

## Supplementary Material

### **Novel Polyurethanes Based on Recycled Polyethylene Terephthalate: Synthesis, Characterization, and Formulation of Binders for Environmentally Responsible Rocket Propellants**

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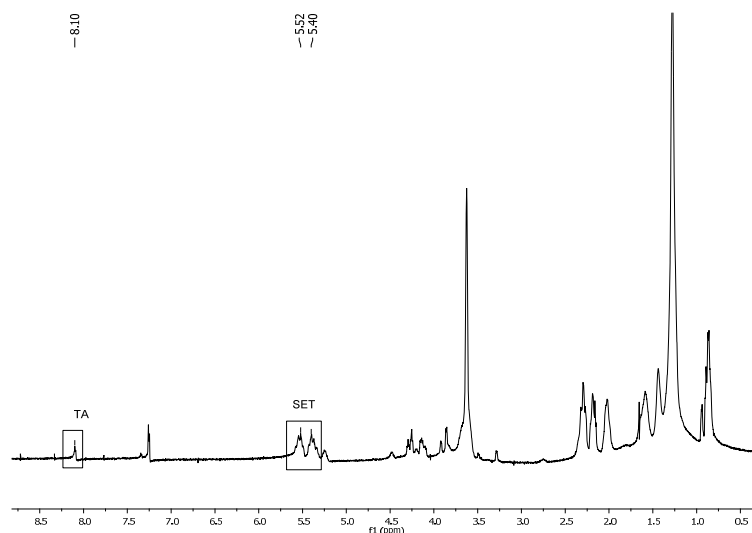
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<sup>‡</sup> co-first authors, equally contributed to this work

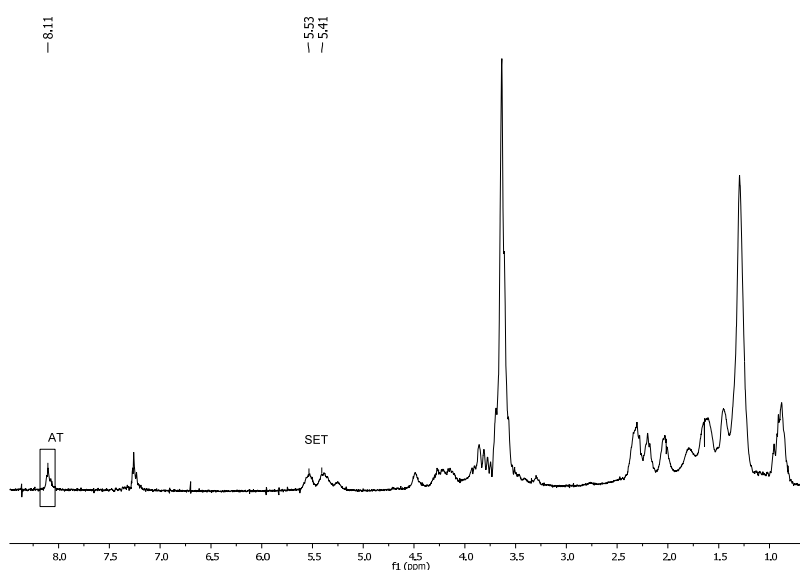
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## Table of Contents

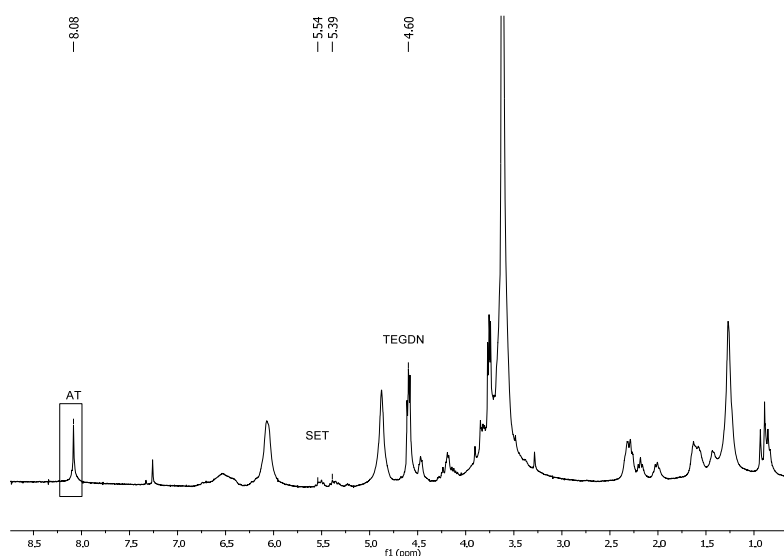
Figure S1 – <sup>1</sup> H- NMR spectra for SRP1	3
Figure S2 – <sup>1</sup> H- NMR spectra for SRP2	3
Figure S3 – <sup>1</sup> H- NMR spectra for SRP1T2	3
Figure S4 – <sup>1</sup> H- NMR spectra for SRP2T2	4
Figure S5 – FT-IR spectra of RP2 polyurethane compounds	4
Figure S6 – Binder characterization regarding aspect and time reaction	5
Figure S7 – SEM images of polyurethanes formulations based on RP1 and RP2 for 3:2 and 2:1 ratio with and without TEGDN	6
Figure S8 EDX mapping of the polyurethane formulations	6
Figure S9 – Tensile charts for PU formulations	7
Figure S10 – Tensile charts for composite propellant formulations	8
Figure S11 – Compressive test charts for composite formulations	9
Figure S12 – SEM-EDX images of composite propellant formulations	11
Figure S13 – EDX spectra for free oxidizer composite formulations	11
Figure S14 – SEM-EDX images of free binder solid mixture	12
Figure S15 – EDX spectra for free binder solid mixture	12
Figure S16 – Stand burner equipped with Subscale Rocket Motor TRM-35	13
Figure S17 – Structural configuration of ECP_D5	13
Figure S18 – Pyrotechnic composition for propellant ignition	13
Figure S19 – Flame configuration of ECP_D5 during the combustion	14
Figure S20 – Behavior of PUs during manual bending	14
Figure S21 – Structural configurations of composite propellants for mechanical analysis	15
Table S1. Weight and atomic composition of free oxidizer composite formulations	15
Table S2. Weight and atomic composition of free binder solid mixture	15
Table S3. Thermal characteristics for polyurethane films, composite propellant formulations, and energetic plasticizer	15
Table S4. Thermal properties of synthesized polyurethanes	15
Table S5. The decomposition process of the polyurethanes (weight loss versus temperature)	16
Table S6. Heat of combustion, specific volume and T <sub>g</sub> for our new composite propellants in comparison with the existing commercial formulations	16



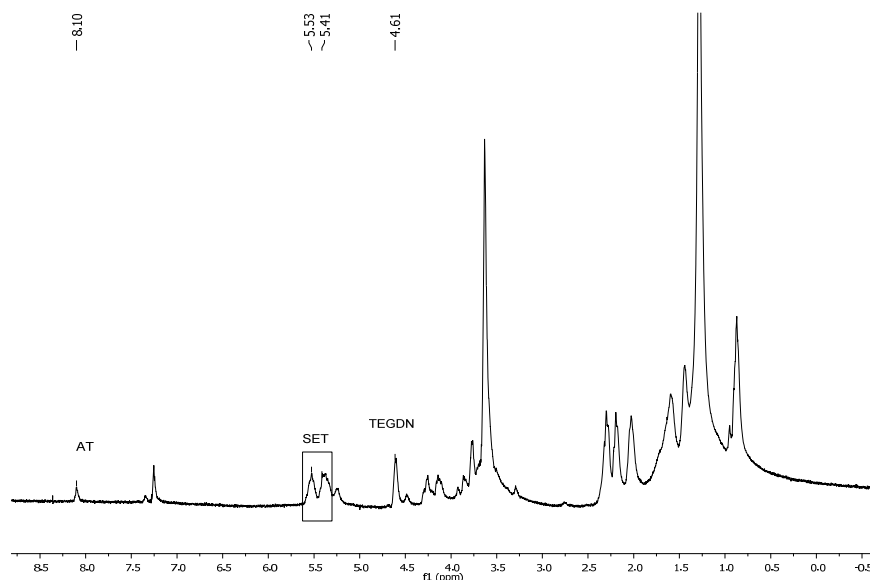
**Figure S1.** –  $^1\text{H}$ - NMR spectra for SRP1.



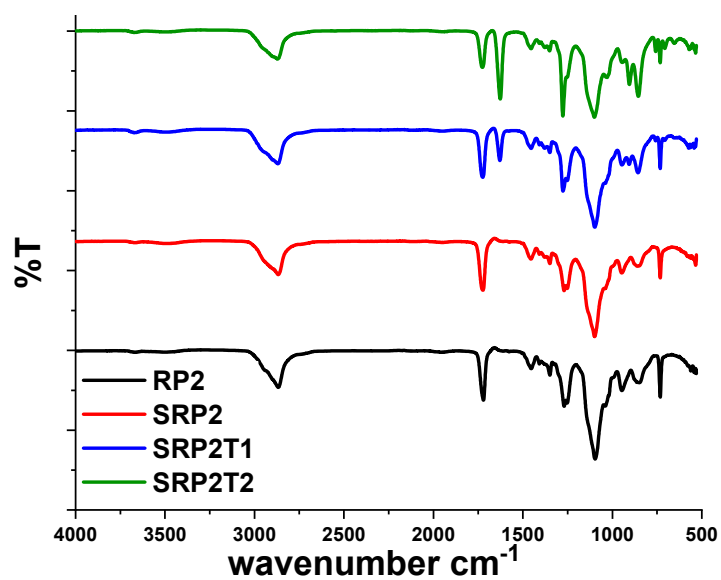
**Figure S2.** –  $^1\text{H}$ - NMR spectra for SRP2.



**Figure S3.** –  $^1\text{H}$ - NMR spectra for SRP1T2.











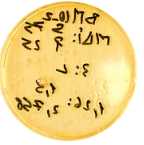

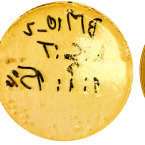


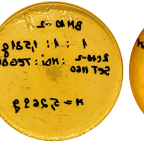


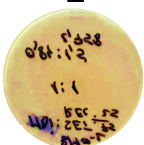
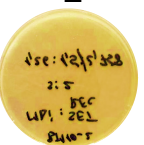

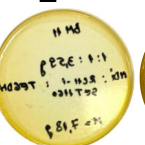




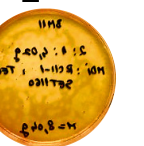


**Figure S4.** –  $^1\text{H}$ - NMR spectra for SRP2T2.

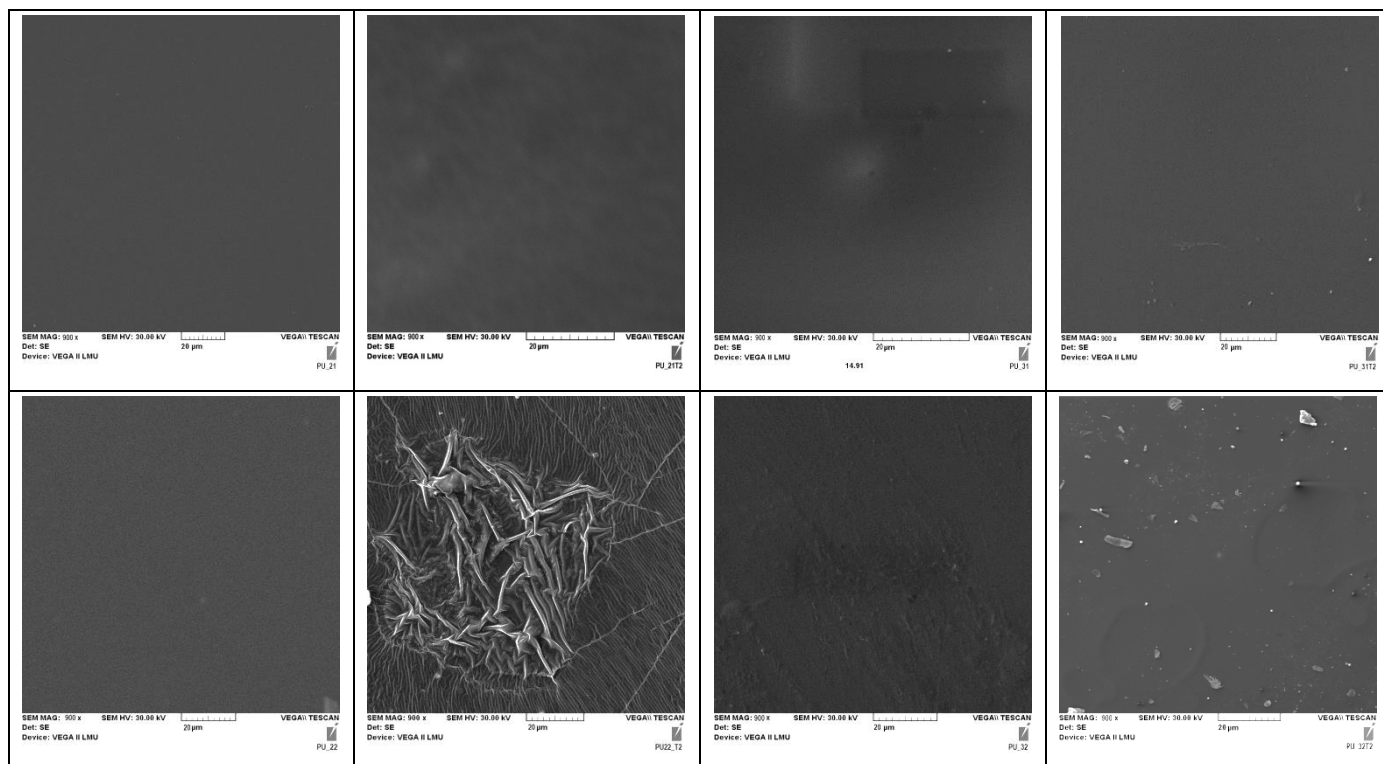


**Figure S5.** – FT-IR spectra of polyurethanes based on RP2.

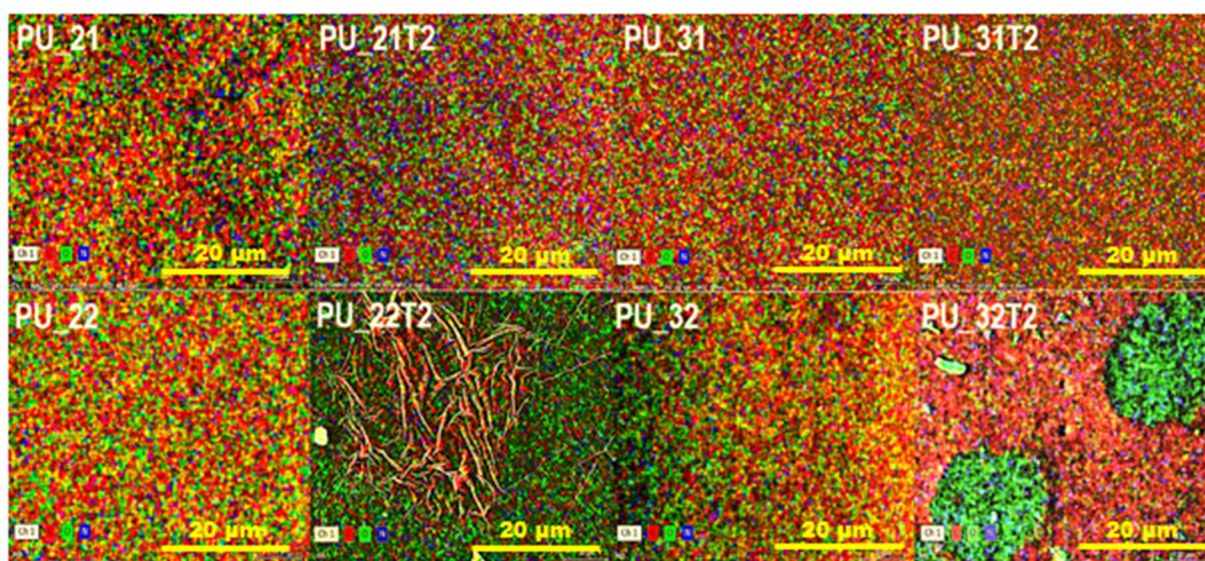


								
Flexible, bubble-free film, compact structure T <sub>c</sub> =72 h	Flexible film, few bubbles, compact structure T <sub>c</sub> =44 h	Not so flexible, few bubbles, compact structure T <sub>c</sub> =18 h	Flexible, bubble-free film, compact structure T <sub>c</sub> =94 h	Flexible, bubble-free film, compact structure T <sub>c</sub> =60 h	Flexible, bubble-free film, compact structure T <sub>c</sub> =24 h	Flexible, bubble-free film, compact structure T <sub>c</sub> =128 h	Flexible, bubble-free film, compact structure T <sub>c</sub> =102 h	Flexible, bubble-free film, compact structure T <sub>c</sub> =50 h
								
Sticky, gelatinous, bubble-free film T <sub>c</sub> =86 h	Flexible film, few bubbles,, compact structure T <sub>c</sub> =52 h	Flexible film, few bubbles,, compact structure T <sub>c</sub> =32 h	Incomplete reaction T <sub>c</sub> =n/a h	Flexible, bubble-free film, compact structure T <sub>c</sub> =102 h	Flexible, bubble-free film, compact structure T <sub>c</sub> =58 h	Incomplete reaction T <sub>c</sub> =n/a h	Flexible, bubble-free film, compact structure T <sub>c</sub> =132 h	Flexible, bubble-free film, compact structure T <sub>c</sub> =96 h
								
Sticky, gelatinous, bubble-free film T <sub>c</sub> =92 h	Flexible film, few bubbles, compact structure T <sub>c</sub> =62 h	Flexible film, few bubbles, compact structure T <sub>c</sub> =38 h	Incomplete reaction T <sub>c</sub> =n/a h	Flexible, bubble-free film, compact structure T <sub>c</sub> =102 h	Flexible, bubble-free film, compact structure T <sub>c</sub> =64 h	Incomplete reaction T <sub>c</sub> =n/a h	Flexible, bubble-free film, irregular surface T <sub>c</sub> =142 h	Flexible, bubble-free film, irregular surface T <sub>c</sub> =102 h

**Figure S6.** – Brief table comprising the aspect, properties, and the time of curing for each polyurethane binder.



**Figure S7.** – SEM images of polyurethanes formulations based on RP1 and RP2 (with and without TEGDN).



**Figure S8.** EDX mapping of the polyurethane formulations from S7.

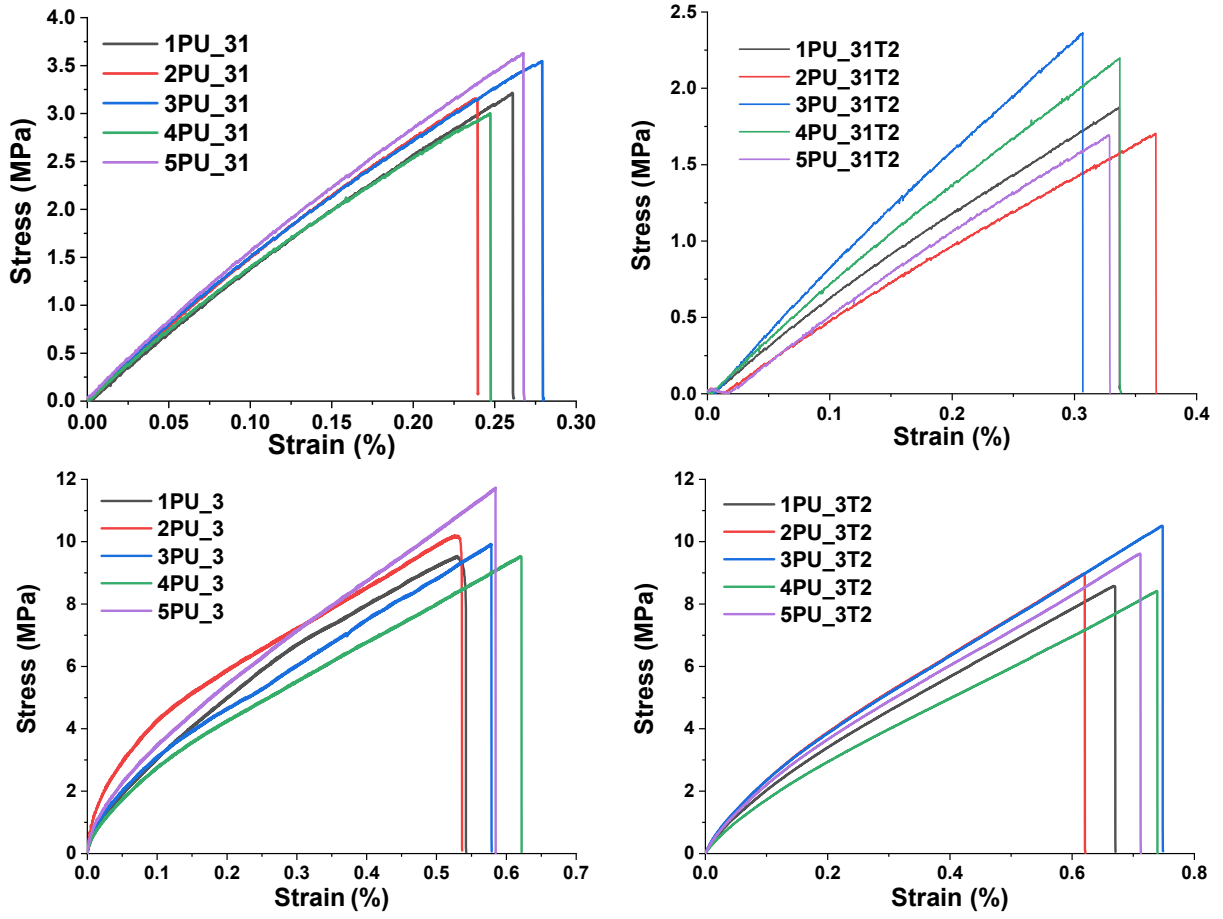
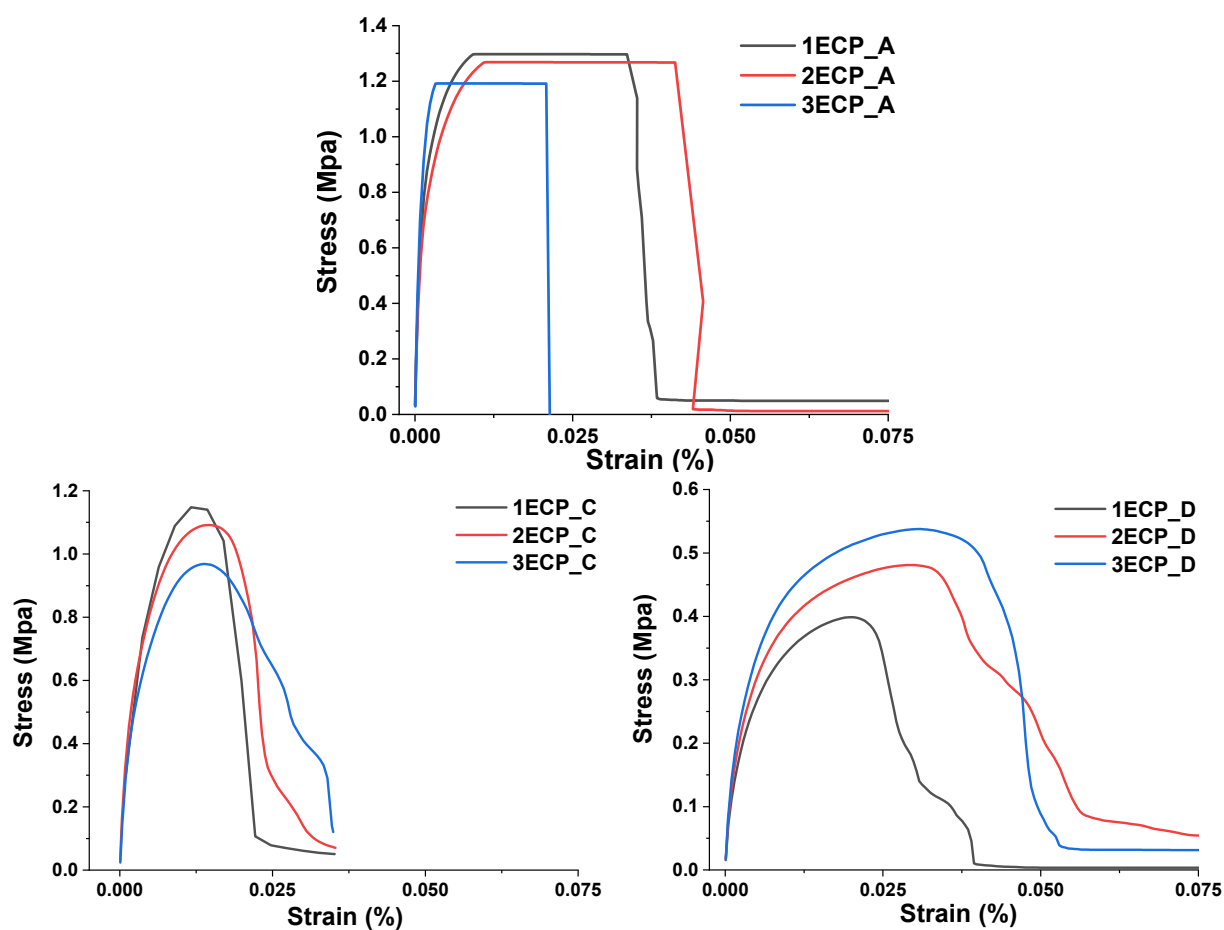


Figure S9. – Tensile stress-strain plots for PU formulations.



**Figure S10.** – Tensile stress-strain plots for composite propellant formulations.

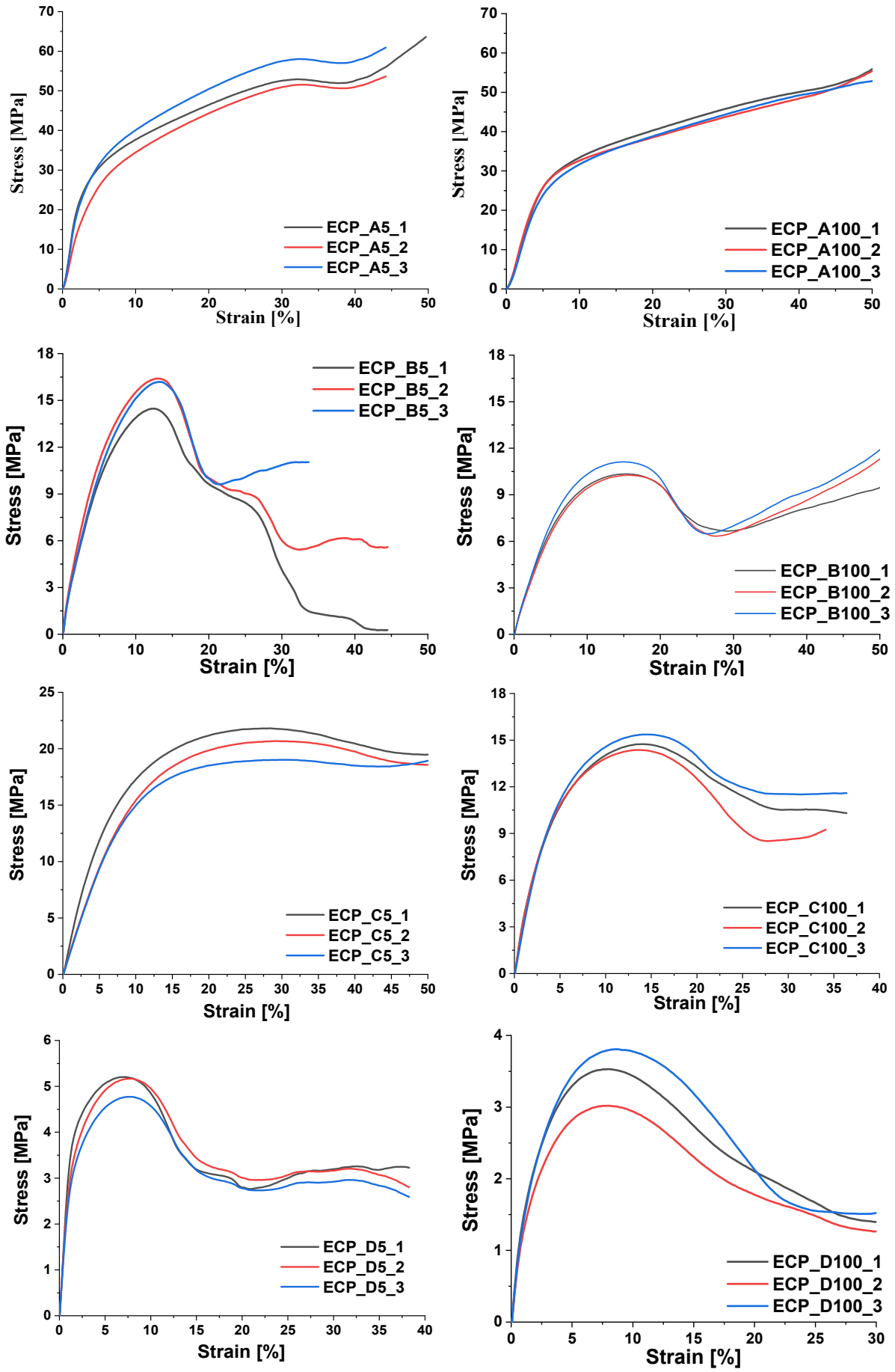
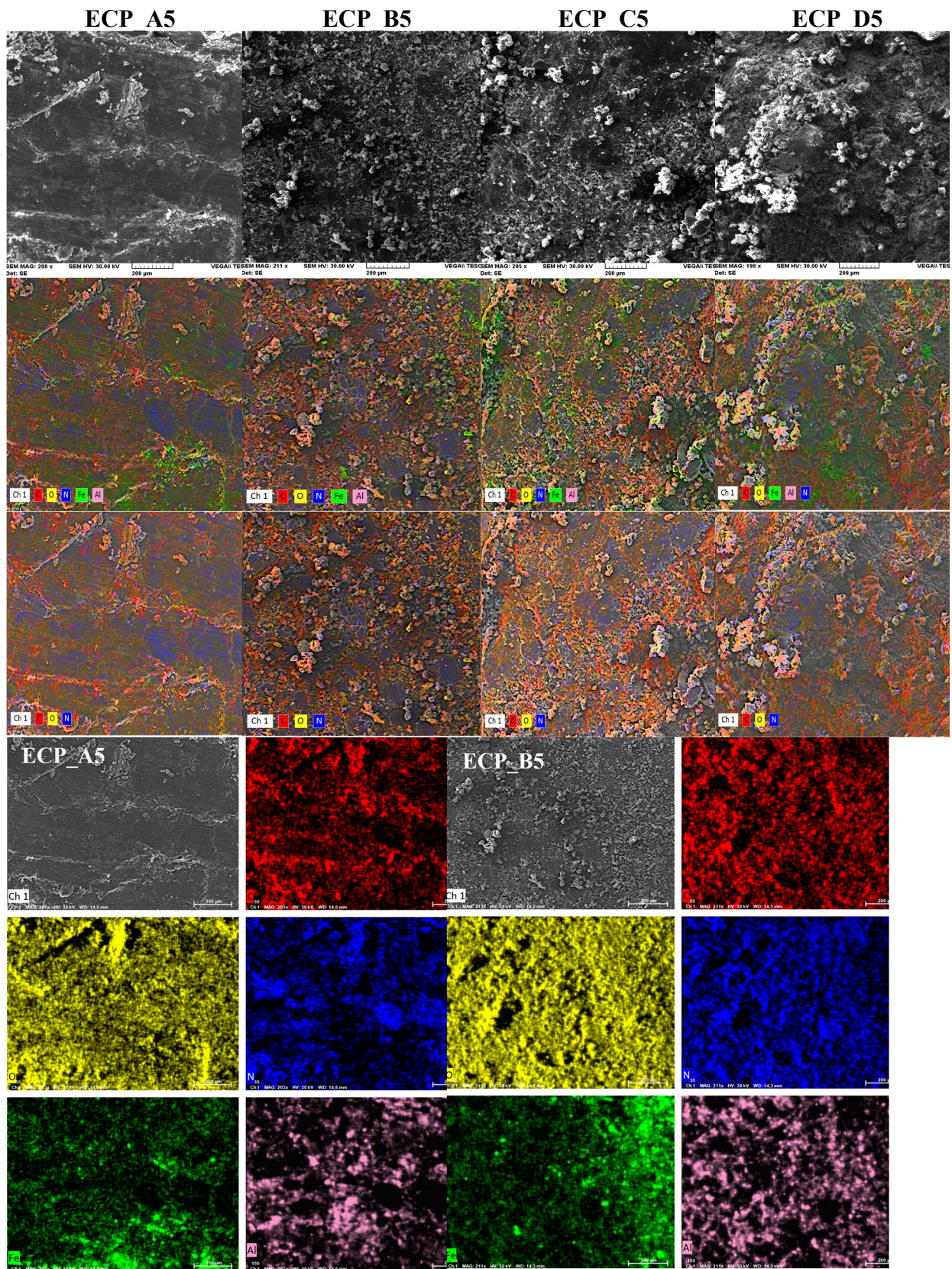
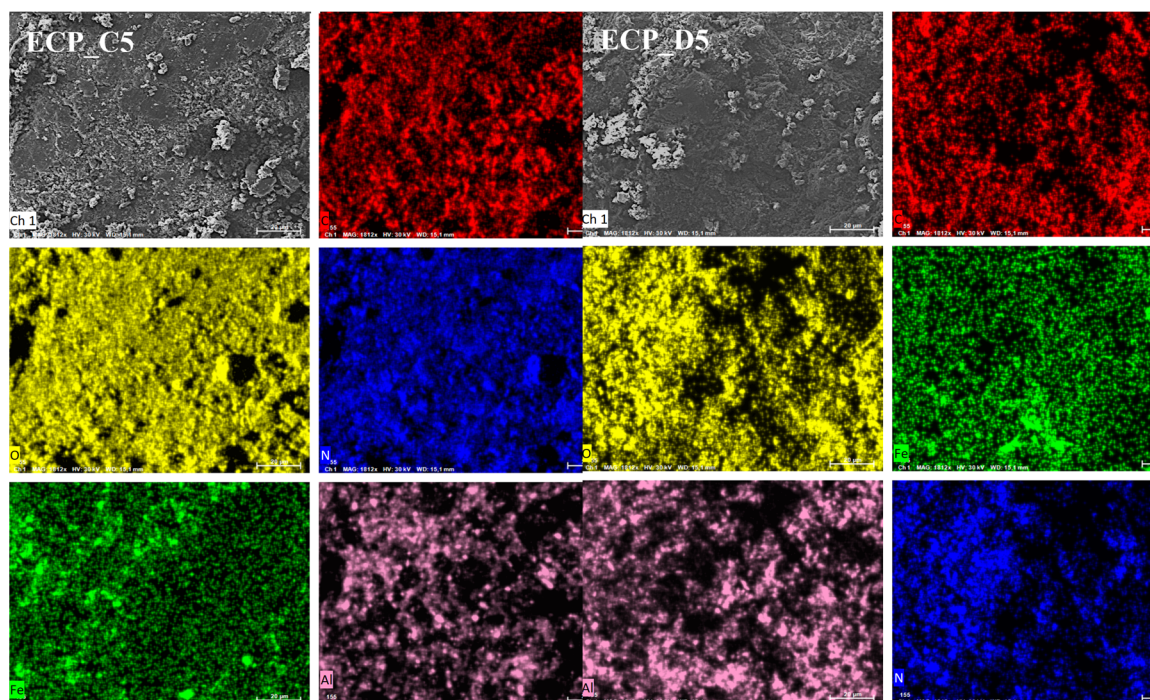


Figure S11. – Compression tests plots for composite formulations.

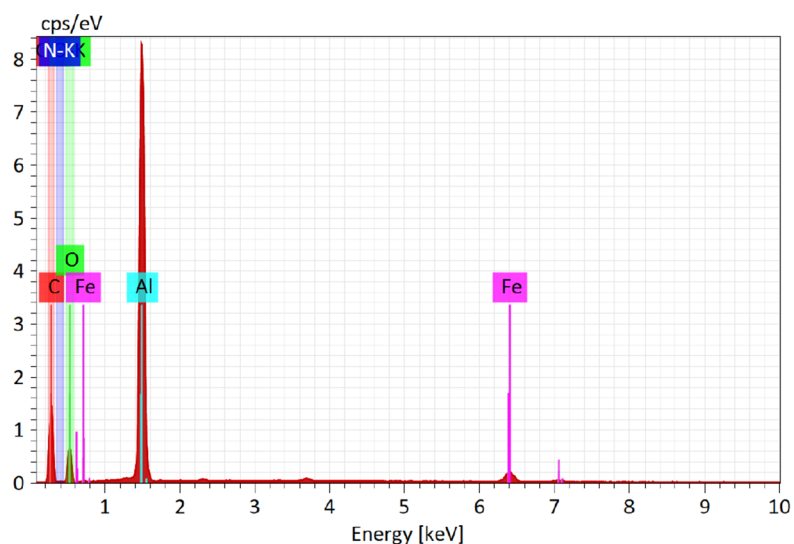








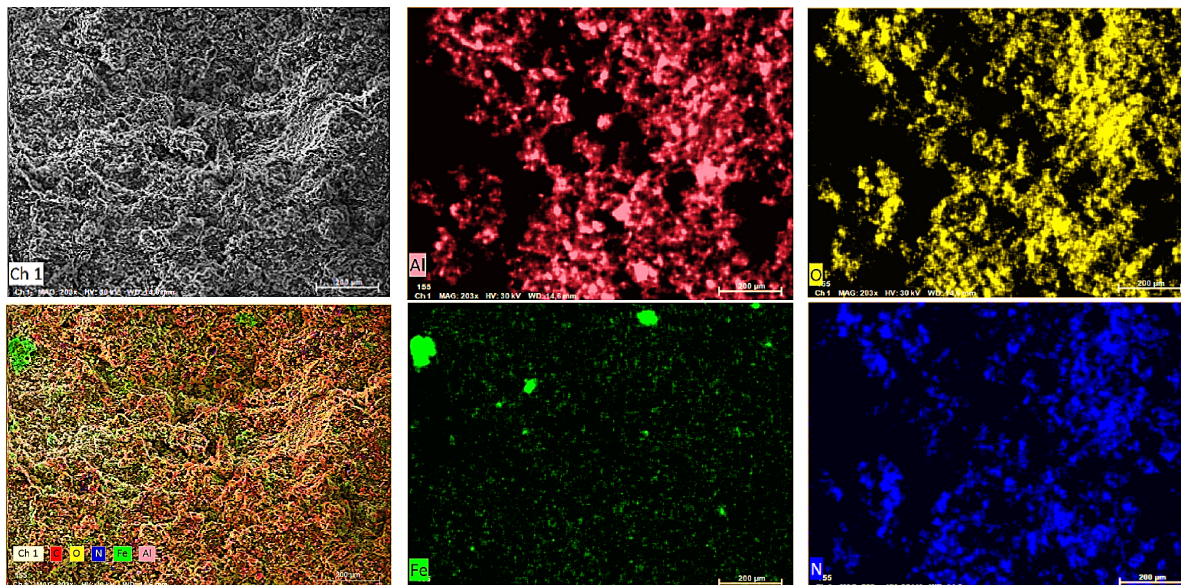
**Figure S12.** – SEM-EDX images of composite propellant formulations.



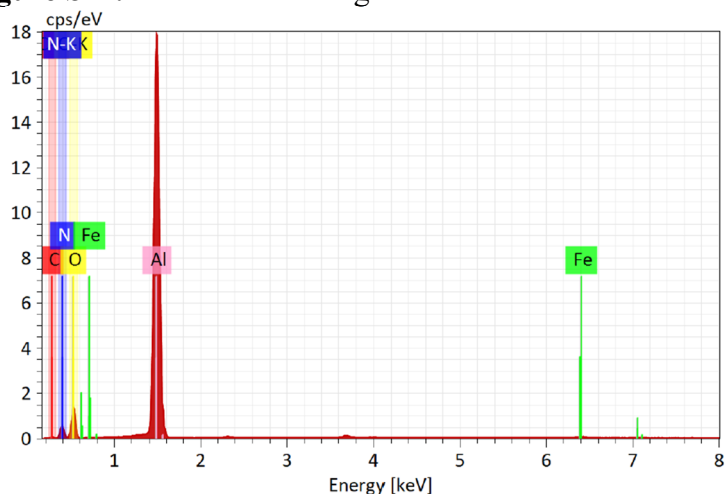
**Figure S13.** – EDX spectra for oxidizer-free composite formulations .

**Table S1.** Weight and atomic composition of oxidizer-free composite formulations.

Element	At. No.	Netto	Mass [%]	Mass Norm. [%]	Atom [%]	abs. error [%] (1 sigma)	rel. error [%] (1 sigma)
Carbon	6	52579	119,49	54,32	65,62	6,53	5,47
Oxygen	8	25392	47,09	21,41	19,42	2,59	5,50
Aluminium	13	426660	41,21	18,74	10,08	1,74	4,21
Nitrogen	7	1755	9,77	4,44	4,60	0,59	6,00
Iron	26	20749	2,39	1,09	0,28	0,05	2,18
		<b>Sum</b>	<b>219,96</b>	<b>100,00</b>	<b>100,00</b>		



**Figure S14.** – SEM-EDX images of binder-free solid mixture.

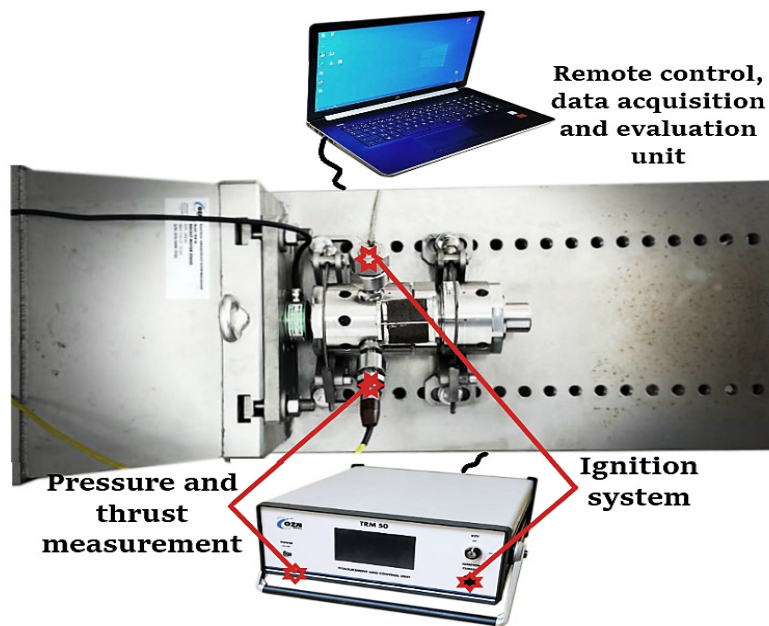


**Figure S15** – EDX spectra for binder-free solid mixture.

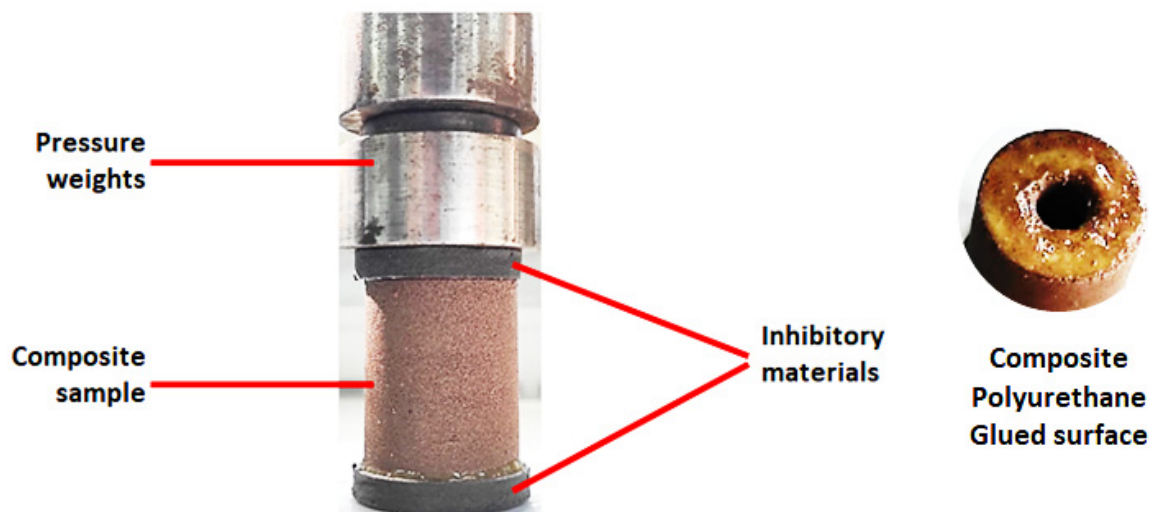
**Table S2.** Weight and atomic composition of free binder solid mixture.

Element	At. No.	Netto	Mass [%]	Mass Norm. [%]	Atom [%]	abs. error [%] (1 sigma)	rel. error [%] (1 sigma)
Aluminium	13	917245	58,74	40,03	28,82	2,30	3,91
Oxygen	8	52068	56,50	38,58	43,88	2,57	5,42
Nitrogen	7	21162	30,96	21,10	27,21	1,69	5,47
Carbon	6	0	0,00	0,00	0,00	0,00	4,36
Iron	26	5482	0,54	0,29	0,09	0,01	3,09
		<b>Sum</b>	<b>146,75</b>	<b>100,00</b>	<b>100,00</b>		

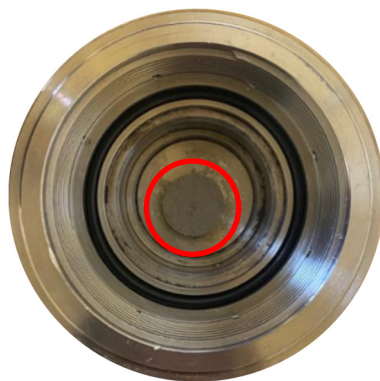




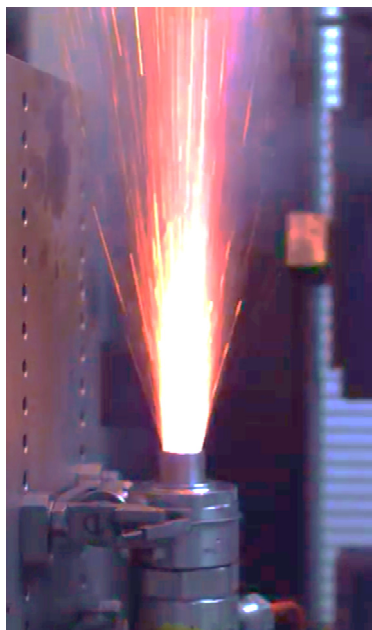
**Figure S16.** – Stand burner equipped with Subscale Rocket Motor TRM-35.



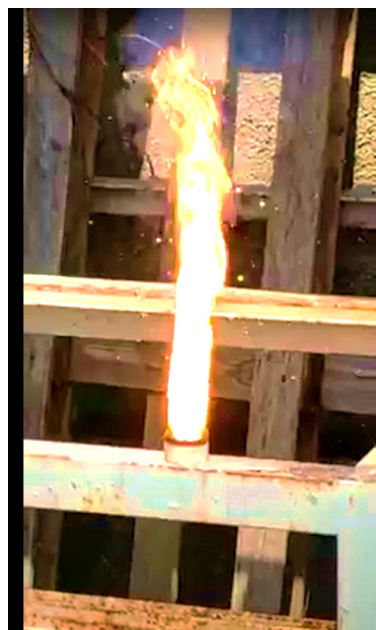
**Figure S17.** – Structural configuration of ECP\_D5.



**Figure S18.** – Pyrotechnic composition for propellant ignition.

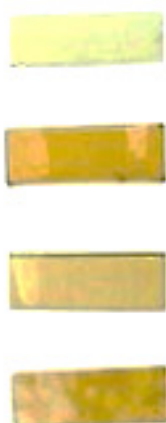


**a.**

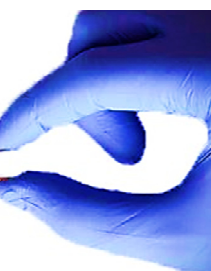


**b.**

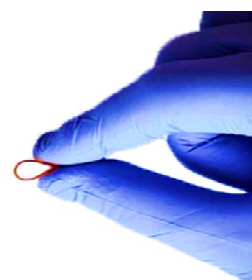
**Figure S19.** – Flame configuration of ECP\_D5 during the combustion (a. at nozzle exit; b. outdoor).



PU\_3



PU\_31

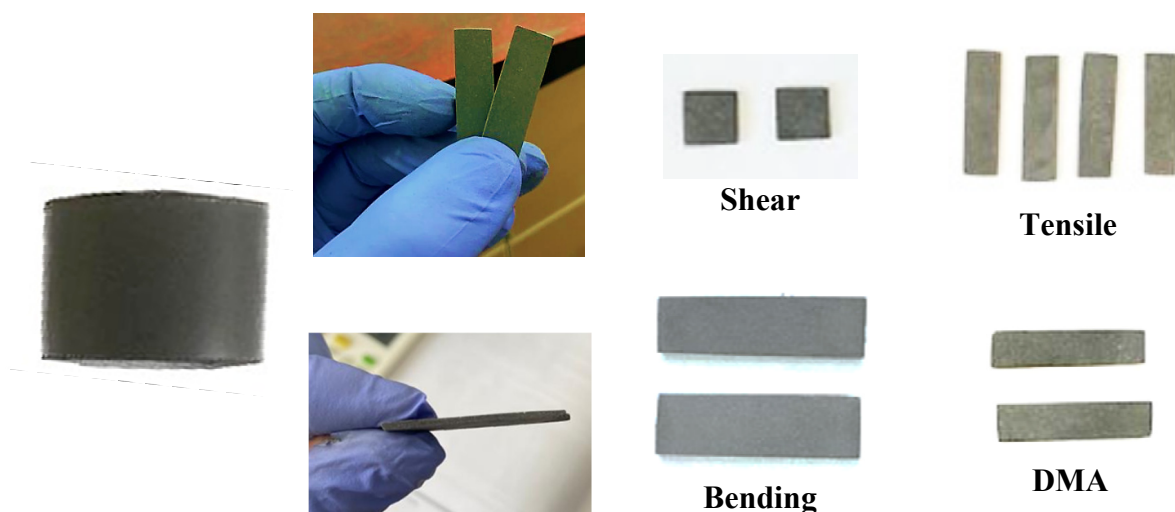


PU\_3T2



PU\_31T2

**Figure S20.** – Behavior of PUs during manual bending.



**Figure S21.** – Structural configurations of composite propellants for mechanical analysis.

**Table S3.** Thermal characteristics for polyurethane films, composite propellant formulations, and energetic plasticizer.

Sample	Evaluation results					
	Beginning temperature	Onset	Top	Max. Difference	End Temperature	Peak Type
PU 2	208.6	231.7	263.7	0.6	283.8	Exotherm
TEGDN	149.4	191.8	193.7	44.6	225.7	Exotherm
PU 2T1	161.5	179.2	200.8	3.9	228.7	Exotherm
PU 2T2	154.0	185.2	196.6	7.8	228.3	Exotherm
PU 21T1	148.3	182.3	199.4	6.2	200.2	Exotherm
PU 21T2	142.2	193.7	194.9	40.3	207.7	Exotherm
PU 3	208.8	239.5	278.7	0.7	289.7	Exotherm
PU 3T1	152.4	179.8	198.6	4.7	233.6	Exotherm
PU 3T2	143.1	185.6	196.2	14.4	227.9	Exotherm
PU 31T1	159.1	180.7	197.4	6.5	229.9	Exotherm
PU 31T2	150.9	190.3	191.2	52.4	202.5	Exotherm
ECP A5	160.5	164.1	54.1	2.5	299.9	Exotherm
ECP B5	155.8	165.3	183.7	32.8	195.8	Exotherm
ECP D5	159.5	176.5	183.8	75.1	194.3	Exotherm

**Table S4.** Thermal properties of synthesized polyurethanes.

Sample	T <sub>10%</sub> <sup>1</sup> [°C]	T <sub>max1</sub> <sup>2</sup> [°C]	T <sub>max2</sub> <sup>2</sup> [°C]	T <sub>max3</sub> <sup>2</sup> [°C]	T <sub>max4</sub> <sup>2</sup> [°C]
PU 3	306.1	313.6	371.1	456.1	-
PU 31	313.5	313.5	410.9	-	-
PU 3T2	197.6	190.1	315.1	370.1	457.6
PU 31T2	181.4	193.9	326.4	408.9	-

<sup>1</sup>decomposition onset temperature (measured at 10 % weight loss).

<sup>2</sup>the maximum decomposition temperatures, corresponding to the maximum of DTG peaks.

**Table S5.** The decomposition process of the polyurethanes (weight loss versus temperature).

Sample	Weight loss [%]								
	200°C	250°C	300°C	350°C	400°C	450°C	500°C	550°C	600°C
<b>PU 3</b>	0.5	0.8	7.1	32.1	60.3	70.3	92.9	94.2	95.1
<b>PU 3I</b>	1.1	1.4	6.4	19.4	43.4	77.4	85.9	87.8	88.6
<b>PU 3T2</b>	10.1	13.8	20.2	37.9	55.9	73.1	91.9	94.3	95.2
<b>PU 3IT2</b>	18.4	25.1	29.5	39.7	58.8	81.7	87.5	88.9	89.7

**Table S6.** Heat of combustion, specific volume and  $T_g$  for our new composite propellants in comparison with the existing commercial formulations.

Sample	Qv [cal/g]	Vsp [l/kg]	$T_g$
<b>HTPB composite propellant</b>	1700	507	-72°C
<b>Double based propellants</b>	1020	798	-35.5°C
<b>ECP D5</b>	1002	693	-32°C
<b>ECP B5</b>	1010	658	-61°C