

Electronic Supplementary Information

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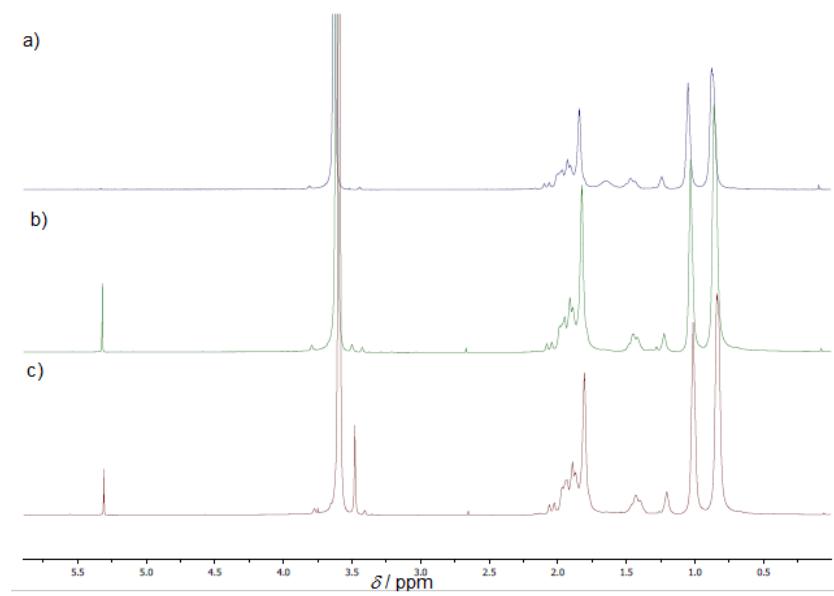


Figure S1. ^1H -NMR (CDCl_3) of $\text{Ta}_x@\text{PMMA}_{\text{brown}}$ copolymer pellets in the oxidised form: a) 1 wt%, b) 2 wt% and c) 3 wt%

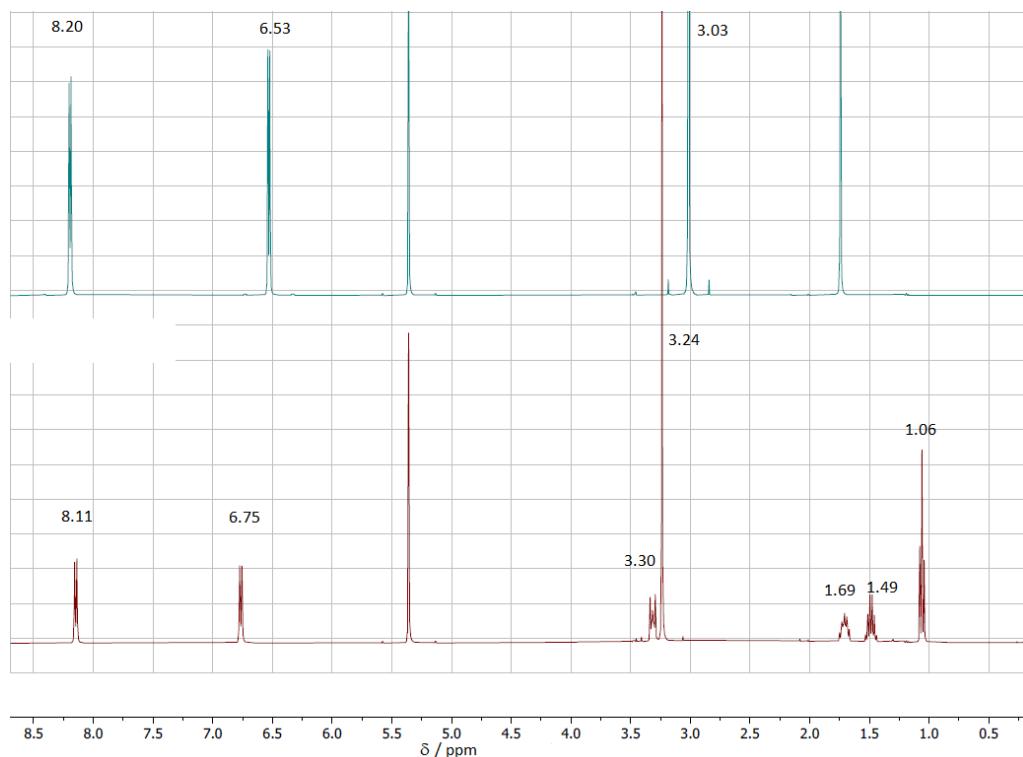


Figure S2. ^1H -NMR spectra (CD_2Cl_2) of DMAP (top) and $(\text{TBA})_2[\{\text{Ta}_6\text{Br}^i_{12}\}\text{Br}^a_6]$ with two equivalents of DMAP (bottom).

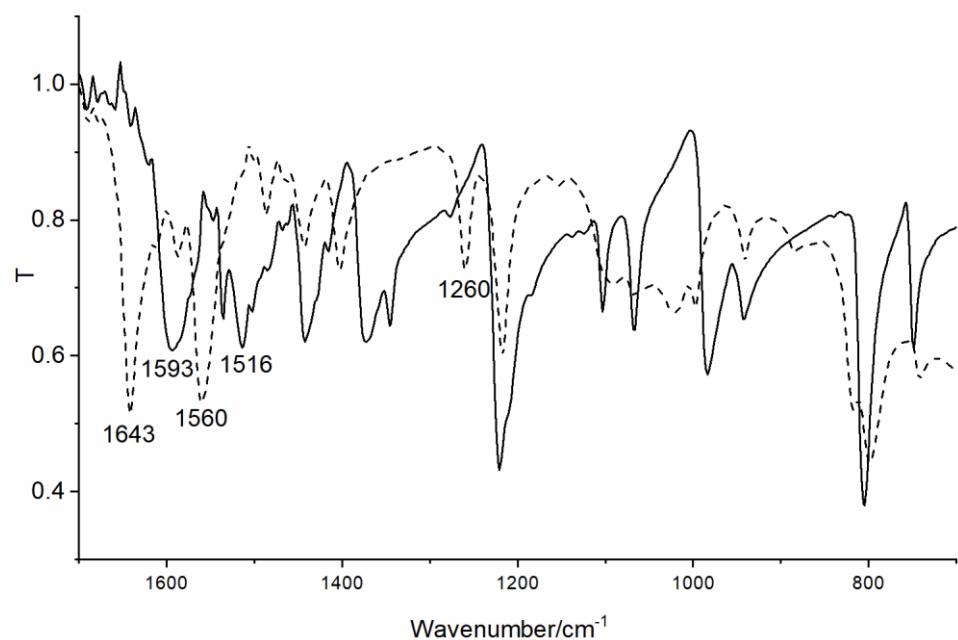


Figure S3. Mid-FTIR spectra of DMAP (solid) and (TBA)₂[{{Ta₆Br}₁₂}Br₃] with two equivalents of DMAP (dash).

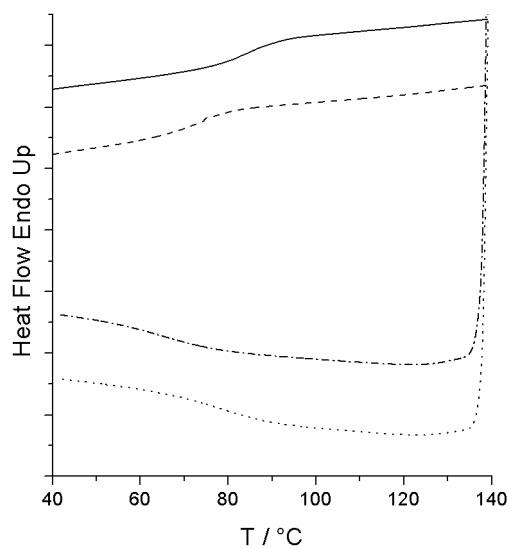


Figure S4. DSC thermograms of $\text{Ta}_2\text{Ta}(\text{Ph}_3)_2\text{Cl}_2\text{Ta}(\text{Ph}_3)_2\text{Cl}_2$. (solid) oxidized state heating cycle and (dot) cooling cycle and (dash) reduced state heating cycle and (dash dot) cooling cycle.

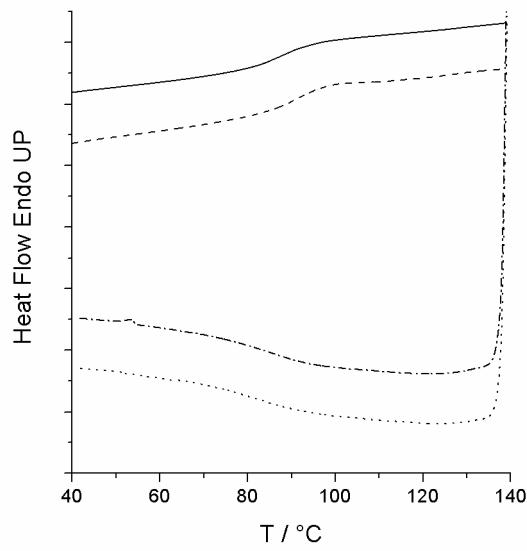


Figure S5. DSC thermograms of Ta₂@PMMA. (solid) oxidized state heating cycle and (dot) cooling cycle and (dash) reduced state heating cycle and (dash dot) cooling cycle.

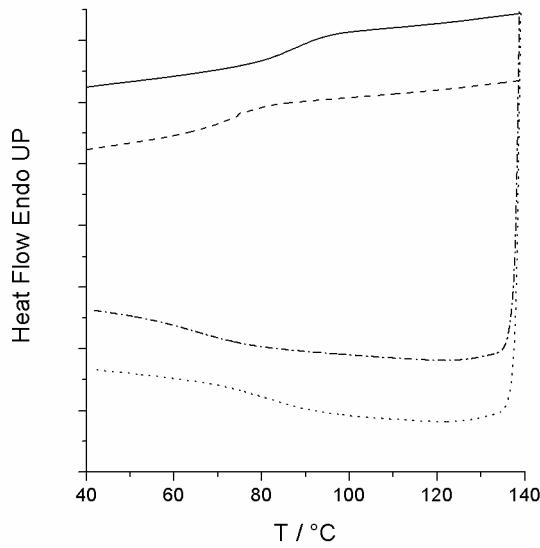


Figure S6. DSC thermograms of Ta₃@PMMA. (solid) oxidized state heating cycle and (dot) cooling cycle and (dash) reduced state heating cycle and (dash dot) cooling cycle.

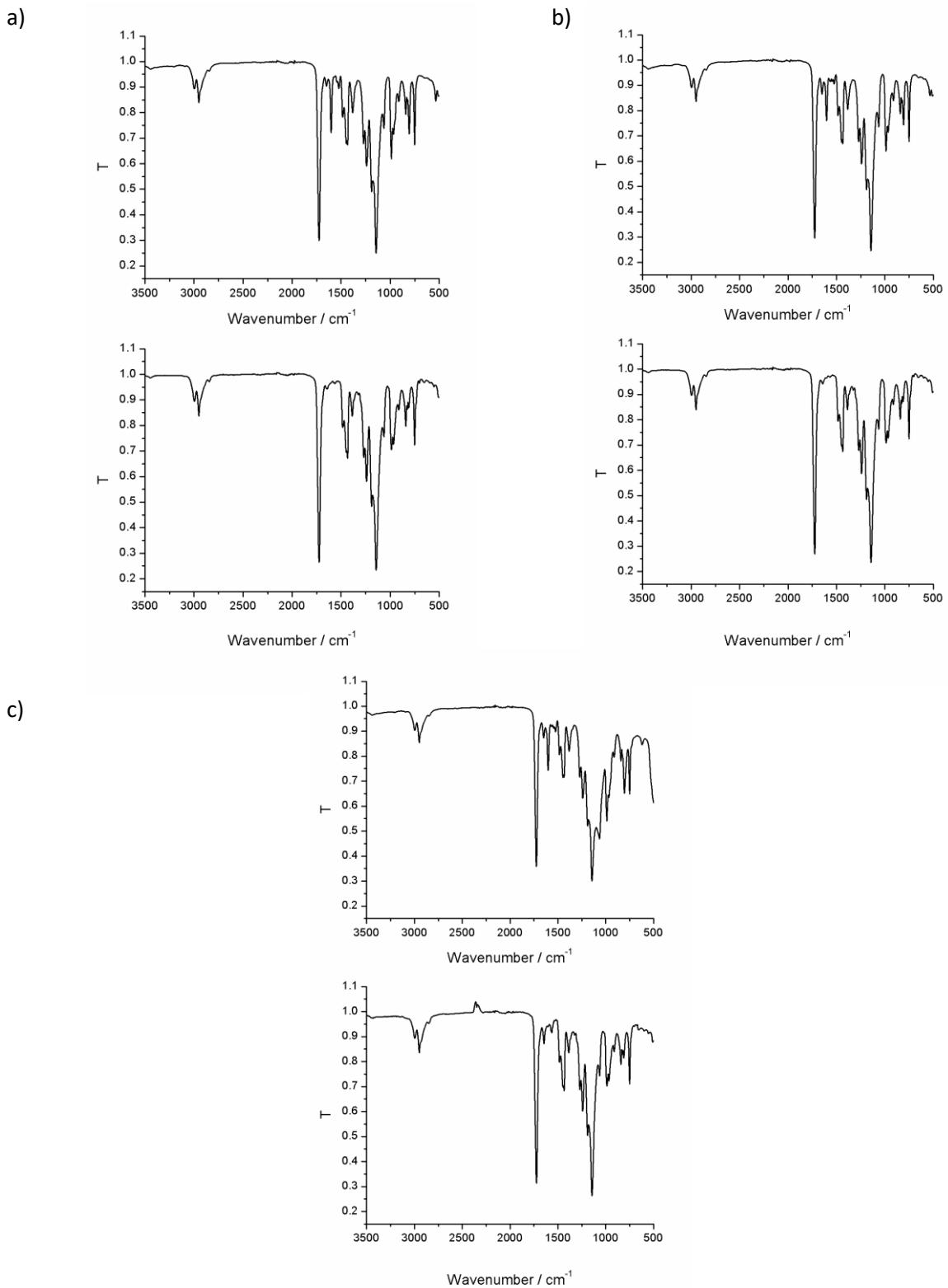


Figure S7. Mid-FTIR a) Ta_1@PMMA_{brown} (bottom), Ta_1@PMMA_{green} (top), b) Ta_2@PMMA_{brown} (bottom), Ta_2@PMMA_{green} (top), c) Ta_3@PMMA_{brown} (bottom), Ta_3@PMMA_{green} (top).

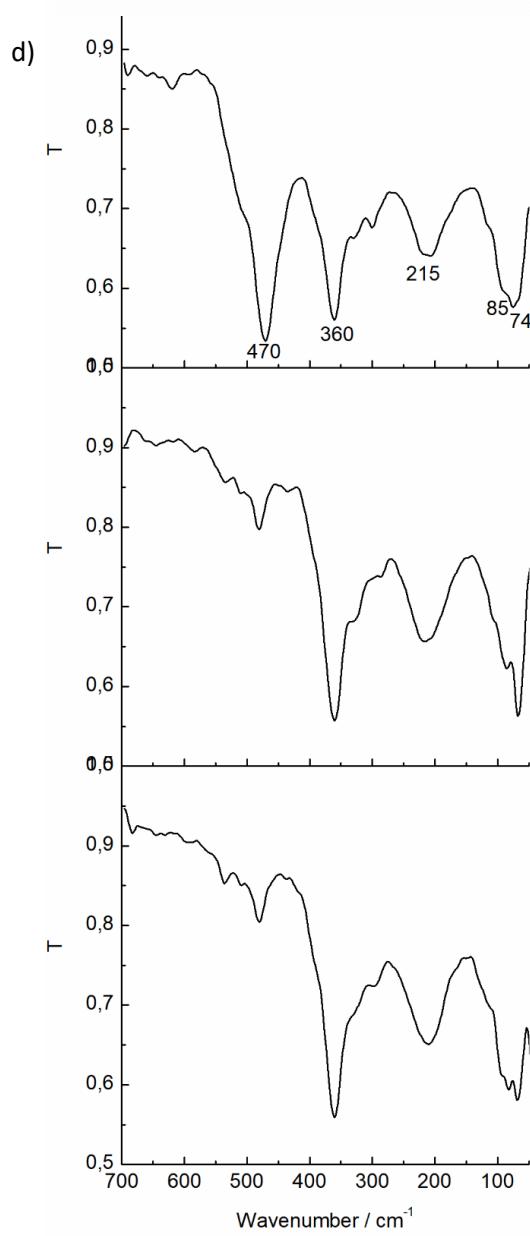
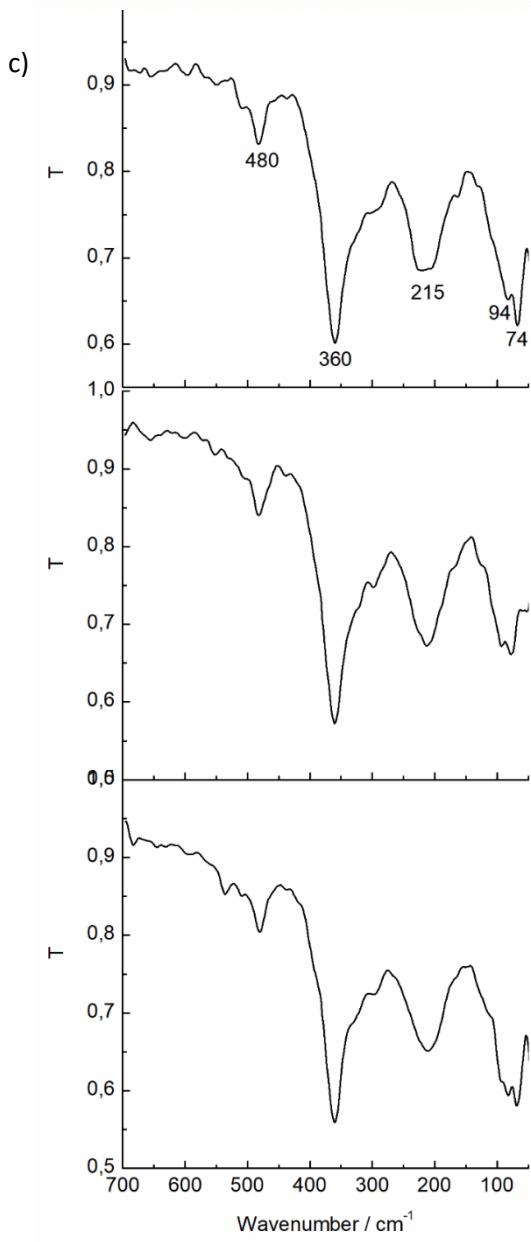
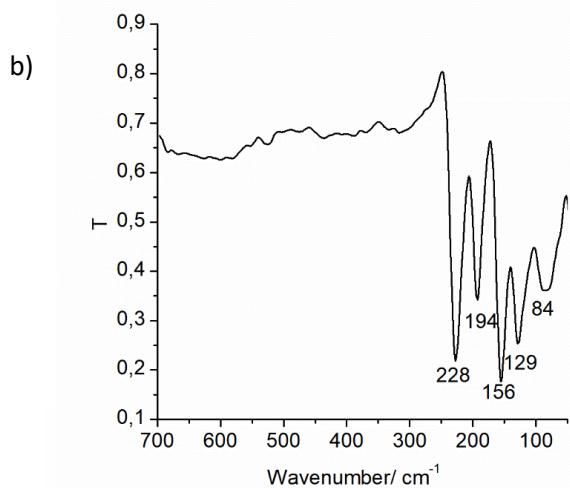
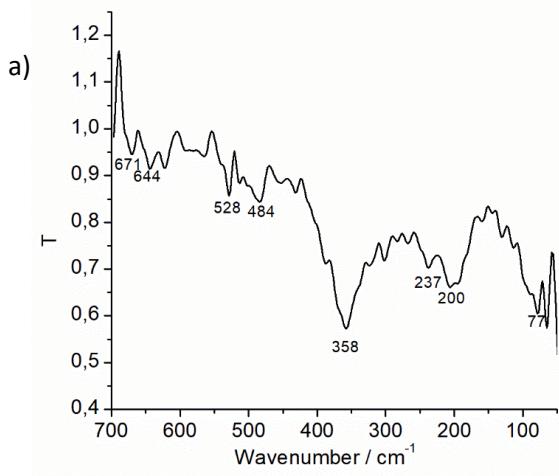


Figure S8. Far IR spectra of a) PMMA, b) TBA₂Ta₆Br₁₈, c) Ta_1@PMMA_{brown} (bottom), Ta_2@PMMA_{brown} (medium), Ta_3@PMMA_{brown} (top), d) Ta_1@PMMA_{green} (bottom), Ta_2@PMMA_{green} (medium), Ta_3@PMMA_{green} (top).

Table S1. Main far IR absorption bands for $[\{Ta_6Br^{i}_{12}\}Br^{a}_6]^{2-}$, PMMA and Ta_2@PMMA composites

Reported for (TEA) ₂ $[\{Ta_6Br^{i}_{12}\}Br^{a}_6]^{1-}$	Found for (TBA) ₂ $[\{Ta_6Br^{i}_{12}\}Br^{a}_6]$	Reported for PMMA	Found for PMMA	Ta- 2@PMMAox	Ta-2@PMMA- red
-----	-----	483 w	484 w	482 w	480 w
-----	-----	365 br, vs	360 br, vs	360 br, vs	360 br, vs
230 vs	228 vs	228 vbr	237 w	227-200 vbr, m	221-200, vbr, m
196 vs	194 vs	----	200 vbr, m	----	----
161 vs	156 vs	----	----	----	----
134 s	129 s	----	----	----	----
98 m	---	----	----	----	----
82 m	84 br, m	----	94 -75 br, m, sp	90 – 75 br, m, sp	95 – 75, br, m, sp

vs: very strong; s: strong; m: medium; w: weak; br: broad; vbr: very broad; sp: splitted

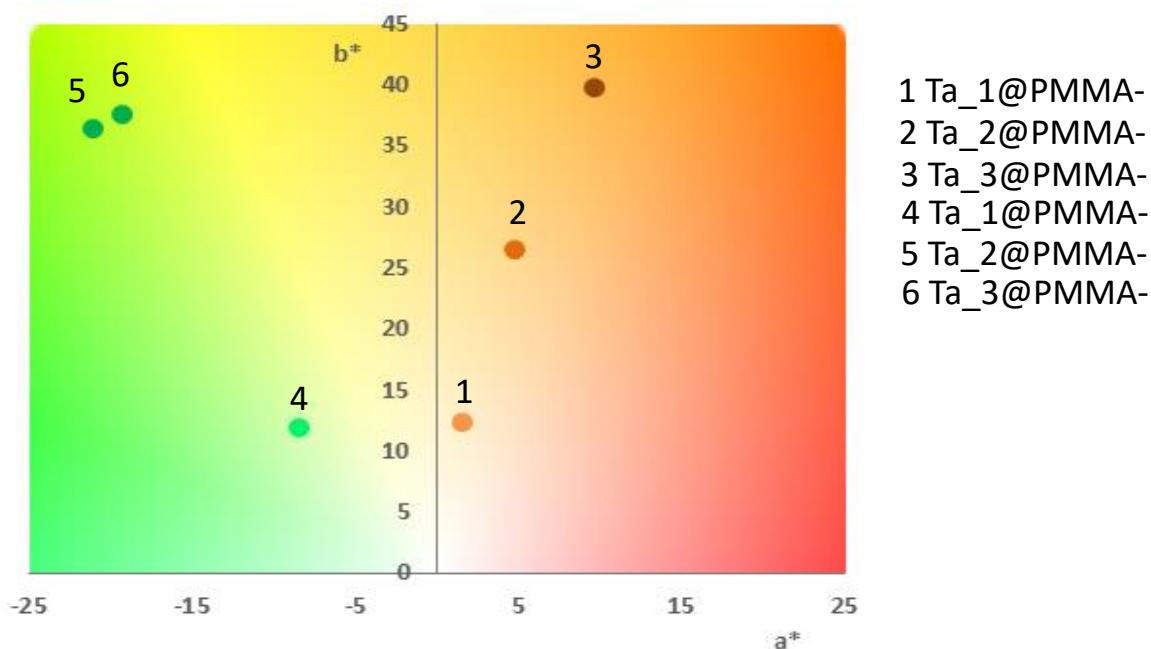


Figure S9. CIE chromaticity coordinates of Ta_x@PMMA (VEC = 14 in the oxidized form and VEC = 16 in the reduced form)

Table S2. L*, a*, b color coordinates of Ta_x@PMMA composites

sample	L*	a*	b*	Color
Ta-1@PMMA _{brown}	92.2	1.49	12.38	
Ta-2@PMMA _{brown}	84.64	4.71	26.45	
Ta-3@PMMA _{brown}	79.18	9.52	39.91	
Ta-1@PMMA _{green}	92.14	-8.53	11.87	
Ta-2@PMMA _{green}	76.47	-21.12	36.36	
Ta-3@PMMA _{green}	71.75	-19.33	37.53	