Fraunhofer Cluster of Excellence Programmable Materials CPM – PT2 Programmable Heat Transfer

Sustainable Shape Memory Polymer Foams

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Thermoreversible Shape Memory Polymer (SMP) Foams

The polyurethane (PU) foams presented can be efficiently produced by reactive foaming processes and their thermal and mechanical properties can be adjusted over a wide range, depending on the building blocks used to produce them. Their ability to undergo temperature-dependent actuation is based on their phase-segregated block copolymer structure and the crystallizability of their soft segments. With a reversible shape change, also referred to as two-way shape memory effect (2W-SME), of up to more than 20% in strain, they represent promising programmable materials for use as adaptive actuators in a wide range of applications (Figure 1). This functionality, combined with good insulating properties, paves the way for novel concepts for material-inherent thermal management.

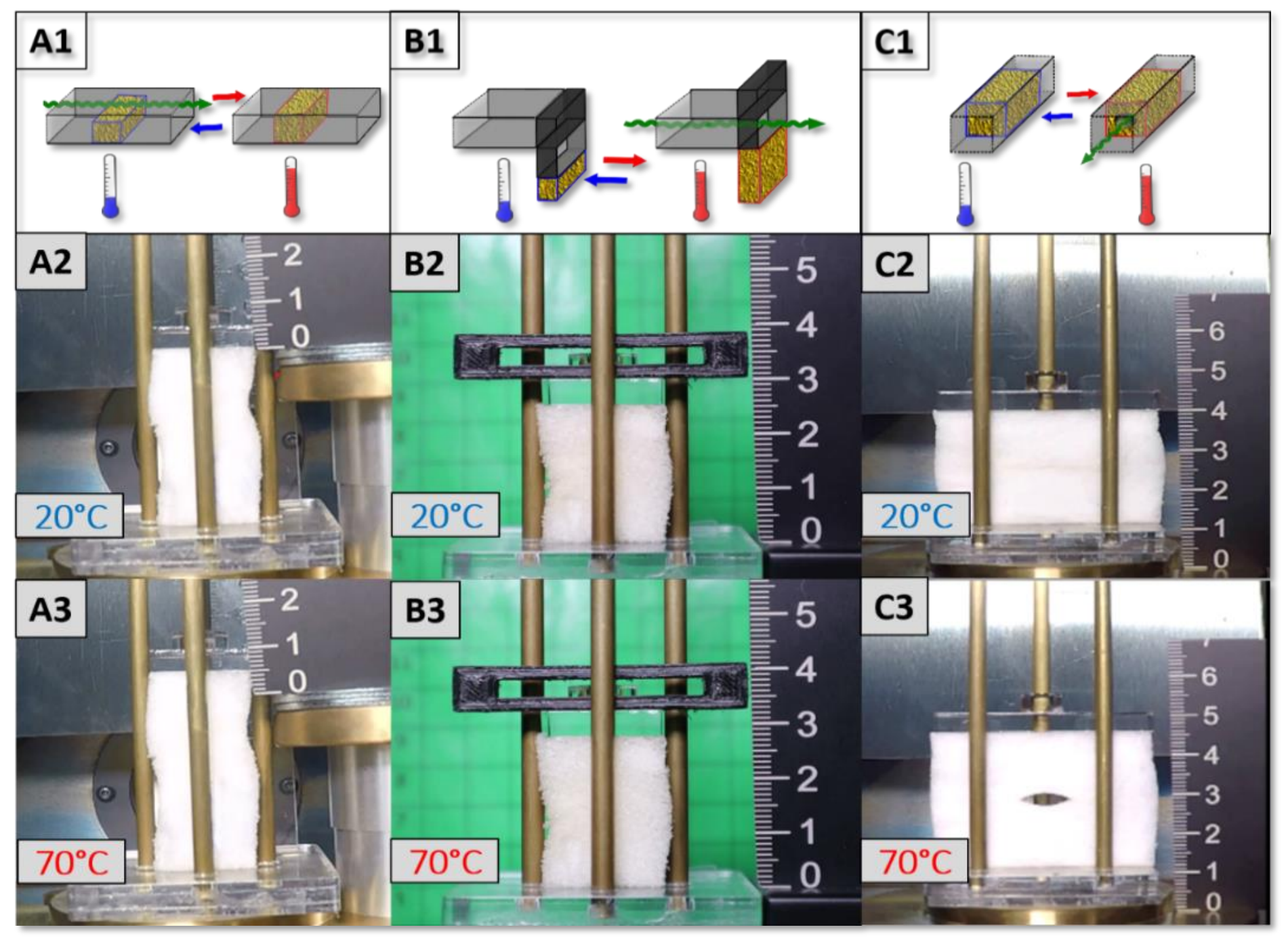


Figure 1 – overview of possible concepts for thermoresponsive foam actuators

Case Study »Adaptive Building Envelope«

In a first case study on an adaptive building envelope, in which such SMP foam elements could be used to achieve a switchable rear ventilation of the façade, the great potential of this innovative concept could be shown.

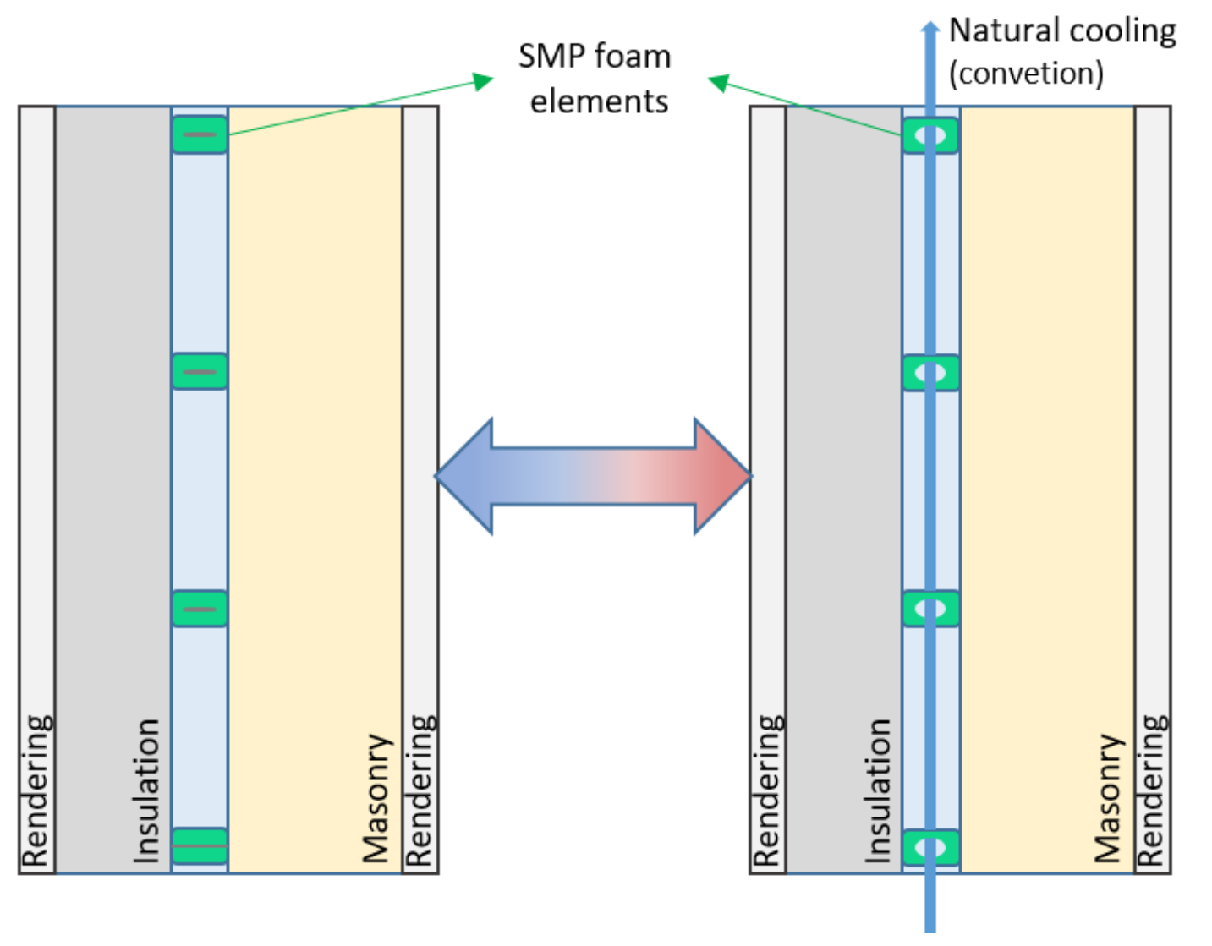


Figure 2 – Concept for an adaptive building envelope

Based on a thermoresponsive SMP foam, such a design allows to regulate the flow of ambient air through the building envelope in order to enable natural cooling of the structure (Figure 2).

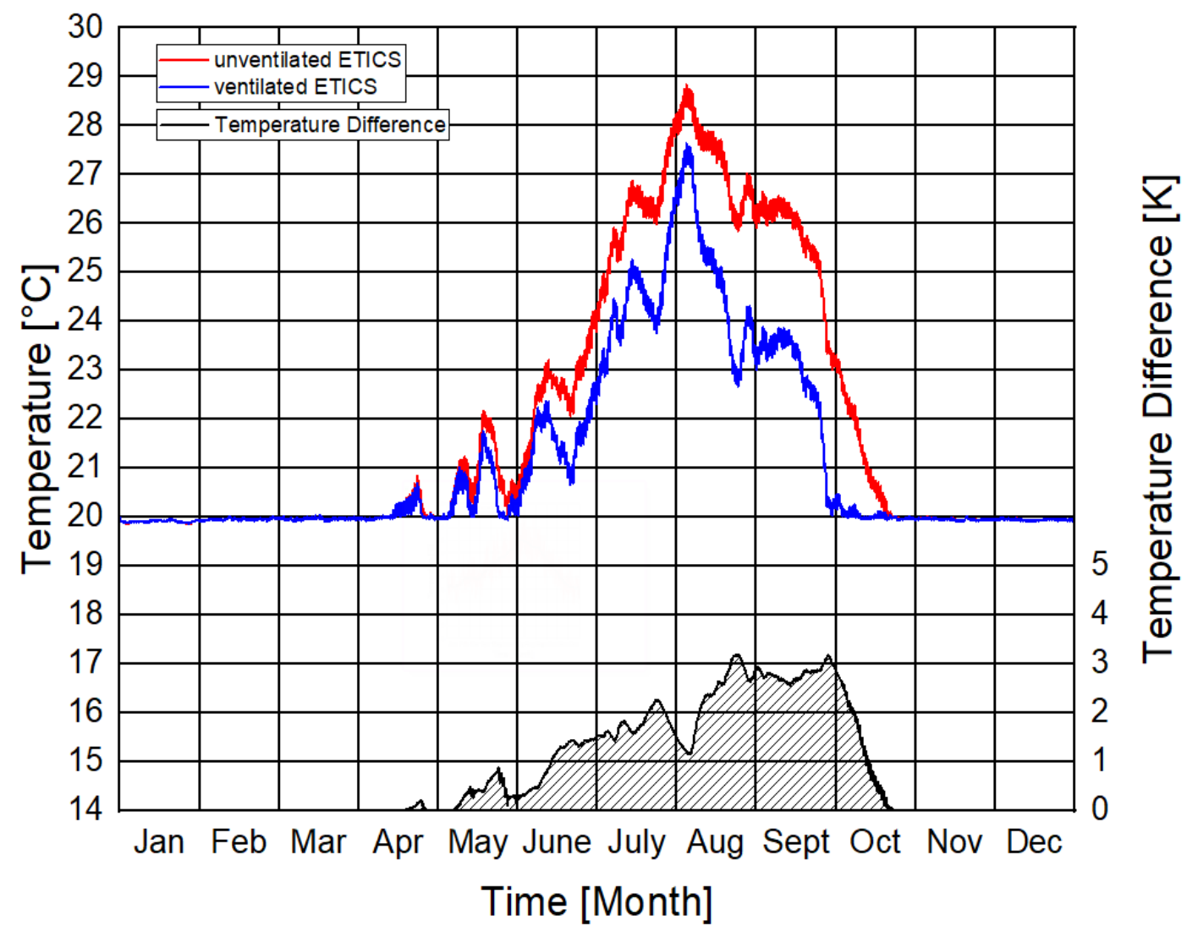


Figure 3 – Comparison of the room temperature for an unventilated (red) and a ventilated facade (blue) as well as plot of the temperature difference (black) from a hygrothermal simulation for Madrid (ES).

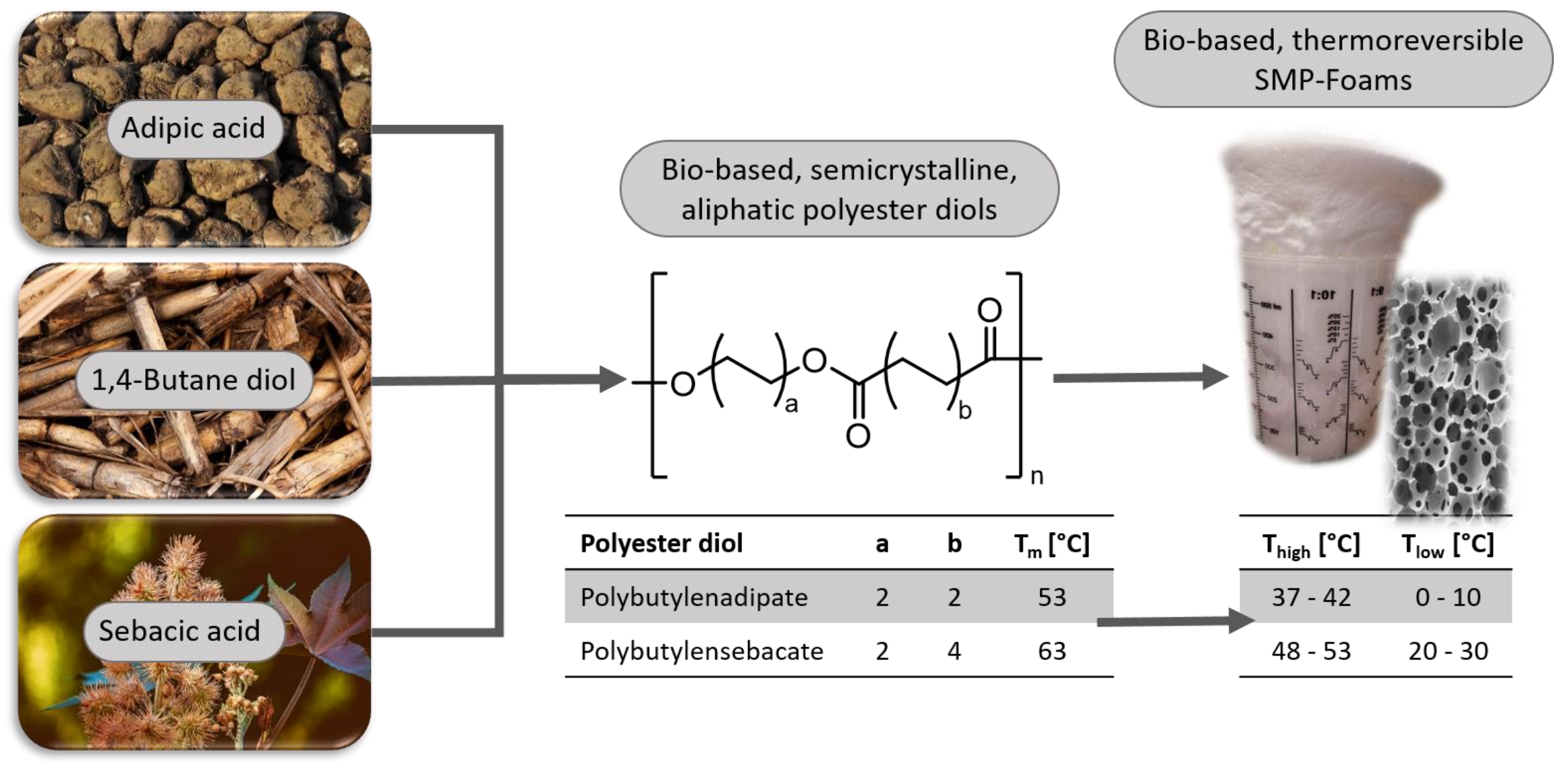
Based on hygrothermal simulation data obtained by the software WUFI® Plus, a significant cooling energy saving potential was revealed (Figure 3).

From an environmental perspective, the great potential of adaptive foam elements in building applications to reduce the impact on climate change could be indicated by means of a Life Cycle Assessment (LCA).

Improving Sustainability

Bio-based Materials

Innovative concepts for saving energy are an important aspect on the way to a more sustainable economy and society. But looking at the application alone is not enough to meet the global challenges. The materials employed must also be subjected to careful assessment in terms of their ecological impact. And here there is still room for improvement in the case of polyurethane (PU) foams, as was shown in a LCA carried out in parallel.



Polyester diol	a	b	T_m [°C]
Polybutylenadipate	2	2	53
Polybutylensebacate	2	4	63

T_{high} [°C]	T_{low} [°C]
37 - 42	0 - 10
48 - 53	20 - 30

Figure 4 – From renewable raw materials to bio-based foams (by the example of 1,4-Butanediol, Adipic acid and Sebacic acid)

A key starting point is to replace petrochemical feedstock by bio-based building blocks. The aliphatic polyester polyols used as soft segments are predestined for this (Figure 4). Meanwhile, several aliphatic diols and dicarboxylic acids, such as Ethylene glycol, 1,3-Propanediol, 1,4-Butanediol, Succinic acid, Adipic acid, Sebacic acid and 1,12-Dodecanoic diacid, are commercially available as bio-chemicals suitable for the preparation of semicrystalline soft segment building blocks.[1]

Table 1 provides a first overview of which bio-based fraction can be achieved in the SMP-PU foams studied so far. Other studies suggest that in this way the carbon foot print can be reduced by more than 20%.[2]

Recyclability

Circularity is the second major issue when it comes to developing new materials. The recycling of reactively foamed PUs is currently limited to downcycling due to the crosslinked structure of the polymer matrix. This can involve chemolysis to obtain new polyol building blocks or mechanical milling and subsequent use of the ground material in composite foams and sheets.

Figure 5 – Recycling/Reuse of linear SMP-PU foams → foam in original (left, left) and compressed state (left,right), ground foam (center) and recycled film (right).

We have succeeded in producing SMP-PU foams with a linear polymer structure using a reactive foaming process, thus paving the way for direct reuse of the material. The synthesized materials show similar pronounced reversible shape memory effect as their crosslinked counterparts (Table 1). Thermoplastic processability was demonstrated by compressing foam granules into TPU films (Figure 5). Currently, we are working on the realization of processing in an extruder. So far, this has led to thermal degradation of the polymer, presumably due to the oxygen introduced into the process from the foam cells.

***Table 1** – Comparison of properties of crosslinked and linear SMP foams and overview on potential renewable carbon content*

	Formulation		Physical properties					1W-SME		2W
	Polyol	NCO	RCC [%]	$T_{m,Peak}$ [°C]	$T_{c,Peak}$ [°C]	OCR [%]	ρ [kg/m³]	$R_f(2)$ [%]	$R_r(2)$ [%]	ε_{rev} [%]
cross-linked	PHA	MDI	37	37.7	5.4	36	72	98.3	98.6	22.7
	PDA/PBA	MDI	46	56.5	37.0	88	133	97.8	99.3	13.7
linear	PBA	MDI	76	30.1	*	96	103	99.4	98.4	14.8
	PHA	MDI	38	35.6	3.7	96	95	99.2	98.4	17.6
	PHA	TDI	41	36.9	-3.9	27	88	99.5	99.5	21.7
	PDA/PCL	MDI	39	54.9	32.9	86	86	98.4	99.6	21.1

NCO – Isocyanate, RCC – renewable carbon content, $T_{m,Peak}$ – melting temperature, $T_{c,Peak}$ – crystallization temperature, OCR – open cell ratio, R_f – fixity ratio, R_r – recovery ratio, ε_{rev} – reversible actuation, PHA – Poly(1,6-hexylene) adipate, PDA – Poly(1,10-decylene) adipate, PBA – Poly(1,4-butylene) adipate, PCL – Poly-ε-caprolactone, MDI – 4,4'-Diphenylmethane diisocyanate, 2,4-/2,6-Toluene diisocyanate, * cold crystallization

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[1] De Jong, E. ; Stichnothe, H.; Bell, G.; Jørgensen, H. Bio-Based Chemicals - A 2020 Update; IEA Bioenergy: Task 42: 2020: 01. ISBN 978-1-910154-69-4
[2] Maga, D., Melchiors, M., Henneken, H. *et al.* Klimaschutz durch biobasierte Klebstoffe. *Adhaes Kleb Dicht* **61,** 16–23 (2017). https://doi.org/10.1007/s35145-017-0072-0