Supporting Information for:

Scalable Solution for High Dispersibility and Low Heterogeneity of High Inorganic Fillers / Polymer Composites: Vitrimer via Reactive Extrusion & Mechanochemical Intercalation

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Various experiments

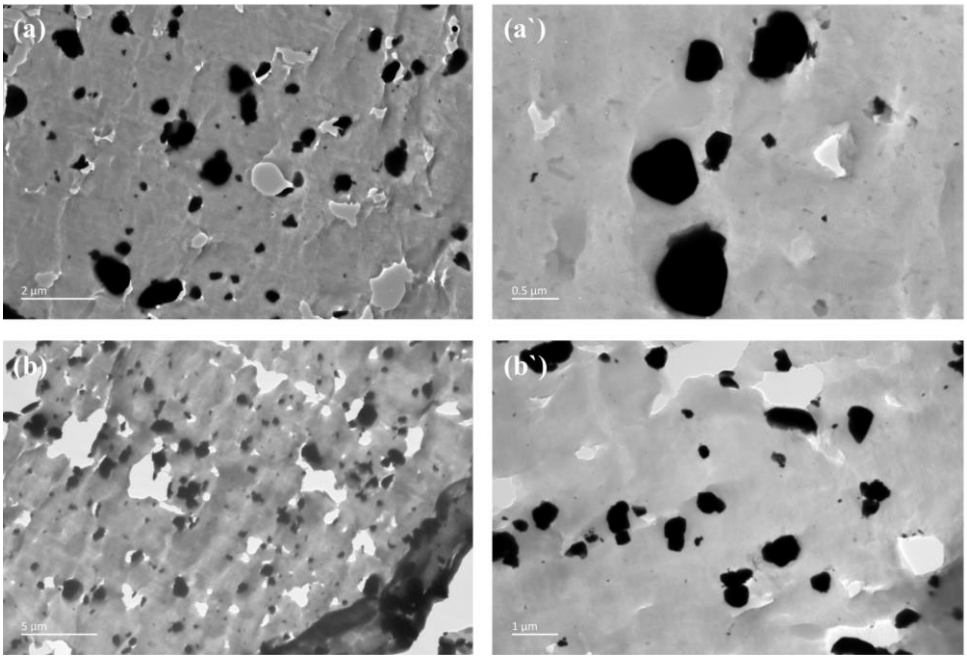

Fig. S1 Single-particle dispersion of KH560-modified tungsten powder in an HDPE-v matrix: TEM graphs (a-a`) W/HDPE-v-0.5%-100rpm-6MPa-200rpm-20min; (b-b`) W/HDPE-v-0.5%-100rpm-6MPa-200rpm-6h.

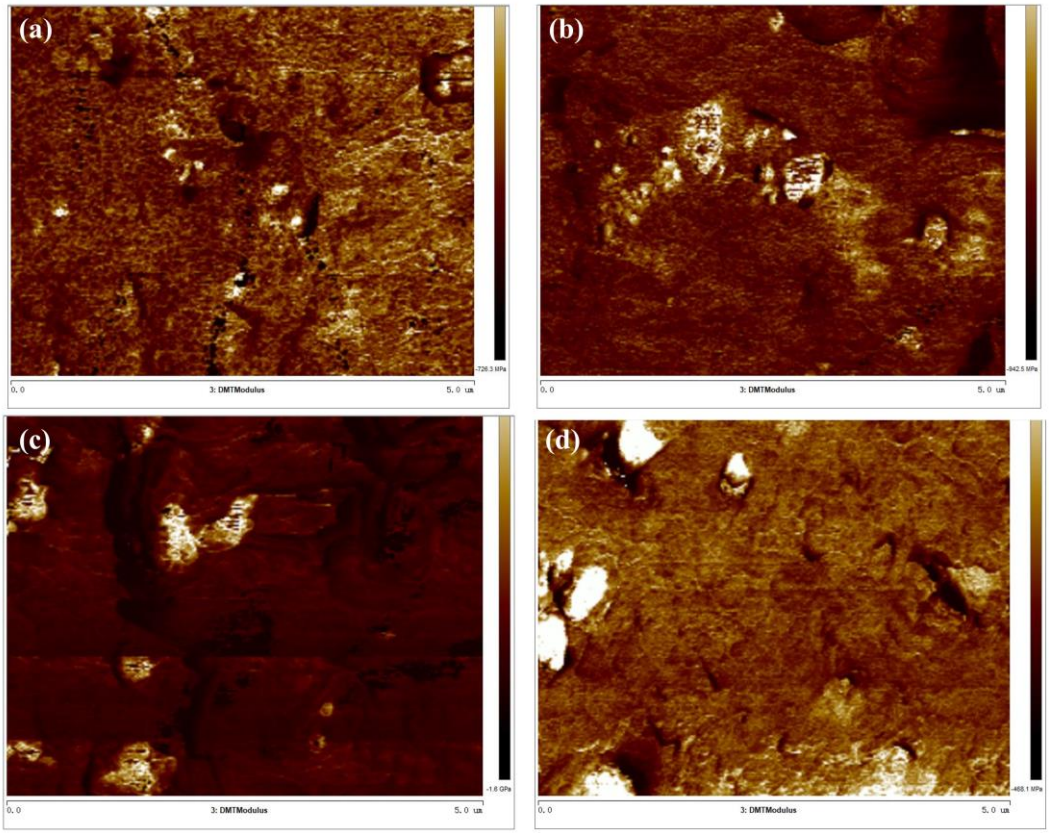

Fig. S2 The modulus of AFM graphs of W/HDPE-v-0.5%-100rpm-6MPa-200rpm-(20min-6h): (a)

20min; (b) 1h; (c) 2h; (d) 6h.

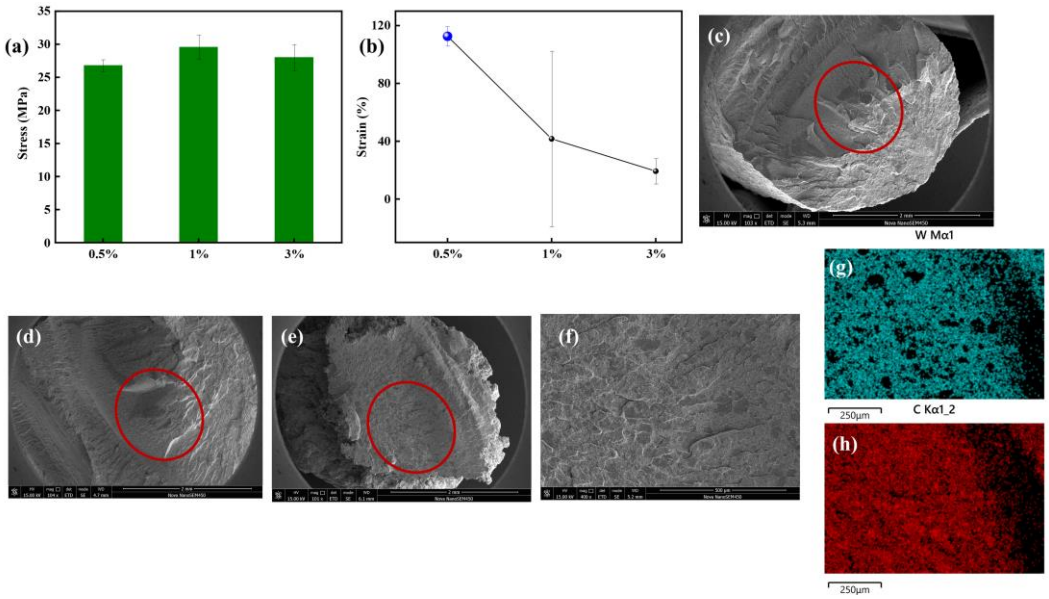

Fig. S3 At a low twin-screw rotational speed (30 rpm) and a low degree of S^3M treatment, the influence of different contents of DGEBA crosslinking agent on W/HDPE-v: (a-b) The mechanical properties of W/HDPE-v-(0.5%, 1%, 3%)-50rpm-3MPa-30rpm-20min; the cross-section morphology of W/HDPE-v-50rpm-3MPa-30rpm twin-screw extrusion particle (c) 0.5%;, (d) 1%, (e) 3%; (f) the magnified graph of 3% particle; EDS mapping of 3% particle`s cross-section (g) W element, (h) C element.

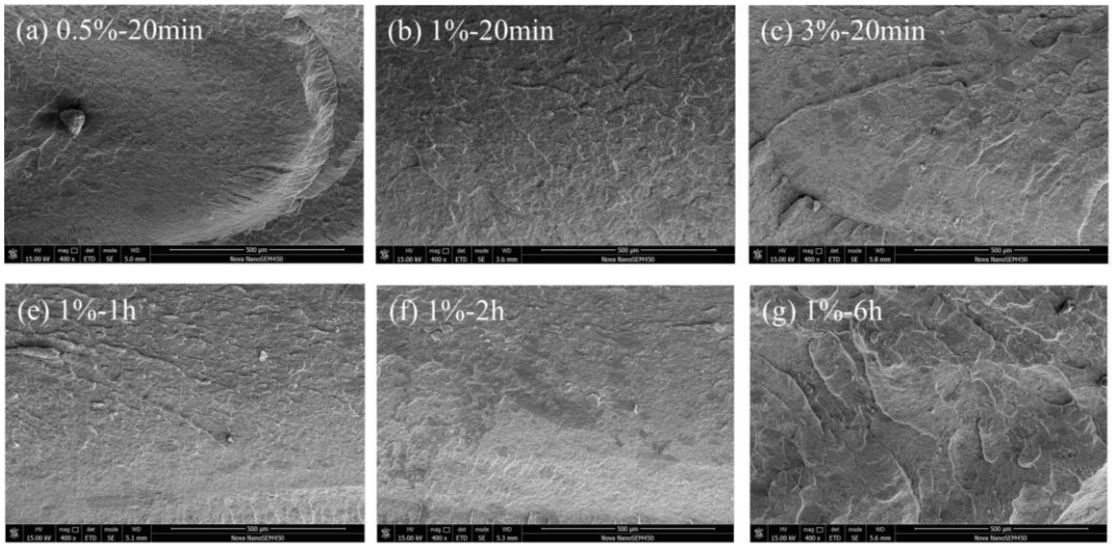

Fig. S4 The macroscopic cross-section morphology of W/HDPE-v-(0.5% (a), 1% (b); (3%) (c))-50rpm-3MPa-30rpm-20min; the macroscopic cross-section morphology of W/DPE-v-1%-50rpm-3MPa-(1h (e), 2h (f), 6h (g)).

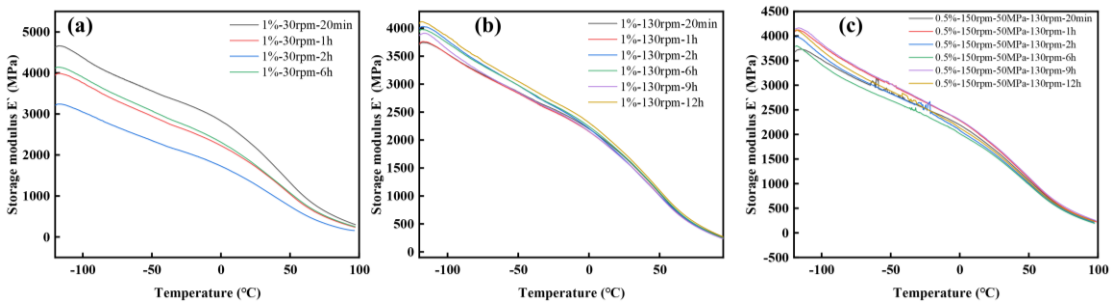

Fig. S5 DMA graphs: the storage modulus of W/HDPE-v-1%-50rpm-3MPa-30rpm-(20min-6h) (a); W/HDPE-v-1%-50rpm-3MPa-130rpm-(20min-12h) (b); and W/HDPE-v-0.5%-150rpm-5MPa-130rpm-(20min-12h) (c).

Table. S1 The DSC data of each sample

	Enthalpy of fusion (J/g)	Crystallinity (%)	Melting point (℃)	Initial melting point(℃)	Crystallization temperature(℃)	Initial crystallization temperature(℃)	Enthalpy of fusion of MA peak (J/g)
HDPE-g-MA	159.7	55.07	130.61	122.41	114.68	117.42	4.91
0.5%-extrusion-50rpm-3MPa-30rpm	47.1	54.14	129.81	121.54	115.48	118.69	1.264
0.5%-20min-50rpm-3MPa-30rpm	46.77	53.76	129.11	121.06	115.7	118.63	1.317
0.5%-1h-50rpm-3MPa-30rpm	49.12	56.45	128.67	121.03	115.49	118.41	1.395
0.5%-2h-50rpm-3MPa-30rpm	47.96	55.12	129.56	121.06	115.36	118.37	1.343

0.5%-6h-50rpm-3MPa-30rpm	48.17	55.36	128.71	121.29	115.35	118.12	1.344
1%-extrusion-50rpm-3MPa-30rpm	49.86	57.31	130.56	121.44	114.56	118.48	1.36
1%-20min-50rpm-3MPa-30rpm	46.48	53.42	129.38	121.59	115.62	118.7	1.259
1%-1h-50rpm-3MPa-30rpm	48.03	55.21	129.42	121.36	114.38	118.52	1.286
1%-2h-50rpm-3MPa-30rpm	47.53	54.63	129.28	121.35	114.9	118.37	1.31
1%-6h-50rpm-3MPa-30rpm	49.24	56.59	128.84	120.91	114.89	118.27	1.355
3%-extrusion-50rpm-3MPa-30rpm	44.5	51.15	130.59	120.41	113.25	118.47	1.252
3%-20min-50rpm-3MPa-30rpm	44.16	50.75	129.28	122.64	114.48	118.9	1.212

3%-1h-50rpm-3MPa-30rpm	45.75	52.58	128.57	119.79	114.02	118.8	1.295
3%-2h-50rpm-3MPa-30rpm	45.66	52.48	128.75	120.34	114.95	119.03	1.477
1%-extrusion-50rpm-3MPa-80rpm	52.23	60.03	129.64	121.22	115.3	118.93	1.38
1%-20min-50rpm-3MPa-80rpm	47.11	54.14	128.30	120.34	116.03	119.01	1.369
1%-1h-50rpm-3MPa-80rpm	46.15	53.04	128.77	120.78	115.33	118.6	1.354
1%-2h-50rpm-3MPa-80rpm	45.13	51.87	128.67	120.71	115.58	118.34	1.338
1%-6h-50rpm-3MPa-80rpm	46.17	53.06	128.39	120.75	115.89	118.37	1.385
1%-extrusion-50rpm-3MPa-130rpm	52.12	59.90	130.31	121.71	116.09	118.93	1.384

1%-20min-50rpm-3MPa-130rpm	49.93	57.39	128.35	120.3	115.43	118.82	1.372
1%-1h-50rpm-3MPa-130rpm	49.58	56.98	127.98	120.55	115.09	118.51	1.459
1%-2h-50rpm-3MPa-130rpm	46.44	53.37	128.48	120.31	115.52	118.56	1.351
1%-6h-50rpm-3MPa-130rpm	47.67	54.79	129.48	121.29	114.39	117.98	1.378
1%-9h-50rpm-3MPa-130rpm	57.14	65.67	127.60	120.34	116.26	118.62	1.725
1%-12h-50rpm-3MPa-130rpm	53.42	61.40	129.83	121.26	114.87	118.26	1.615
0.5%-extrusion-150rpm-5MPa-130rpm	55.35	63.62	128.92	121.55	117.5	119.02	1.447
0.5%-20min-150rpm-5MPa-130rpm	52.98	60.89	128.74	120.47	116.25	118.96	1.556

0.5%-1h-150rpm-5MPa-130rpm	51.13	58.77	127.58	121.16	117.2	118.95	1.473
0.5%-2h-150rpm-5MPa-130rpm	50.81	58.40	127.04	120.48	117.17	118.78	1.45
0.5%-6h-150rpm-5MPa-130rpm	51.54	59.24	128.69	121.13	115.88	118.41	1.541
0.5%-9h-150rpm-5MPa-130rpm	52.69	60.56	127.53	121.1	117.18	118.51	1.471
0.5%-12h-150rpm-5MPa-130rpm	53.39	61.36	127.34	120.73	116.47	118.33	1.532

Table. S2 The optimal process parameters

Processing name	Processing parameters
Reactive twin-screw extrusion processing section	Extruder parameters:180℃, 185℃ 190℃, 190℃, 190℃,185℃； 30rpm； (Zn(acac)2): 1 wt%; DGEBA: 0.5wt%, HDPE-g-MA: 98.5 wt%
S^3M processing section	70 wt% W, 30 wt% HDPE-v; 5MPa, 150rpm; 10 milling cycles
Twin-screw extrusion processing section	Extruder parameters: 180℃, 185℃, 190℃, 190℃, 190℃,185℃,130rpm
Hot-pressing section	200°C, 10 MPa, 2h