Supporting Information for

Enhancing the Recyclability of a Vegetable Oil-Based Epoxy Thermoset through Initiator Influence

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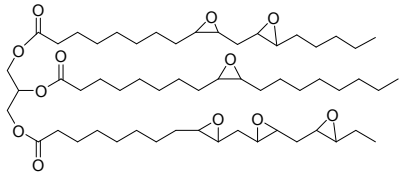
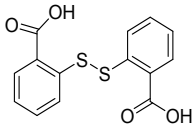
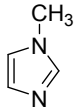
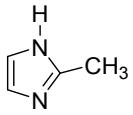
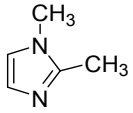
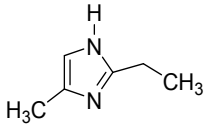
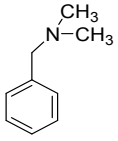
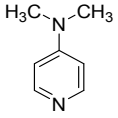
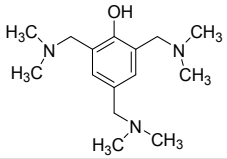
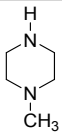
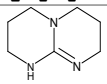
Table S1. Chemical structures and physico-chemical characteristics of selected reagents.

Acronym	Structure and name	Molar mass [g/mol]	Melting temp. [°C]	pKa	Functionality
ELO	 **Epoxidized Linseed Oil**	980	-	-	Matrix **5,5** (5,6 meq/g)
DTBA	 **2,2′-Dithiodibenzoic acid**	306.35	287-290	-	Hardener **2**
IM	 **Imidazole**	68.07	89-91	6.95	Initiator
1-MI	 **1-Methylimidazole**	82.10	-60	7.21	Initiator
2-MI	 **2- Methylimidazole**	82.10	145	7.85	Initiator
1,2 - DMI	 **1,2 Dimethylimidazole**	96.13	37 - 39	8.21	Initiator
2E4MI	 **2-Ethyl 4-Methylimidazole**	110.16	47 - 54	8.68	Initiator
DMBA	 **N,N-Dimethylbenzylamine**	149.19	43 - 45	8.97	Initiator
DMAP	 **Dimethylaminopyridine**	122.17	108 - 110	9.2	Initiator
DMP30		265.39	< -20	8.41 9.12 9.75	Initiator

	2,4,6 Tris (dimethylaminomethyl)phenol				
1-MP	 **1-Methylpiperazine**	100.16	-6	9.14	Initiator
TBD	 **1,5,7-Triazabicyclo[4.4.0]dec-5-ene**	139.20	125 - 130	14.5	Initiator

The epoxy/hardener formulation was optimized using an equimolar ratio between epoxide and acid, according to the Equation S1:

$$R = \frac{Epoxy\ group}{Acid\ group} \qquad (S1)$$

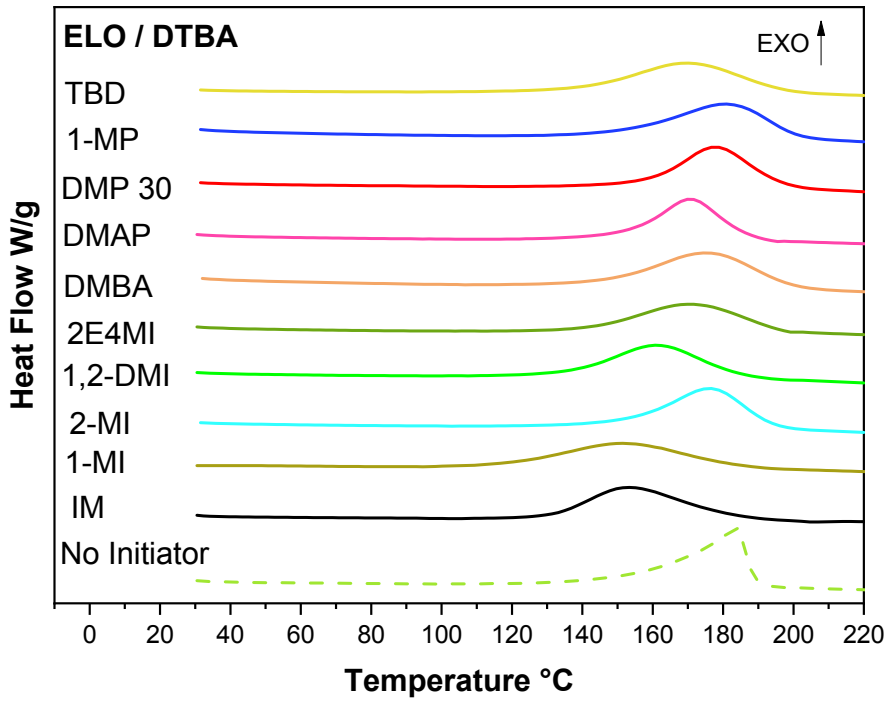


Figure S1. Dynamic DSC scans of the reactions of ELO/DTBA with and without initiators.

Table S2. DSC results during ELO/DTBA curing: Enthalpy of reaction, T_{peak} and Reaction interval.

ELO/DTBA	***ΔH (J/g)***	***T_{peak} (°C)***	***Reaction Interval***	***Curing***	***Post-curing***
No Initiator	248	184	167 - 189	150 °C – 90 Minutes	180°C – 30 Minutes
IM	214	154	130 - 184	130°C – 60 Minutes	170°C – 30 Minutes
1-MI	206	152	117 - 184	120°C – 60 Minutes	170°C – 30 Minutes

2-MI	240	177	151 – 195	150°C – 60 Minutes	180°C – 30 Minutes
1,2-DMI	208	162	135 - 186	130°C – 60 Minutes	170°C – 30 Minutes
2E4MI	186	171	142 - 197	140°C – 60 Minutes	180°C – 30 Minutes
DMBA	260	176	145 - 200	140°C – 60 Minutes	180°C – 30 Minutes
DMAP	206	171	150 - 188	140°C – 60 Minutes	170°C – 30 Minutes
DMP30	220	178	158 - 197	150°C – 60 Minutes	180°C – 30 Minutes
1MP	244	181	152 - 202	150°C – 60 Minutes	180°C – 30 Minutes
TBD	189	170	141 - 198	140°C – 60 Minutes	180°C – 30 Minutes

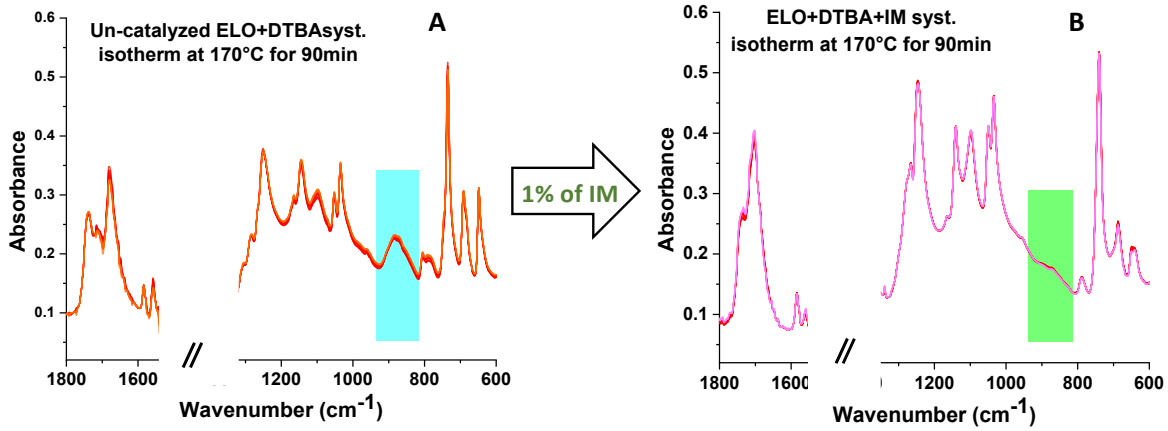


Figure S2. Isothermal ATR-FTIR spectra, collected each 5 min during 90 min for the ELO/DTBA thermoset without initiator (A) and with 1% of imidazole (B).

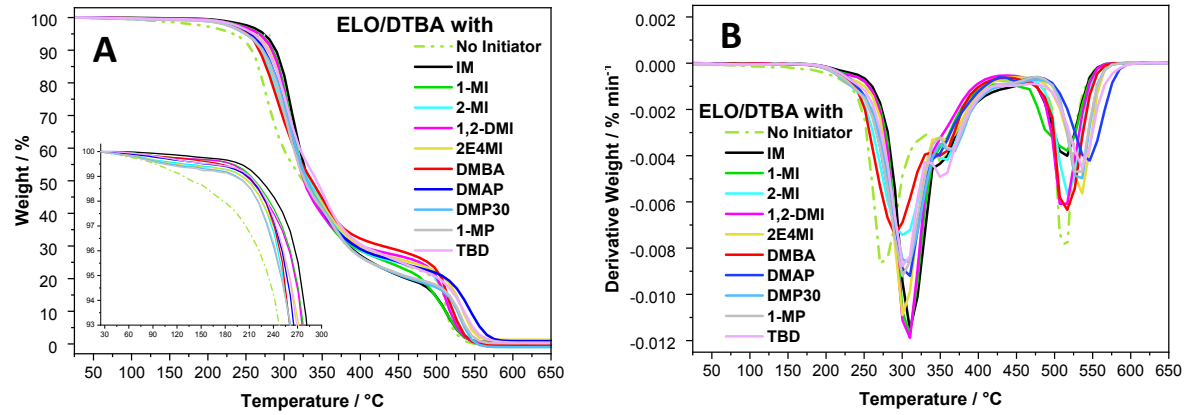


Figure S3. TGA (A) and DTG (B) thermograms of ELO-DTBA curing mixtures.

Table S3. TGA results for ELO/DTBA curing mixtures .

ELO/DTBA	*$T_{5\%}$ (°C)*	*Residue 1st (%)*	*1st Degradation Peak (°C)*	*Final Residue (%)*	*2nd Degradation Peak (°C)*
No Initiator	230	25.2	271	0.39	510
IM	275	19.4	307	0.67	517
1MI	265	24.7	307	0.37	515
2MI	250	25.0	302	0.08	518
1,2DMI	265	27.8	307	0.39	512
2E4MI	260	25.4	302	1.22	535
DMBA	255	29.7	290	0.22	510
DMAP	245	22.2	305	0.77	540
DMP30	245	18.6	305	0.23	530
1MP	250	19.6	302	0.03	535
TBD	260	26.6	305	0.44	540

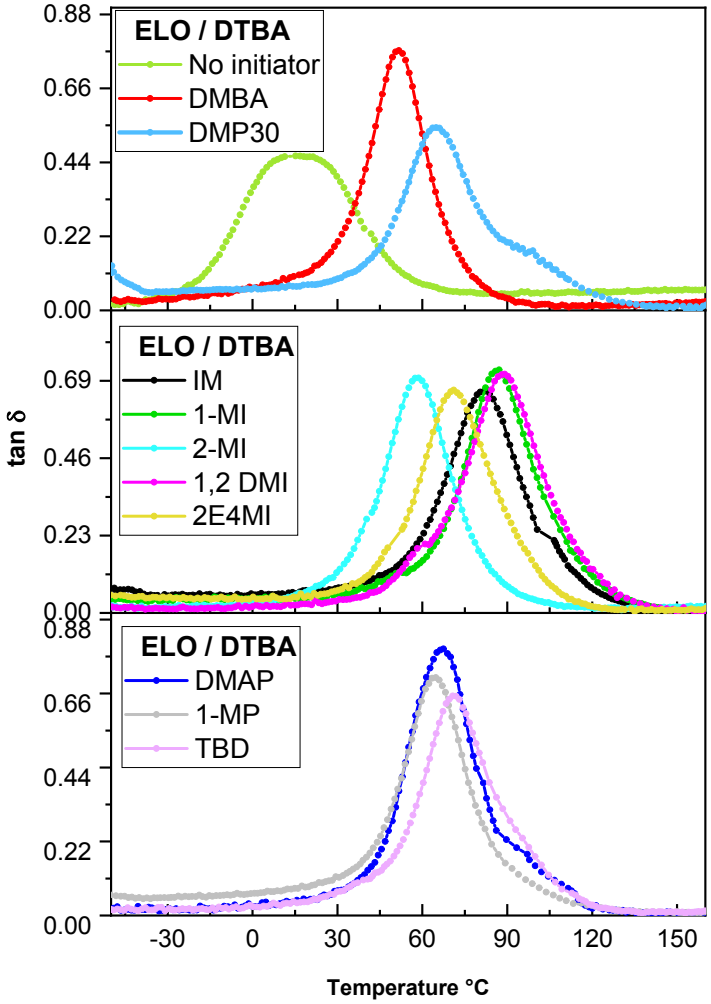


Figure S4. DMA results: tan δ *vs.* temperature for ELO/DTBA thermosets prepared without and with initiators.

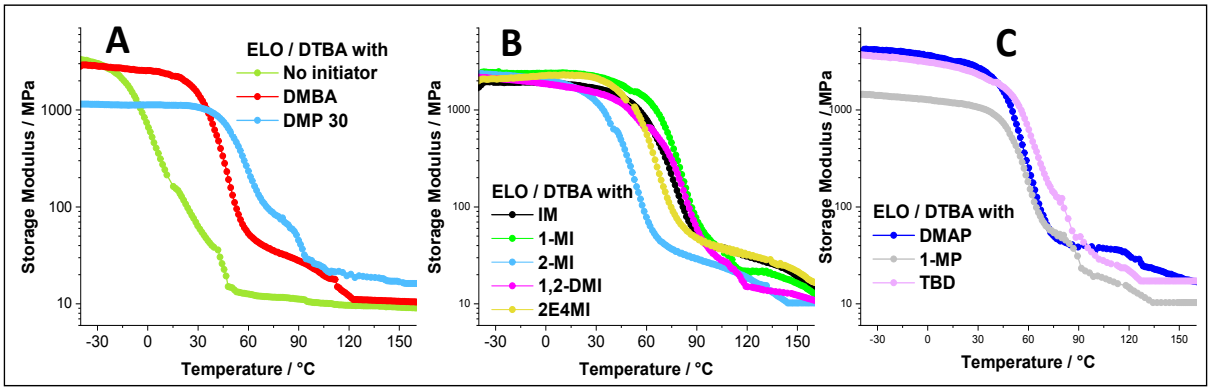


Figure S5. DMA results: storage modulus *vs.* temperature for ELO/DTBA thermosets prepared without and with initiators.

Table S4. DMA data and networks properties for ELO/DTBA thermosets.

	tan *δ* (°C)	**(tan *δ*) max**	***E' at -50 °C (MPa)***	***E' at 160°C (MPa)***	***Crosslinking density (mmol/cm³)***	***Density (g/cm³)***	***M_c (g/mol)***

No Initiator	15	0.45	3390	9.01	0.82	0.905	1104
IM	83	0.66	1560	15.7	1.25	1.025	820
1-MI	87	0.72	2030	11.2	1.03	0.988	876
2-MI	59	0.70	2100	6.1	1.14	0.788	691
1,2-DMI	89	0.71	1820	7.23	1.09	0.743	708
2E4MI	71	0.66	1900	15.6	1.84	0.935	508
DMBA	52	0.77	2660	7.2	0.98	0.896	914
DMAP	66	0.79	2380	8.2	1.18	0.913	774
DMP30	65	0.54	800	16.1	1.97	0.782	400
1-MP	65	0.70	1230	10.3	0.96	0.775	807
TBD	71	0.65	2560	17.17	1.62	0.837	517

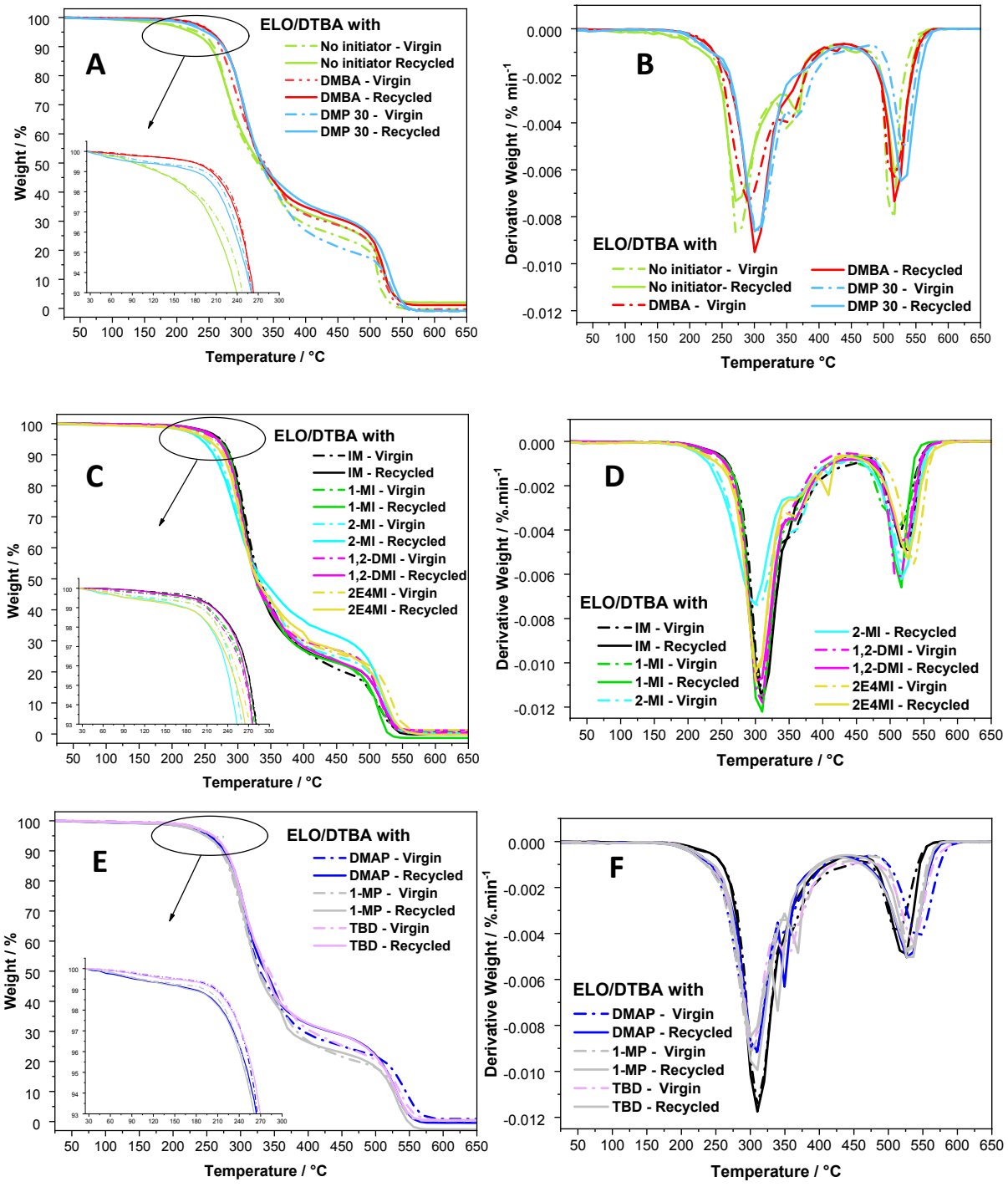


Figure S6. TGA and DTG of the virgin and recycled thermosets: No initiator, DMBA and DMP30 (A and B), IM, 1-MI, 2-MI, 1,2-DMI and 2E4MI (C and D) and DMAP, 1-MP and TBD (E and F).

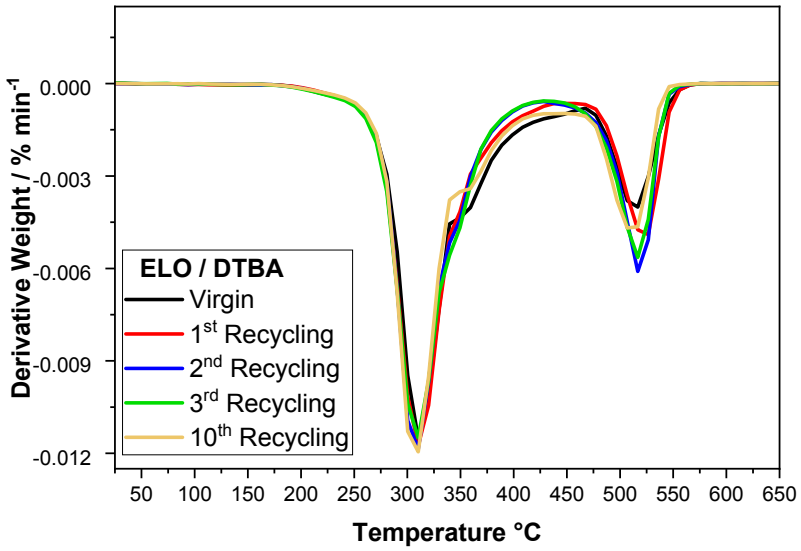


Figure S7. DTG curves of the virgin and recycled ELO/DTBA/IM materials after several reprocessing steps.

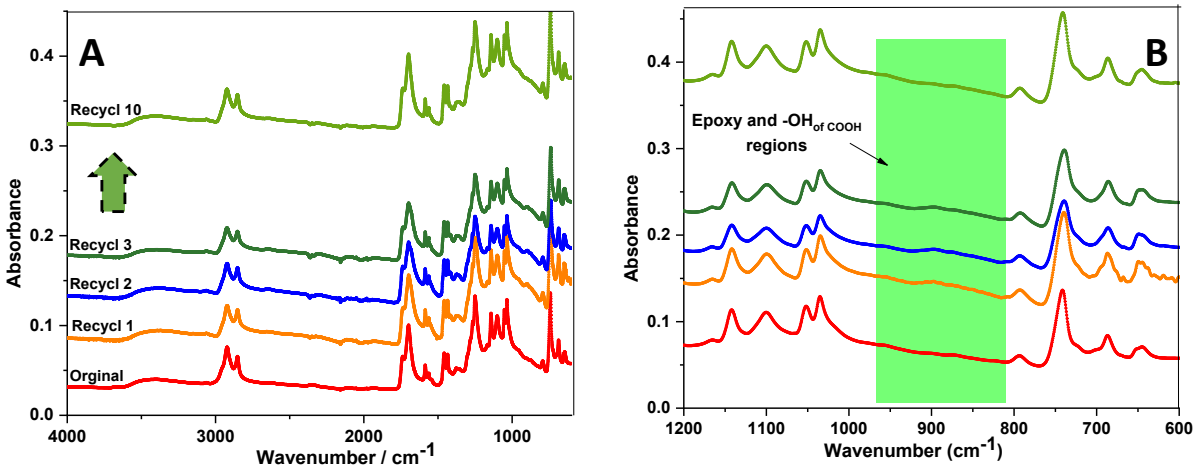


Figure S8. ATR-FTIR spectra of the original and reprocessed thermosets (ten consecutive cycles were applied): (A): whole spectra from 4000-600 cm^{-1}; (B): zoom in the region of epoxy groups and -OH of carboxylic groups (DTBA) from 1200 to 600 cm^{-1}.

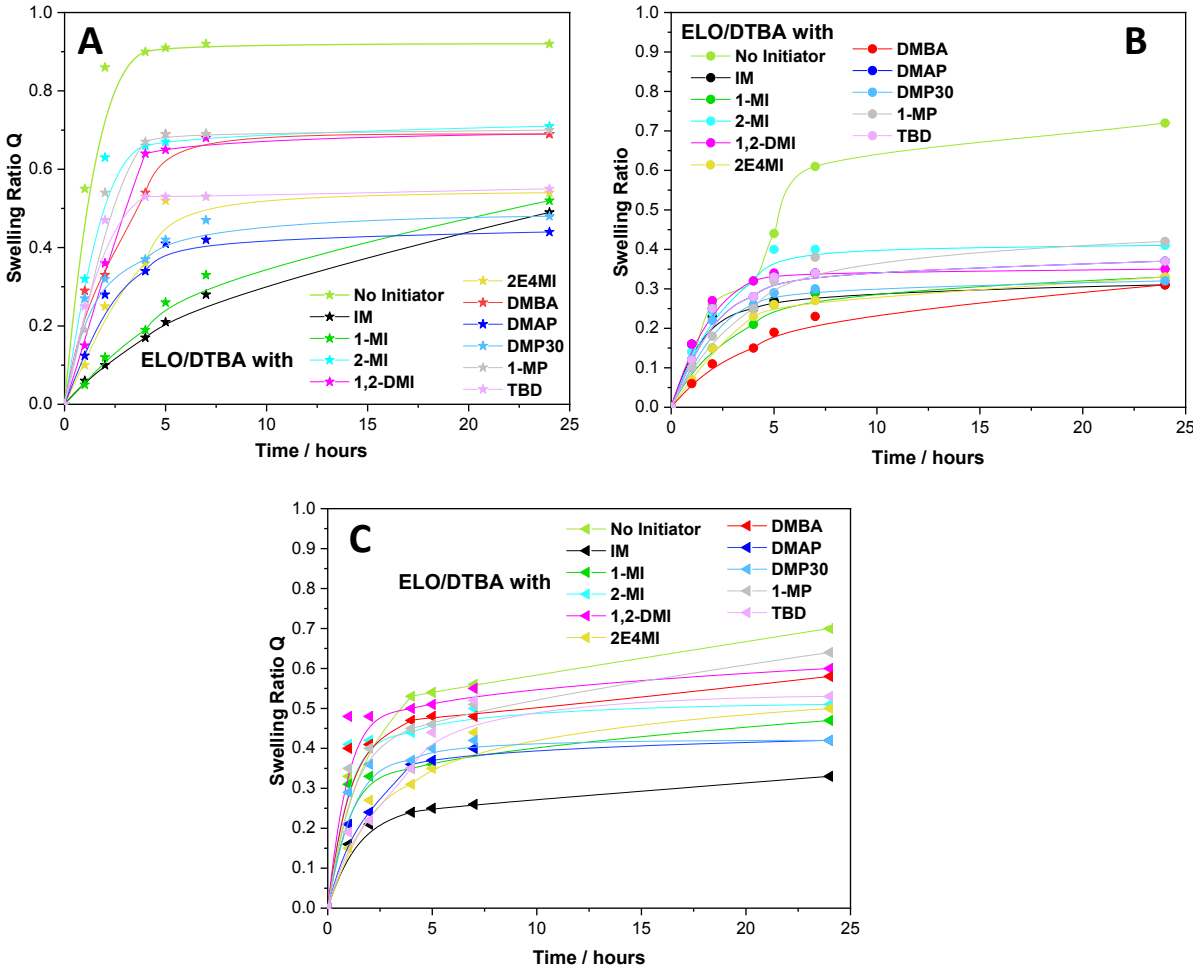


Figure S9. Swelling ratio of the virgin ELO/DTBA thermosets prepared with and without initiator in Toluene (**A**), Methanol (**B**) and Acetone (**C**) for 24 hours.

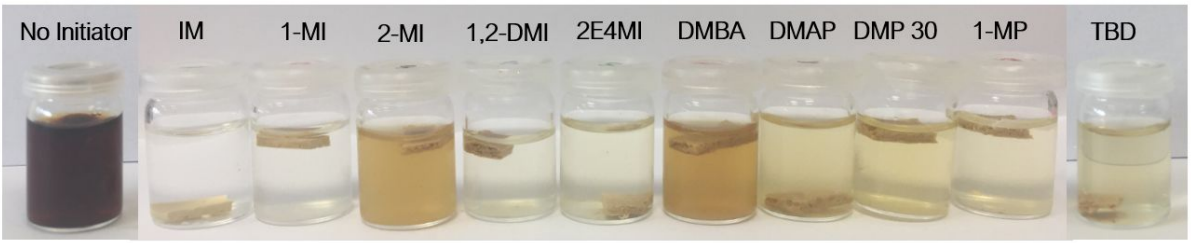


Figure S10. Solvent test after 24 hours in NaOH solution 1N.

Table S5. The reprocessing conditions (temperature and duration) performed at a pressure of 60 bars

ELO/DTBA formulations	**Temperature (°C)**	**Time (minutes)**
No Initiator	170	10
IM	160	30
1-MI	150	20
2-MI	170	40
1,2-DMI	160	30
2E4MI	170	40
DMBA	170	30
DMAP	160	20
DMP30	170	30
1-MP	170	40
TBD	170	30

Table S6. Properties of the virgin and recycled thermoset resins.

		Tg (°C) by DSC	***tan δ (°C)***	***Crosslink density (mmol/cm^3)***	***$T_{5\%}$ (°C)***
No Initiator	Virgin	13	15	0.82	230
	Recycled	12	13	0.30	220
IM	Virgin	76	83	1.25	275
	Recycled	74	97	0.81	275
1-MI	Virgin	74	87	1.03	265
	Recycled	72	95	0.38	265
2-MI	Virgin	46	59	1.14	250
	Recycled	42	63	0.34	250
1,2-DMI	Virgin	70	89	1.05	265
	Recycled	64	87	0.35	265
2E4MI	Virgin	64	71	1.84	260
	Recycled	59	77	0.38	252
DMBA	Virgin	37	52	0.98	255
	Recycled	36	58	0.21	252
DMAP	Virgin	55	66	1.18	255
	Recycled	50	63	0.79	245
DMP30	Virgin	65	65	1.97	245
	Recycled	56	69	0.58	245
1-MP	Virgin	64	65	0.96	250
	Recycled	57	73	0.23	245
TBD	Virgin	61	71	1.62	260
	Recycled	57	57	0.53	260

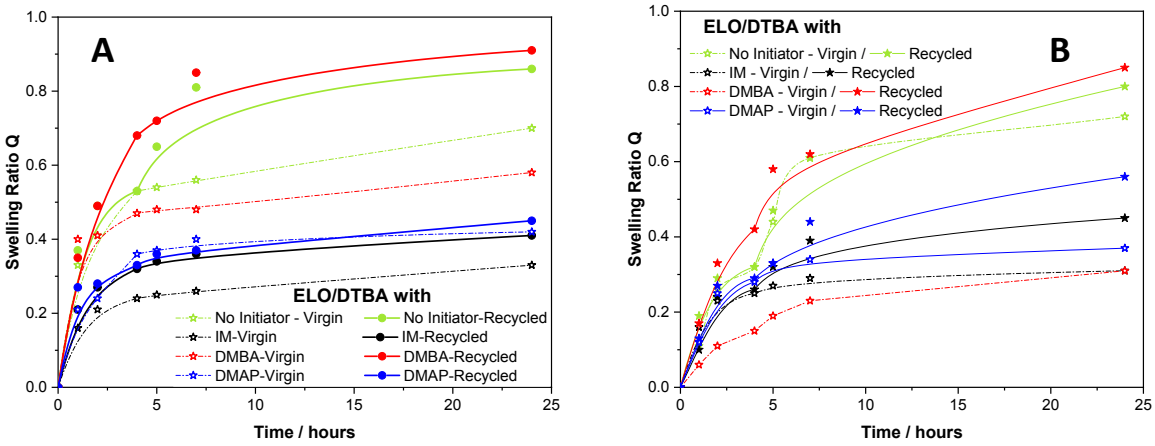


Figure S11. Swelling test in Acetone (**A**) and Methanol (**B**) for the virgin and reprocessed thermoset resins.

Table S7. ELO/DTBA/IM thermoset resin after several reprocessing cycles.

Virgin	1st Recycling	2nd Recycling	3rd Recycling	10th Recycling