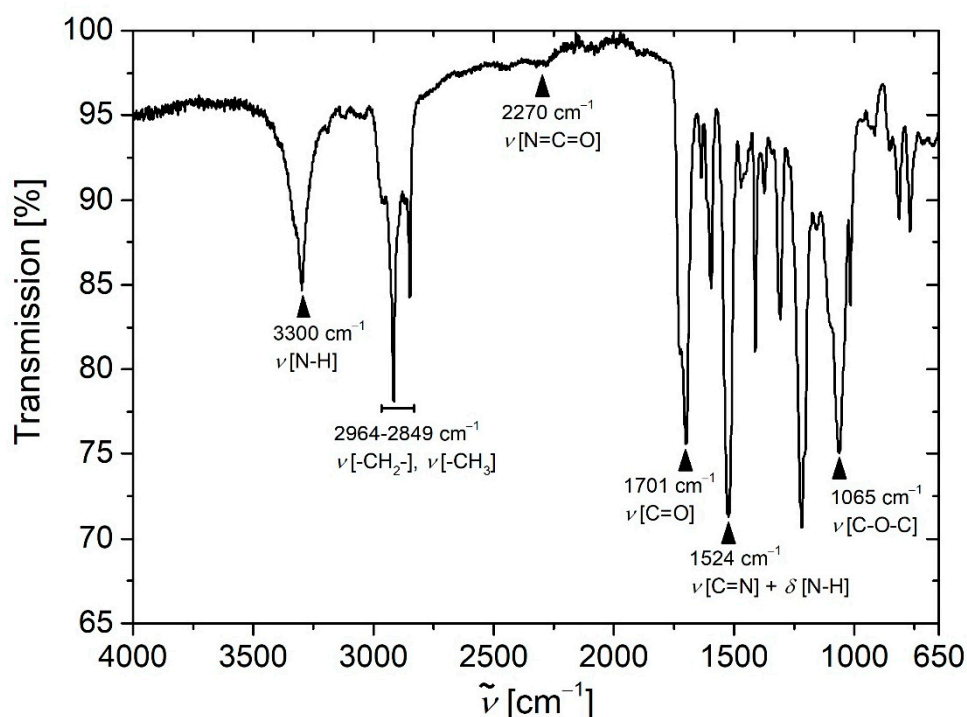


# Dual Stimuli-Responsive Orthodontic Aligners: An In Vitro Study

## Fourier-transform infrared (FTIR) spectroscopy

In the FT-IR spectrum of the synthesized TPU, a very weak signal occurred at about  $2270\text{ cm}^{-1}$ , indicating that only a few free isocyanate groups were available [1]. This speaks to an almost complete reaction. At the same time, vibration modes are present that are characteristic of the formation of a poly(ether urethane) (Figure S1).



**Figure S1.** FT-IR spectrum of polypropylene glycol (PPG)-based poly(ether urethane), including the assignment of vibration modes and the specification of their wavenumbers.

A more detailed analysis of the FT-IR spectrum shows overlapping absorbances between  $2964\text{ cm}^{-1}$  and  $2849\text{ cm}^{-1}$ , which can be assigned to asymmetric and symmetric stretching vibrations of  $\text{CH}_2$  alkyl entities together with  $\text{CH}_3$  entities of the PPG side chain [2,3]. The absorbance band at  $1065\text{ cm}^{-1}$  can be assigned to the  $\nu[\text{C-O-C}]$  stretching vibrations of the PPG soft segment [3,4]. The stretching vibrations  $\nu[\text{N-H}]$  at  $3300\text{ cm}^{-1}$ ,  $\nu[\text{C=O}]$  in the carbonyl stretching region at  $1701\text{ cm}^{-1}$ , as well as an amide peak ( $\nu[\text{C=N}] + \delta[\text{N-H}]$ ) at  $1524\text{ cm}^{-1}$ , indicate the formation of the hard segment. The signals can be attributed to the formation of urethane species [3,5,6]. As a result, the reaction seemed to be successful.

## Cyclic thermomechanical measurements

**Table S1.** Dependence of shape memory properties of the synthesized TPU on cycle number. For  $T_{\text{low}}$  and  $T_{\text{high}}$ , 23 °C and 80 °C were selected, respectively, while  $\epsilon_{\text{max}}$  was set to 100%.

Cycle number N	Fixed strain $\epsilon_u$	Recovered strain $\epsilon_r$	Shape fixity ratio $R_f(N)$	Shape recovery ratio $R_r(N)$
1	100%	37%	>99%	63%
2	100%	41%	>99%	94%
3	100%	43%	>99%	97%
4	100%	44%	>99%	98%
5	100%	45%	>99%	98%

**Table S2.** Dependence of shape memory properties of the synthesized TPU on  $\epsilon_{\text{max}}$ . For  $T_{\text{low}}$  and  $T_{\text{high}}$ , 23 °C and 80 °C were selected, respectively.

Maximum strain applied $\epsilon_{\text{max}}$	Cycle number N	Fixed strain $\epsilon_u$	Recovered strain $\epsilon_r$	Shape fixity ratio $R_f(N)$	Shape recovery ratio $R_r(N)$
25%	1	25%	20%	>99%	20%
	2	24%	20%	96%	>99%
50%	1	50%	23%	>99%	90%
	2	50%	25%	>99%	93%
100%	1	100%	34%	>99%	88%
	2	100%	37%	>99%	95%
200%	1	199%	56%	>99%	88%
	2	199%	61%	>99%	97%

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