabrication of energy harvesting components



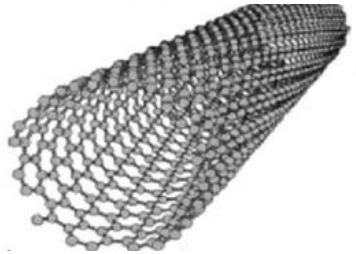
PEEK



PC

Polymer matrices

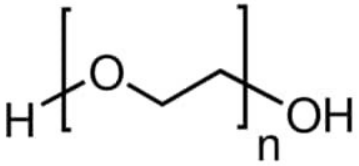
- PEEK (Polyether ether ketone)
- PC (Polycarbonate)



SWCNTs

Conductive filler

- SWCNTs (singlewalled carbon nanotubes)



PEG

Switching additive

- PEG (Polyethylene glycol)

$$S = \frac{U}{dT}$$

S = Seebeck coefficient
U = Thermovoltage
dT: Temperature difference

$$PF = S^2 \cdot \sigma$$

PF = Power factor
σ = Volume conductivity

p-type composites (S>0) : polymer matrix (PEEK or PC) + conductive filler (SWCNTs)

n-type composites (S<0) : polymer matrix/SWCNTs + Switching additive (PEG)

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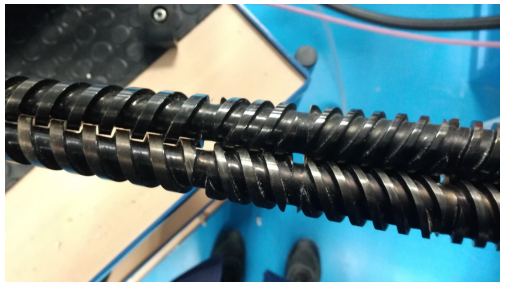
HAAKE PolyLab QC: Rheomex QC (Extruder – AIMEN's facilities)

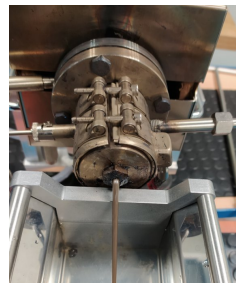
APPLICATIONS:

- Extrusion of thermally critical materials such as PVC
- Compounding of fillers and reinforcing materials
- Blending of polymers
- Pilot plant production of master batches
- Continuous mixing of recycling materials
- Mixing of nano clays and carbons
- Discharge for further process state

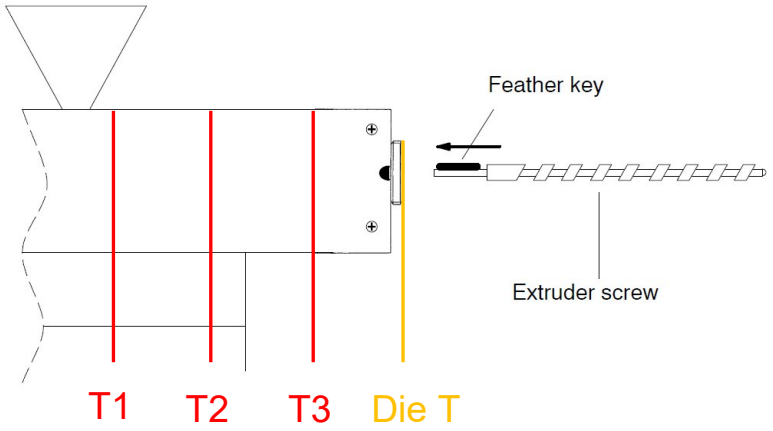
Conical twin-screw extruder

- Continuous compounding & plasticizing
- Counter rotating conical twin-screw compounder, with intermeshing screws => well-defined residence time for faultless production of process-critical polymers.





HAAKE PolyLab QC – Rheomex QC (Extruder)



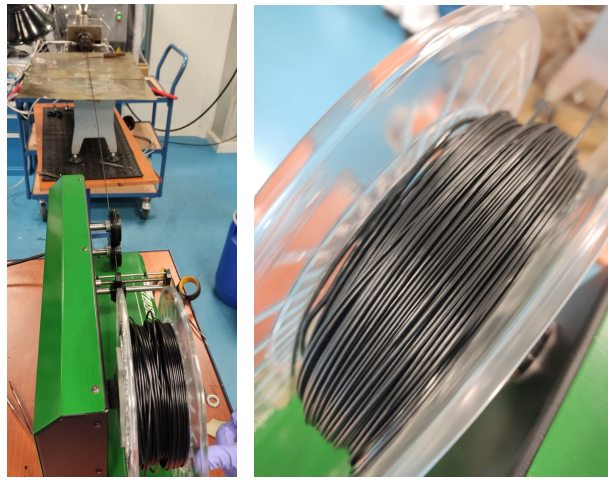
- Extruders: melting, conveying and compounding polymers under temperature, pressure and shear.
- Extruded polymer pressed through a die to form a solid material such as fibre.
- The extrusion process characterized by monitoring processing power (torque), temperature, pressure and throughput

Specifications	***Value***
Dimensions	(L x W x H) 290 mm x 750 mm x 210 mm
Weight	42.9 kg (only base extruder)
Screw diameter	conical
L/D	
Gear ratio	1:1
Rotating direction	counter
Max screw speed	250 min^{-1}
Max temperature	450 °C
Max pressure	700 bar
Max torque	200Nm
Heating zones	3
Cooling	air
External heaters	2
Feeding zone	cooled

Scale-up process

- PC/SWCNTs (2.5 wt%) composites (***p-type***)
  - PC dried at 120 °C during 4h
  - 0.5 h of mixing time PC/SWCNTs (2.5 wt%) in turbula
  - PC/SWCNTs added to feeder of extruder machine
  - Control of the four temperature ranges, speed and torque

MATRIX	FILLER (wt%)	TS1 (°C)	TS2 (°C)	TS3 (°C)	TD (°C)	SPEED (rpm)	TORQUE (Nm)
PC	SWCNTs (2.5 wt%)	220	230	240	210	40	45



Scale-up of p-type PC/SWCNTs (2.5 wt%) composite

Composite	Vol. conductivity [S/m]	Seebeck coefficient [μV/K]	Power factor [μW/(m·K²)]
PC/SWCNTs (2.5 wt%)	23.1	53.2 ± 0.3	$6.5 \cdot 10^{-2}$

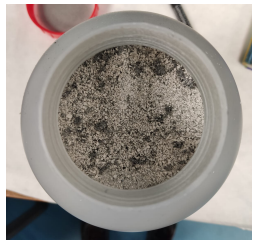
Selection for TPEG

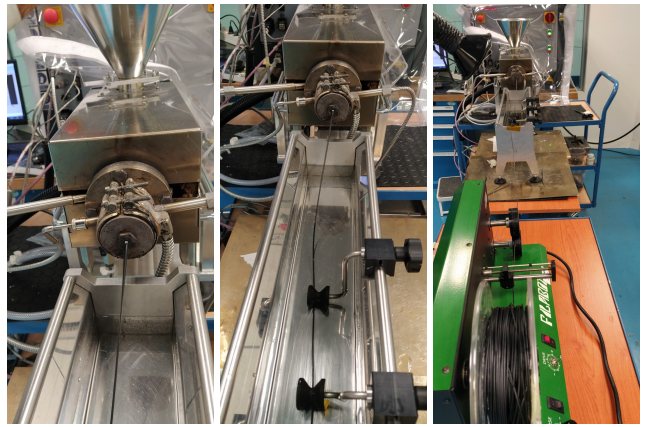
Scale-up process

➢ PEEK/SWCNTs (2.5 wt%) composites (***p-type***)

Conditions

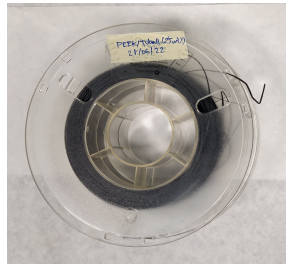
- PEEK dried at 120ºC for 4h
- Premix: PEEK + SWCNTs during 1h





Nº	MATRIX	FILLER (wt%)	TS1 (°C)	TS2 (°C)	TS3 (°C)	TD (°C)	SPEED (rpm)	TORQUE (Nm)
3	PEEK	SWCNTs (2.5 wt%)	335	360	360	340	40-55	40

✓ Continuous extruded filament for p-type PEEK/SWCNTs composites (bobbins).



Composite	Vol. conductivity [S/m]	Seebeck coefficient [μV/K]	Power factor [μW/(m·K²)]
PEEK/SWCNTs (2.5 wt%)	14.6	47.0 ± 0.3	$3.2 \cdot 10^{-2}$

Selection for TEG

Scale-up process

➢ **N-type composites** (PC and PEEK)

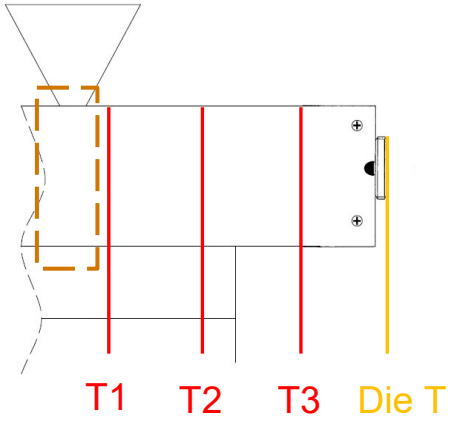
Conditions

- Dry of polymer matrix at 120ºC during 4h

- Premixing of n-type composites:
  1) SWCNTs + Additive* (15 min) & 2) SWCNTs/Additive* + Polymer matrix (30 min)

  *Additive: PEG
  *Polymer matrix: PC or PEEK

- Problems with the extrusion when adding PEG (low melting temperature = 65 °C) and high extrusion temperatures needed for PC or PEEK matrix.
- Formation of a paste in the first region of the extruder (stack) which difficult the extrusion of a continuous filament.
- Need to manipulate the feeding process by hand to be able to extrude the compounding.



Scale-up process

➢ **N-type** PC/SWCNTs (2 wt%)/PEG (15 wt%)

✓ Improvement of Seebeck coefficient and homogeneity of samples by following second extrusion

1) Filament/compounding obtained after the first extrusion was pelletized
2) Pellets fed into the hoper
3) 2nd extrusion followed

Nº	MATRIX	FILLER (wt%)	ADDITIVE (wt%)	TS1 (°C)	TS2 (°C)	TS3 (°C)	TD (°C)
1	PC	SWCNTs (2 wt%)	PEG (15 wt%)	190	220	230	220

PC/SWCNT (2 wt%) /PEG(15 wt%)

- Observation of an increased torque values from the fist (Nm) to the second extrusion (Nm)
- Not possible to extrude a continuous filament of n-type PC composite.
- Small extruded segments of PC/SWCNT(2 wt%)/PEG(15 wt%) were achieved. Britle samples.

Composite	Vol. conductivity [S/m]	Seebeck coefficient [μV/K]	Power factor [μW/(m·K²)]
PC/SWCNT(2 wt%)/PEG(15 wt%) 1st extrusion	52.0 ± 4.7	-26.0 ± 8.9	$3.5 \cdot 10^{-2}$
PC/SWCNT(2 wt%)/PEG(15 wt%) 2nd extrusion	68.0 ± 20.1	-35.1 ± 3.1	$8.4 \cdot 10^{-2}$

Scale-up process

➢ **N-type** PEEK/SWCNTs(2.5 wt%)/PEG(15 wt%)

✓ Improvement of Seebeck coefficient and homogeneity of samples by following second extrusion

1) Filament/compounding obtained after the first extrusion was pelletized
2) Pellets fed into the hoper
3) 2nd extrusion followed

MATRIX	FILLER (wt%)	ADDITIVE (wt%)	TS1 (°C)	TS2 (°C)	TS3 (°C)	TD (°C)	Speed (rpm)
PEEK	SWCNTs (2.5 wt%)	PEG (15 wt%)	340	370	360	335	20-40

PEEK/SWCNTs (2.5 wt%)/PEG (15 wt%) – 2nd extrusion step feeding

- Observation of an increased torque values from the to the second extrusion (from 5 to 15 Nm)
- Not possible to extrude a continuous filament of n-type PC composite.
- Small extruded segments of PEEK/Tuball(2.5 wt%)/PEG(15 wt%) were achieved. Britle samples.

1st & 2nd extrusion of PEEK/SWCNTs (2.5 wt%)/PEG (15 wt%)

Composite	Vol. conductivity [S/m]	Seebeck coefficient [μV/K]	Power factor [μW/(m·K²)]
PEEK/Tuball(2.5 wt%)/PEG(15 wt%) 1st extrusion	50.6 ± 10.4	-33.6 ± 0.3	$5.7 \cdot 10^{-2}$
PEEK/Tuball(2.5 wt%)/PEG(15 wt%) 2nd extrusion	32.8 ± 0.6	-37.1 ± 3.1	$4.5 \cdot 10^{-2}$

Conclusions

- Scale-up of continuous filament/bobbins of p-type PC & PEEK composites was successfully achieved
- Best thermoelectric characteristics for p-type PC composites correspond to a filler content of 2 wt% of SWCNTs
- Best thermoelectric properties for p-type PEEK composites with a SWCNT content of up to 2.5 wt%
- Problems with the extrusion of polymer matrices (PC/PEEK) with PEG (as switching additive)
- A second extrusion of n-type PC and PEEK composites improved the homogeneity of extruded segments and thermoelectric features, but brittleness of samples observed.
- The scale-up of n-type composites containing PEG needs to be improved.

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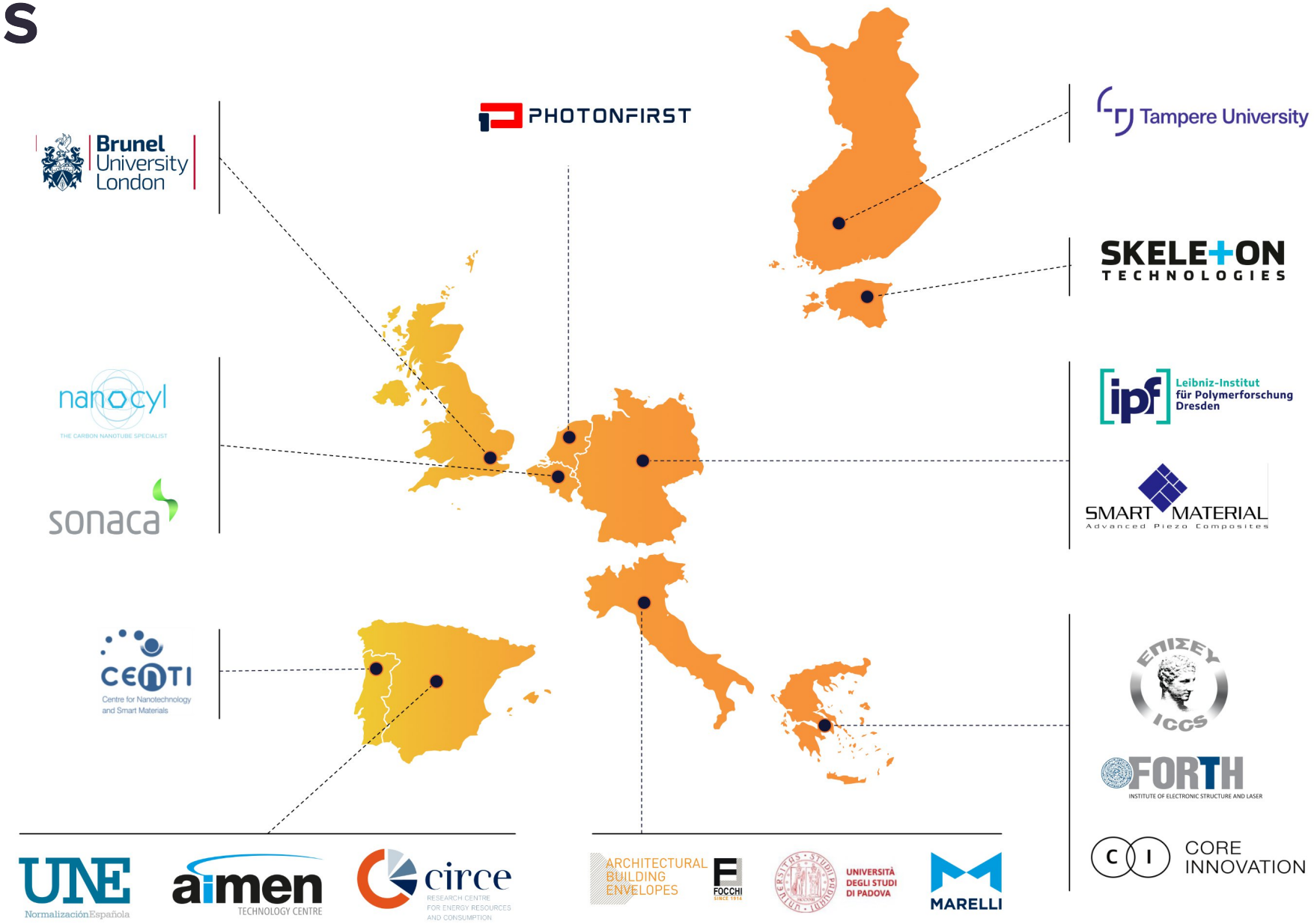
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Innovative polymer-based composite systems
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Thank you

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