

## Surface-Initiated Reversible Addition Fragmentation Chain Transfer of fluoromonomers: an efficient tool to improve interfacial adhesion in piezoelectric composites

### Supporting Information

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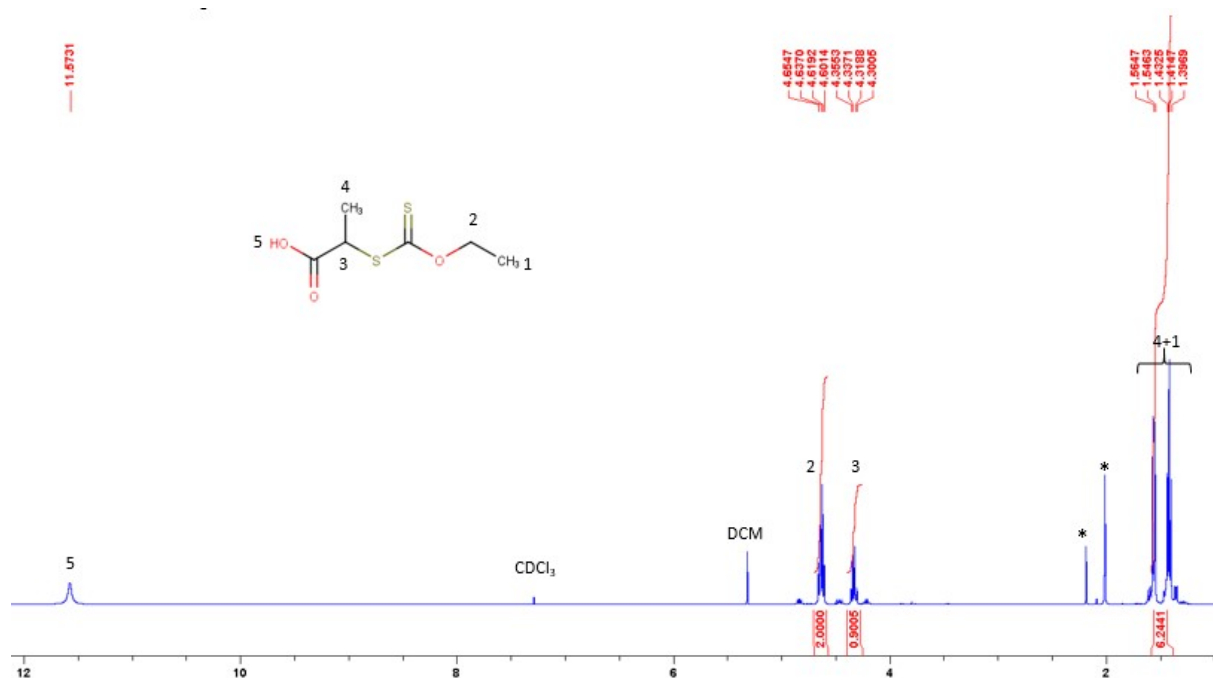


Figure S1. <sup>1</sup>H NMR spectrum of XA-acid recorded in CDCl<sub>3</sub>. \* signals of residual acetone and 2-bromopropionic acid.

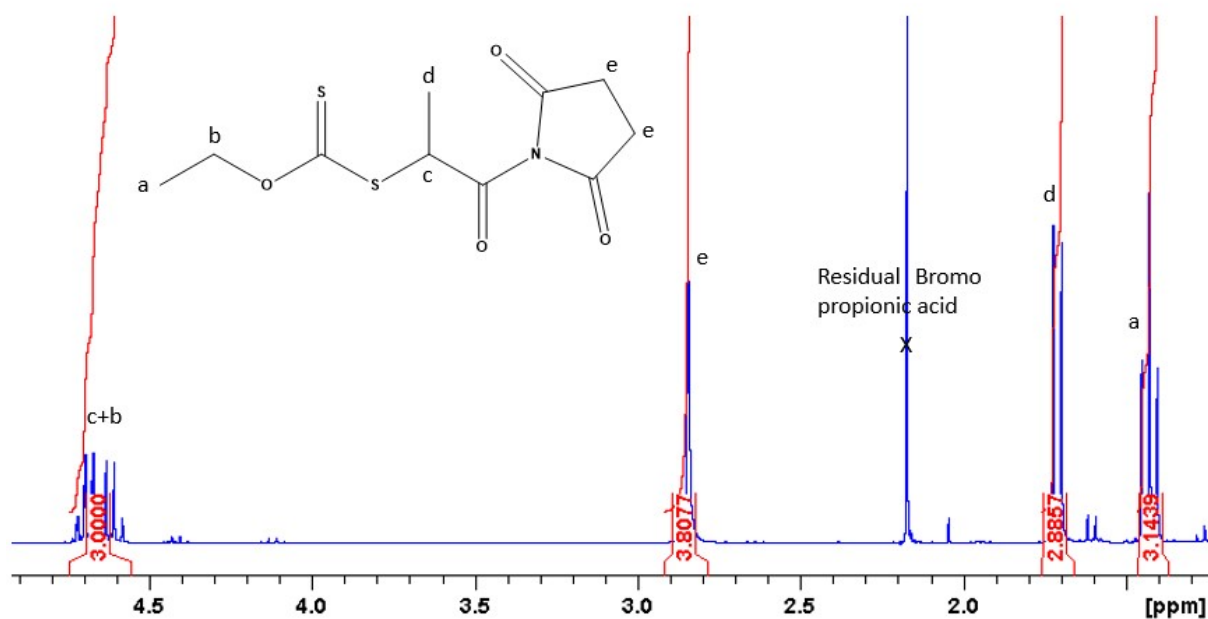


Figure S2. <sup>1</sup>H NMR spectrum of NHS-XA recorded in CDCl<sub>3</sub>.

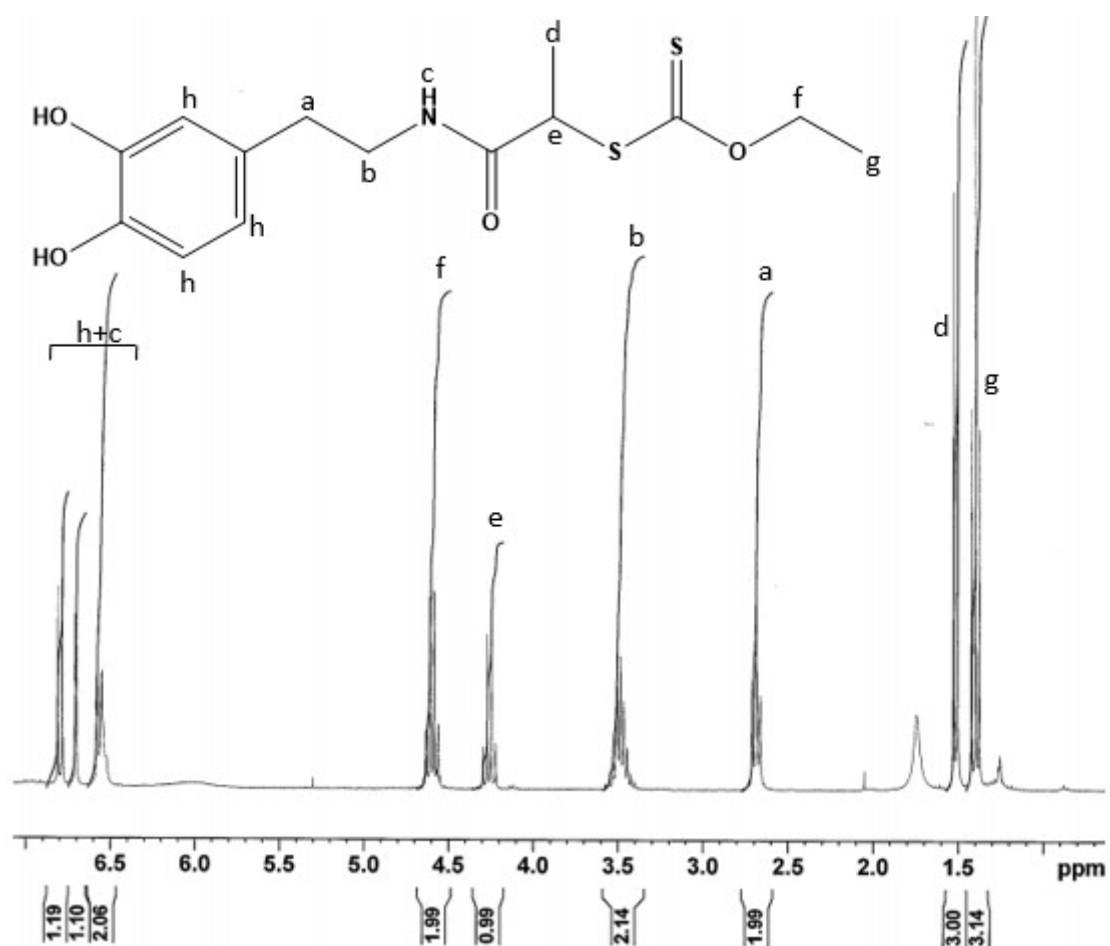


Figure S3. <sup>1</sup>H NMR spectrum of the DOPA-XA, recorded in CDCl<sub>3</sub>.

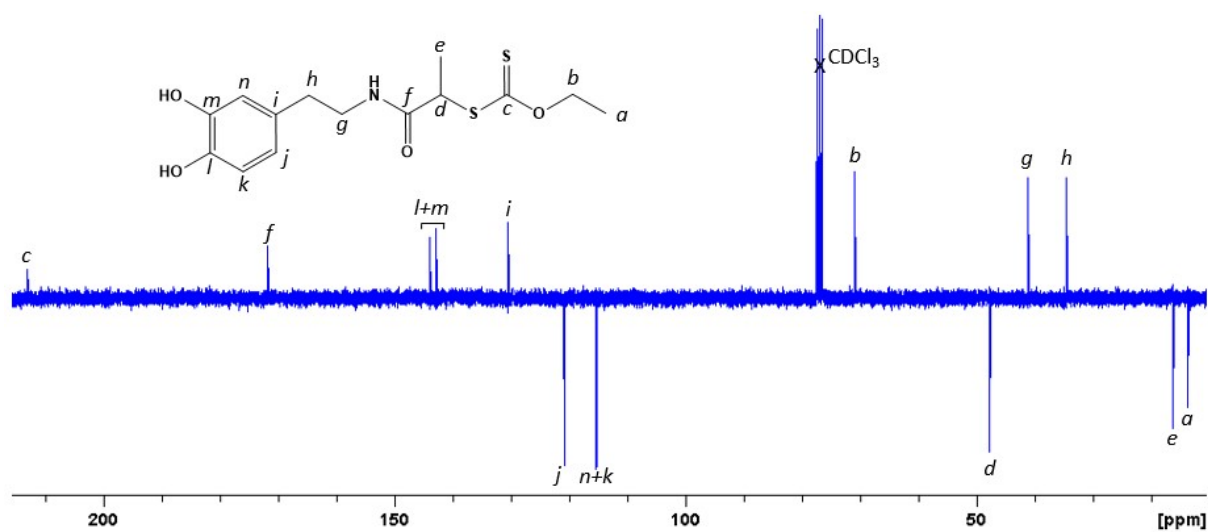


Figure S4.  $^{13}\text{C}$  APT NMR experiment of the DOPA-XA, recorded in  $\text{CDCl}_3$ .

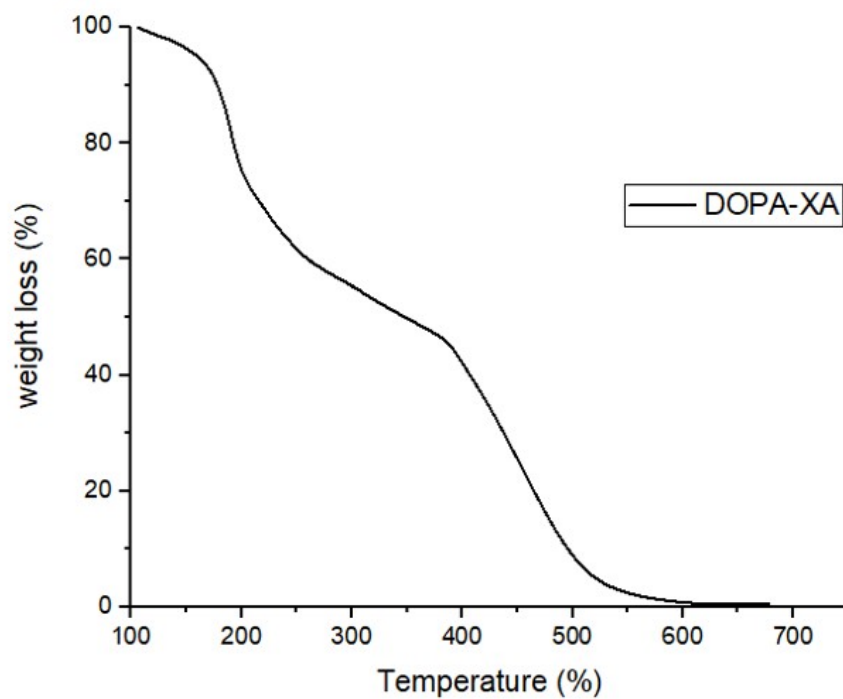


Figure S5. TGA thermogram of DOPA-XA.

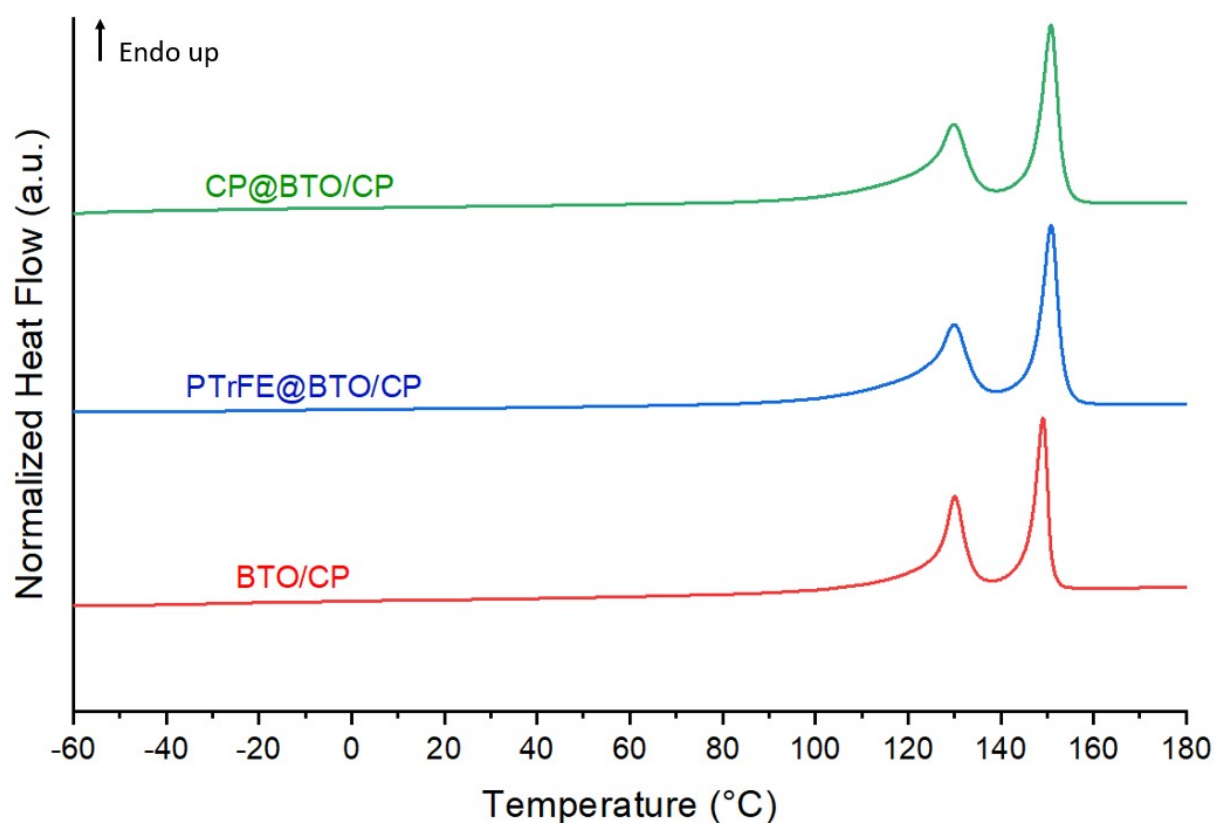


Figure S6. DSC thermograms of the composites with non-modified BTO particles, the PTrFE@BTO particles and with P(VDF-co-TrFE)@BTO particles. The filler content is around 20wt%.

Table S1. XPS quantification (%) of the fonctionnalized particles.

Sample	BTO	DOPA-XA@BTO	PTrFE@BTO	P(VDF-co-PTrFE)@BTO
O 1s	38.6	27.0	16.7	24.7
C 1s	41.4	61.8*	42.0	41.0
Ba 3d	9.0	2.7	3.8	6.1
Ti 2p	11.1	4.1	5.1	8.3
N 1s	-	1.3	0.8	0.8
S 2p	-	3.1	1.5	1.5
F 1s	-	-	30.1	17.6

\* The relatively high carbon ratio of DOPA-XA surface is probably due to contamination of the sample. It affects the relative atomic ratio and increases the apparent “masking effect” of DOPA-XA on the BTO surface.

Equation S1. Grafting density calculations. Weight losses are calculated using TGA. WL(polymer) stands for polymer weight loss and WL(residue) stands for weight loss of raw BTO.

$$\text{Graft density (Polymer)} = \frac{N(\text{polymer})}{S(\text{BTO})} \quad S(\text{BTO}) = \text{Surface BTO}$$

$$\text{Graft density (Polymer)} = \frac{n(\text{polymer}) * Na}{S(\text{BTO})} \quad Na = 6.022 * 10^{23}$$

$$\text{Graft density (Polymer)} = \frac{m(\text{polymer}) * Na}{S(\text{BTO}) * Mn(\text{polymer})}$$

$$\text{Graft density (Polymer)} = \frac{m(\text{polymer}) * Na}{S(\text{BTO}) * Mn(\text{polymer})}$$

$$\frac{m(\text{polymer})}{m(\text{BTO})} = \frac{WL(\text{polymer}) - \frac{WL(\text{residue})}{100 - WL(\text{residue})} * (100 - WL(\text{Polymer}))}{100 - WL(\text{polymer})}$$

$$\text{Graft density (Polymer)} = \frac{m(\text{BTO}) * Na}{S(\text{BTO}) * Mn(\text{polymer})} * \frac{WL(\text{polymer}) - \frac{WL(\text{residue})}{100 - WL(\text{residue})} * (100 - WL(\text{Polymer}))}{100 - WL(\text{polymer})}$$

$$\text{Graft density (Polymer)} = \frac{\text{Specific surface (BTO)} * Na}{Mn(\text{polymer})} * \frac{WL(\text{polymer}) - \frac{WL(\text{residue})}{100 - WL(\text{residue})} * (100 - WL(\text{Polymer}))}{100 - WL(\text{polymer})}$$