

Electronic Supplementary Information (ESI)

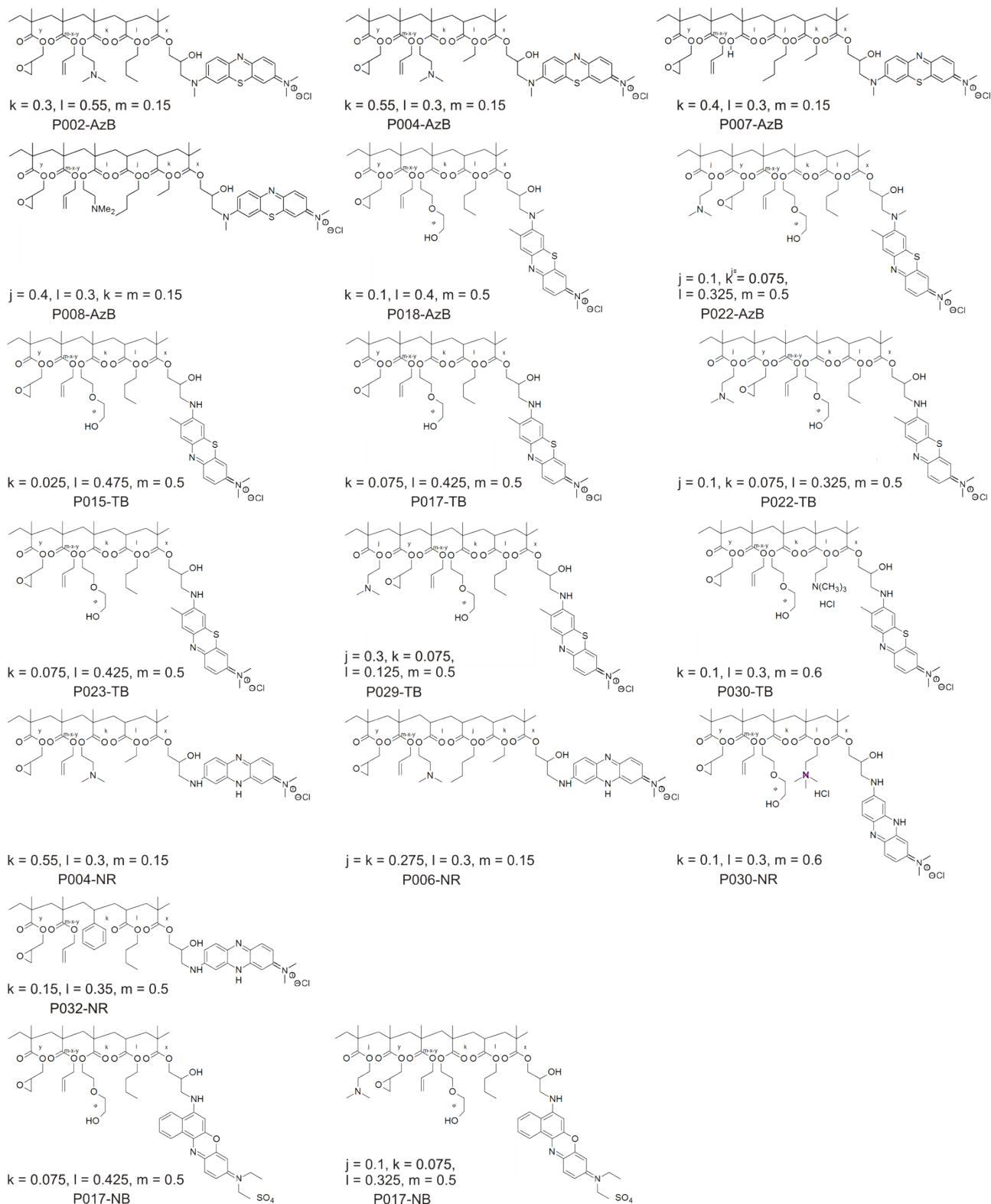


Fig. S1 Structures and indices of the phenothiazine modified redox hydrogels in the investigated library. AzB (azure blue), TB (toluidine blue), NR (neutral red), NB (nile blue).

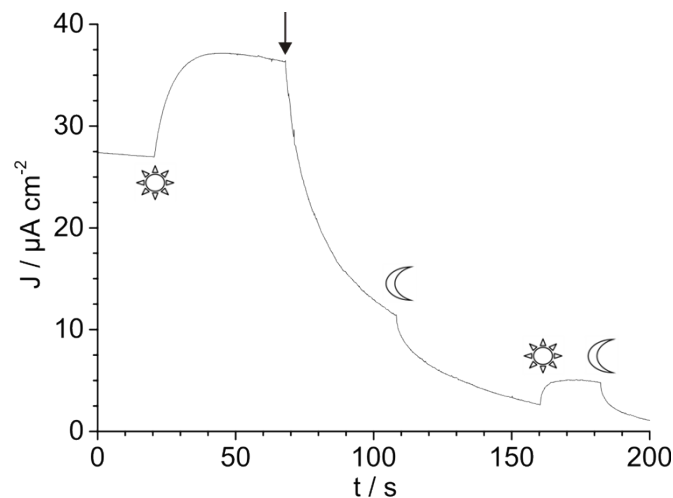


Fig. S2 Chronoamperometry of the PS2/P023-TB photoanode upon addition of the inhibitor dinoterb (100 μM , black arrow). Measurements in buffered electrolyte (50 mM MES pH 6.5, 10 mM MgCl_2 , 10 mM CaCl_2 and 0.03 % [w/v] β -DM) with an applied potential of +500 mV vs. SHE and an illumination of 34.9 mW cm^{-2} at 685 nm (sun = light on, moon = light off).

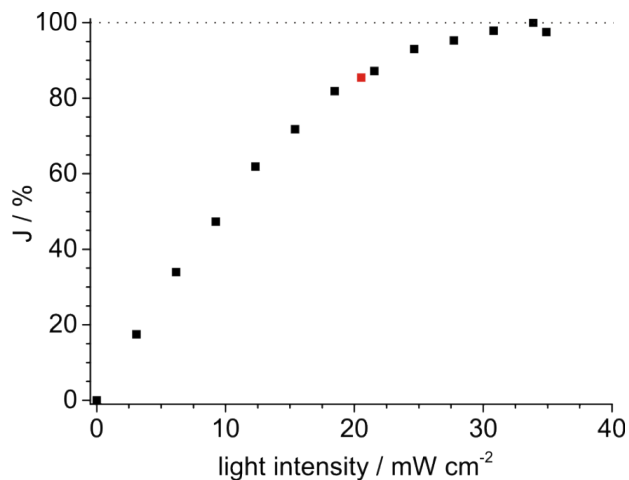


Fig.S3 Determination of light saturation conditions at the PS2/P023-TB photoanode by plotting the percentage of the maximum photocurrent density ($J / \%$) against the light intensity (mW cm^{-2}). Measurements were performed in buffered electrolyte (50 mM MES, pH 6.5, 10 mM MgCl_2 , 10 mM CaCl_2 and 0.03 % [w/v] β -DM) with stepwise increased light intensity (at 685 nm). Light intensity used routinely in biophotovoltaic measurements is marked red.

Table S1 Used compounds and providers

acrylic acid (AA)	Fluka, Buchs, D	2,2'-ethylenedioxydiethylamine	Sigma-Aldrich,Steinheim, D
ethylacrylate(EA)	Acros Organics, Geel, B	inhibitorremover	Sigma-Aldrich,Steinheim, D
allylmethacrylate(AIIMA)	Acros Organics, Geel, B	potassium hexachloroosmat	Sigma-Aldrich,Steinheim, D
azurB (AzB)	Sigma-Aldrich,Steinheim, D	potassium peroxomonosulfate triplesalt	Sigma-Aldrich,Steinheim, D
2,2'-azo-bis(isobutyronitrie) (AIBN)	Fluka, Buchs, D	methacrylicacid (MA)	Acros Organics, Geel, B
2,2'-bipyridin (bpy)	Acros Organics, Geel, B	dimethylaminoethylmethacrylate(DMAEM A)	Sigma-Aldrich,Steinheim, D
n-butanol	J. T. Baker, Deventer, NL	methanol	J. T. Baker, Deventer, NL
n-butylacrylate(BA)	Fluka, Buchs, D	neutralred(techn.) (NR)	Sigma-Aldrich,Steinheim, D
chlorethylaminehydrochloride	Acros Organics, Geel, B	nileblueA (NB)	Sigma-Aldrich,Steinheim, D
hydrogenchloride(4M in 1,4-dioxane)	Sigma-Aldrich,Steinheim, D	poly-(ethylenglycole)methacrylate (PEGMA)	Sigma-Aldrich,Steinheim, D
dichloromethane	J. T. Baker, Deventer, NL	2-propanol	J. T. Baker, Deventer, NL
di-tert-butylidicarbonate	Sigma-Aldrich,Steinheim, D	styrene (St)	Fluka, Buchs, D
diethylether	J. T. Baker, Deventer, NL	toluidineblueO (90 %) (TB)	Sigma-Aldrich,Steinheim, D
dimethylformamide	J. T. Baker, Deventer, NL	triethylamine	J. T. Baker, Deventer, NL
ethanol	J. T. Baker, Deventer, NL	[2-(methacryloyloxy)ethyl]	Sigma-Aldrich,Steinheim, D
2,2'-ethylenedioxydiethanthiole	Sigma-Aldrich,Steinheim, D	trimethylammonium chloride (80% w/w in H2O) /TMAEMA)	

5 All copolymer backbones were synthesized according to procedures mentioned in the manuscript, using the indicated monomer mixtures. The given weight concentration refers to the final copolymer suspension used for the epoxidation. Also the epoxidation as well as the modification with the respective redox dye (NB, 10 NR, AzB, TB) was performed following analogue procedures as described for the redox polymers P023-TB and P022-AzB, always using 25 mg (dry mass) of the polymer backbone for the epoxidation and 30 mg of the redox dye for the modification step.

Table S2 Parameters of copolymers P002 to P008

Copolymer No.	AIIMA [mmol]	BA [mmol]	EA [mmol]	MA [mmol]	AA [mmol]	DMAEMA [mmol]	Weight-% [w/w]
P002	1.50	5.50	/	/	/	3.00	4.9
P004	1.50	/	5.50	/	/	3.00	6.3
P006	1.50	2.75	2.75	/	/	3.00	5.8
P007	1.50	4.00	1.50	3.00	/	/	5.4
P008	1.50	4.00	1.50	/	/	3.00	5.7

TableS3 Parameters of copolymers P015 to P018

Copolymer No.	AIIMA [mmol]	PEGMA [mmol]	BA [mmol]	Weight-% [w/w]
P015	5.00	0.25	4.75	13.3
P017	5.00	0.75	4.25	23.9
P018	5.00	1.00	4.00	9.5

Table S4 Parameters of copolymers P022 to P032

Copolymer No.	AIIMA	BA	PEGMA	TMAEMA	St	DMAEMA	Weight -%
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	[mmol]	[mmol]	[mmol]	[mmol]	[mmol]	[mmol]	[w/w]
P022	5.00	3.25	0.75	/	/	1.00	5.8
P023	25.00	21.30	3.75	/	/	/	12.5
P029	25.00	6.028	3.75	/	/	15.00	13.2
P030	11.00	/	2.00	6.00	/	/	18.7
P032	10.00	7.00	/	/	3.00	/	16.6