Enhancing thermoelectric efficiency of GeTe through process improvement assisted by active learning and Bayesian optimization

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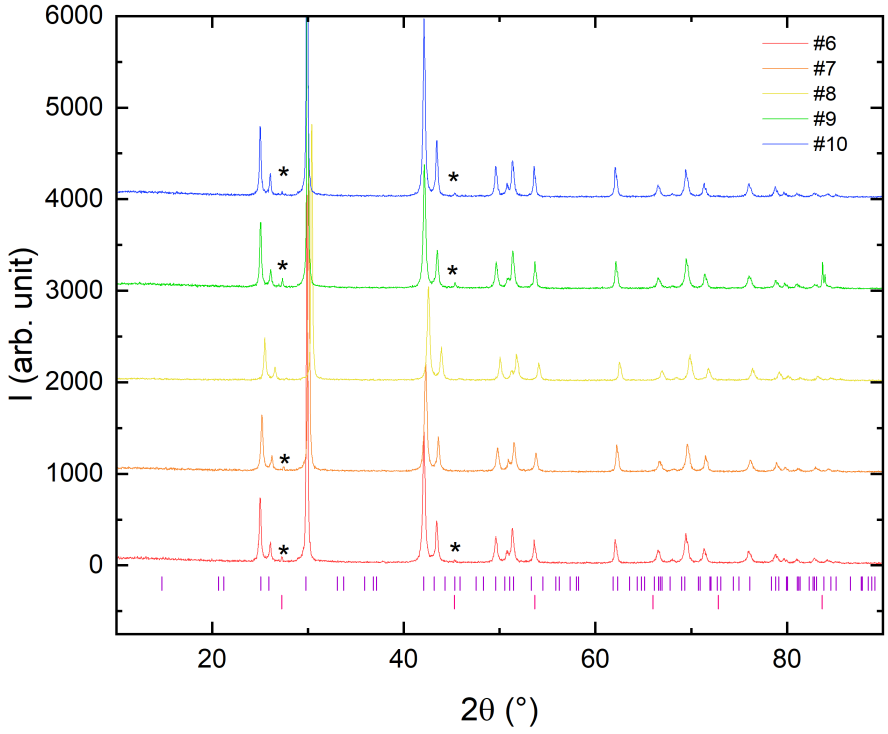
Supplementary information

Table S1: Process parameters of the samples.

Sample#	**heating.rate** ($K.h^{-1}$)	**T_annealing** (K)	**t_duration** (h)	**cooling.rate** ($K.h^{-1}$)	**mass_powder** (g)	**milling.time.1** (min)	**heating.rate.1** ($K.min^{-1}$)	**T_annealing.1** (K)	**t_duration.1** (min)	**cooling.rate.1** ($K.min^{-1}$)	**Uniaxial pressure** (MPa)
0	25	1123	16	175	4.25	1	25	850	40	150	70
1	50	1323	16	75	5.75	1	100	800	20	175	40
2	50	1273	12	175	4.5	1	75	900	55	50	60
3	50	1073	12	50	5.25	1	50	750	30	75	50
4	115.625	1223	12	154.16667	6.7	15	75.33	750	5	75.33	60
5	25	1273	24	25	5.1982	10	50	900	30	25	40
6	25	1323	14	75	5.8184	6	150	750	15	75	50
7	75	1423	18	25	5.9029	9	50	938	0	140	40
8	50	1473	22	75	5.1522	6	200	850	35	200	50
9	75	1423	24	50	5.2986	4	175	850	25	25	50
10	10.5	1348	24	75	4.52333	2	201	700	25	48	40
11	30	1348	24	75	5.5103	8	138	850	25	139	45
12	10.5	1448	0.1	75	4.52489	2	167	800	25	123	40
13	30	1123	16	25	6.05351	4	151	750	25	47	50
14	20	1348	24	50	5.75132	6	96	875	15	100	55
15	52.4	1348	18	25	4.61276	9	27.2	900	25	105.4	60
16	125	1373	6	100	4.10237	5	184	800	15	47.4	60
17	113.9	1273	22	25	5.19576	15	138	800	10	106.8	55
18	97.5	1223	6	77.3	3.74733	13	113	700	10	109	50
19	32	1273	22	25	4.12751	3	27.4	850	25	159	65
20	120	1098	22	75	5.252	3	125	750	5	175	45
21	10	1448	2	100	4.505	12	150	850	10	100	55
22	80	1398	22	100	5.752	3	125	900	0	50	50
23	130	1023	6	100	5.25	11	175	900	5	125	40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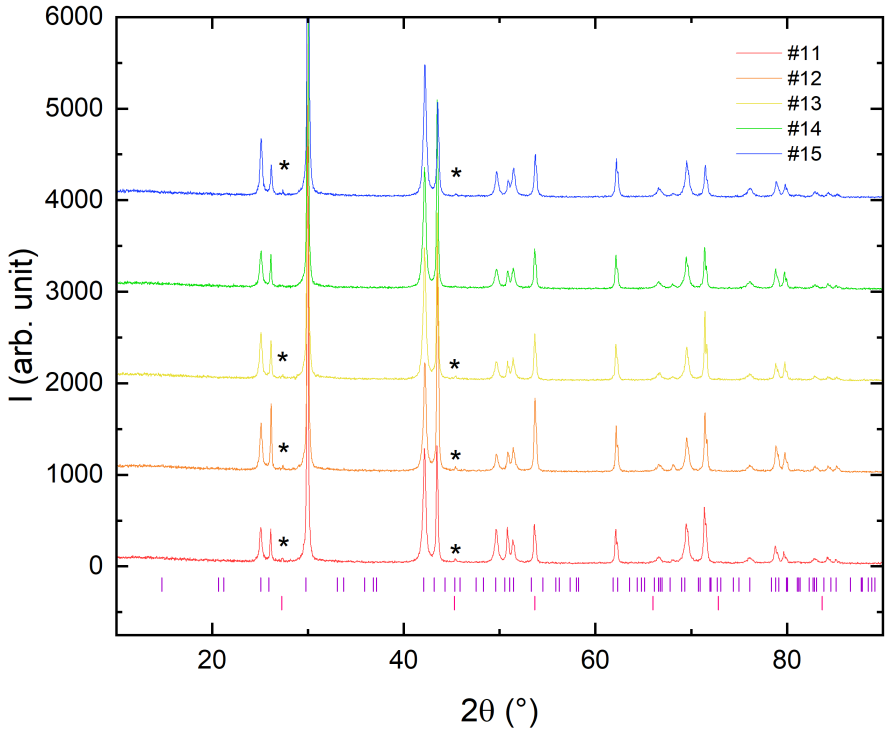
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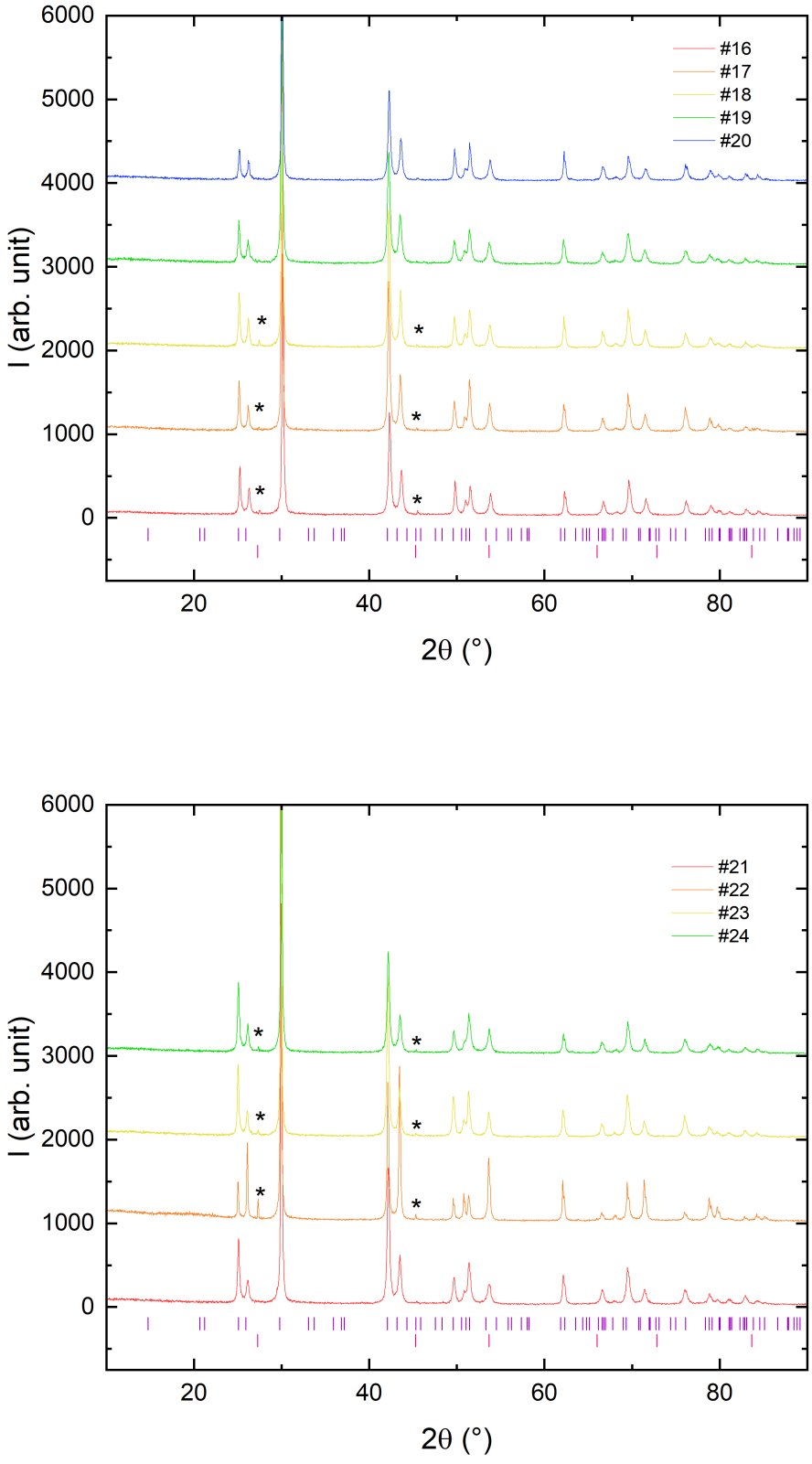

Figure S1: X-ray diffraction patterns associated with the Bragg peak positions of trigonal GeTe ($R3m$) and cubic Ge ($Fd\bar{3}m$). The * denotes the presence of elemental Ge.

Table S2: Le Bail refinement parameters extracted from XRD patterns.

Sample	**a (Å)**	**(a) (Å)**	**c (Å)**	**(c) (Å)**	$\langle d \rangle$ **(Å)**	$\langle (d) \rangle$ **(Å)**	**Chi^2**	**R_Bragg**	**R_F**
0	8.32898	7E-5	10.67244	1.3E-4	755.81	0.87	2.45	1.8	3.31
1	8.3309	1.6E-4	10.6736	2.7E-4	637.31	0.05	3.47	2.38	3.73
2	8.33097	7E-5	10.66495	1.8E-4	750.39	0.05	2.83	4.65	10.52
3	8.32922	9E-5	10.6701	2.1E-4	550.67	0.57	3.18	2.73	5.2
4	8.33326	6E-5	10.6763	2E-4	305.8	0.25	3.69	2.03	5.68
5	8.32907	1.2E-4	10.67576	1.6E-4	488.49	0.33	2.36	5.52	10.02
6	8.33442	1.8E-4	10.67958	2.6E-4	412.33	0.28	1.69	1.66	2.43
7	8.33812	1.3E-4	10.69258	1.8E-4	471.05	0.68	2.03	2.32	6.2
8	8.32745	1E-4	10.67811	1.6E-4	524.09	0.53	2.59	1.51	2.71
9	8.33116	8E-5	10.67306	1.5E-4	569.24	0.59	2.23	1.15	2.45
10	8.33295	9E-5	10.67242	2.2E-4	520.25	0.1	3.46	2.65	5.35
11	8.32997	8E-5	10.67417	1.9E-4	692.56	0.92	4.32	1.68	4.63
12	8.33044	9E-5	10.67576	2.1E-4	596.8	0.66	4.43	1.73	4.06
13	8.33358	9E-5	10.67232	2.2E-4	538.64	0.25	3.94	1.96	3.34
14	8.32923	1.1E-4	10.68117	2E-4	511.68	0.48	3.75	1.58	4.31
15	8.33184	1.1E-4	10.68418	1.8E-4	431.48	0.12	2.54	1.45	4.52
16	8.33128	1E-4	10.67865	1.6E-4	491.05	0.48	2.79	4.25	7.61
17	8.3315	1.1E-4	10.6801	1.8E-4	474.62	0.18	2.98	1.87	4.37
18	8.33429	1.4E-4	10.67578	2.4E-4	357.57	0.44	3.15	2.02	7.07
19	8.33074	1.2E-4	10.68204	1.8E-4	474.67	0.43	2.66	1.34	2.43
20	8.3337	1.3E-4	10.68167	1.9E-4	411.69	0.61	3.07	2.84	6.18
21	8.32752	6E-5	10.67397	1.4E-4	898.95	0.56	3.02	1.65	2.71
22	8.33156	9E-5	10.67583	1.6E-4	451.86	0.65	2.6	1.71	4.48
23	8.32592	1.3E-4	10.67917	2E-4	413.46	0.56	3.16	2.58	5.79

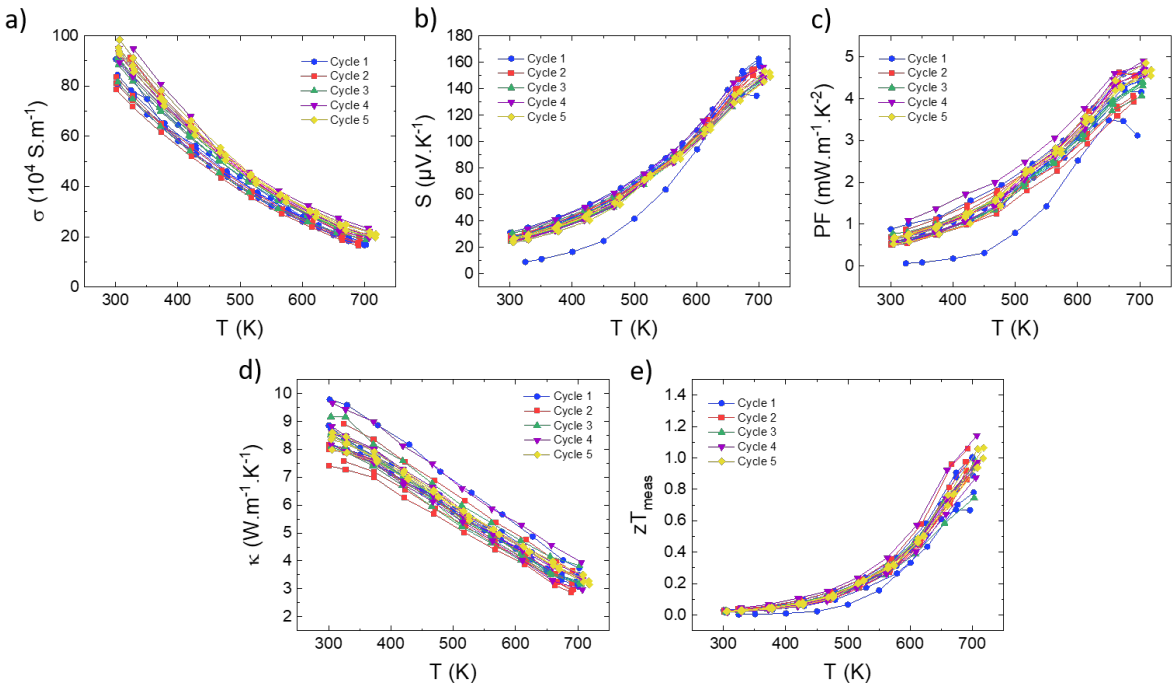

Figure S2: Thermoelectric properties of the samples as a function of temperature from cycle 1 to cycle 5. Electrical conductivity (a), Seebeck coefficient (b), power factor (c), thermal conductivity (d), and figure of merit (e).

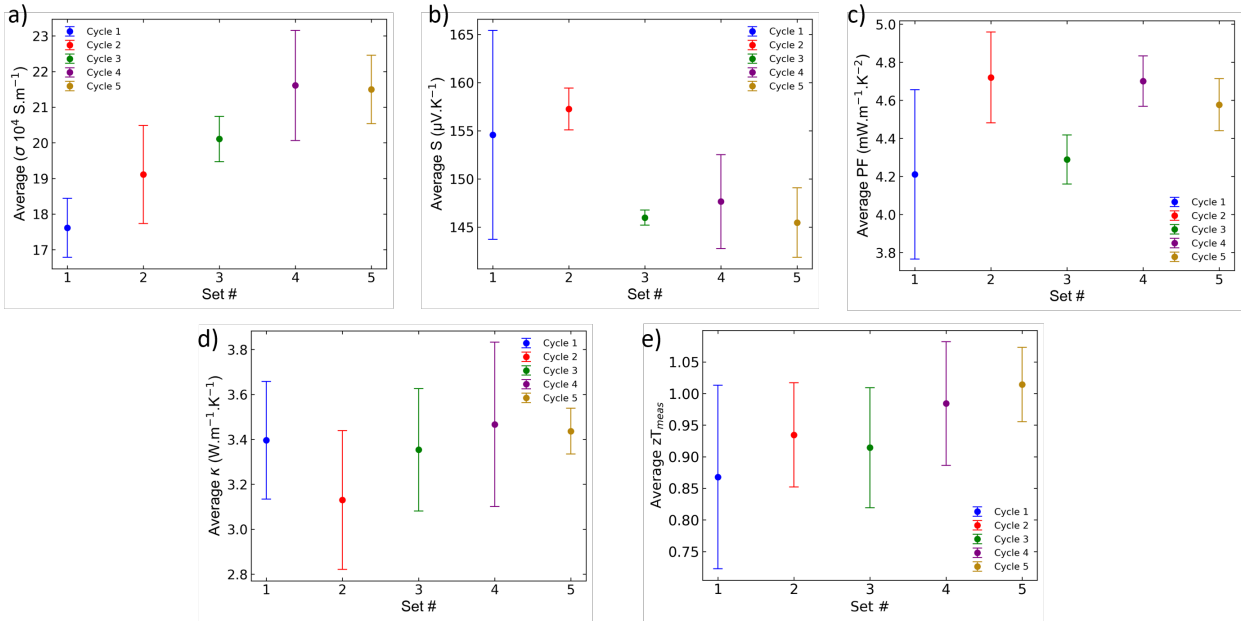

Figure S3: Average of the thermoelectric properties at 700 K as a function of sample number from cycle 1 to cycle 5. Electrical conductivity (a), Seebeck coefficient (b), power factor (c), thermal conductivity (d), and figure of merit (e).

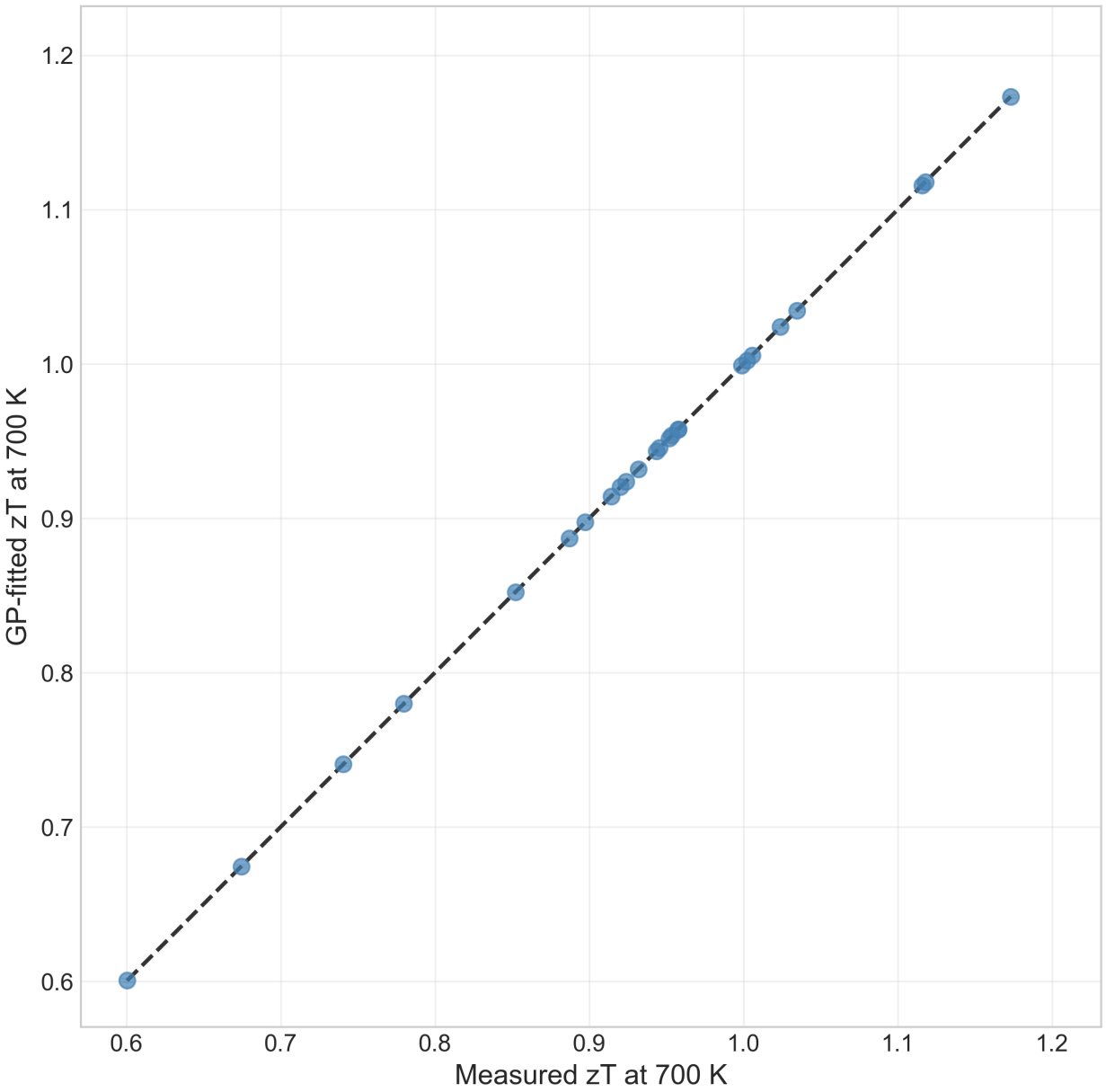

Figure S4: GP-fitted zT at 700 K as a function of measured zT at 700 K for all 25 GeTe samples. The perfect prediction line is shown in black. The error bars, large of 0.001 in average and therefore not visible, represent a single standard deviation of the GP-fitted zT at 700 K.

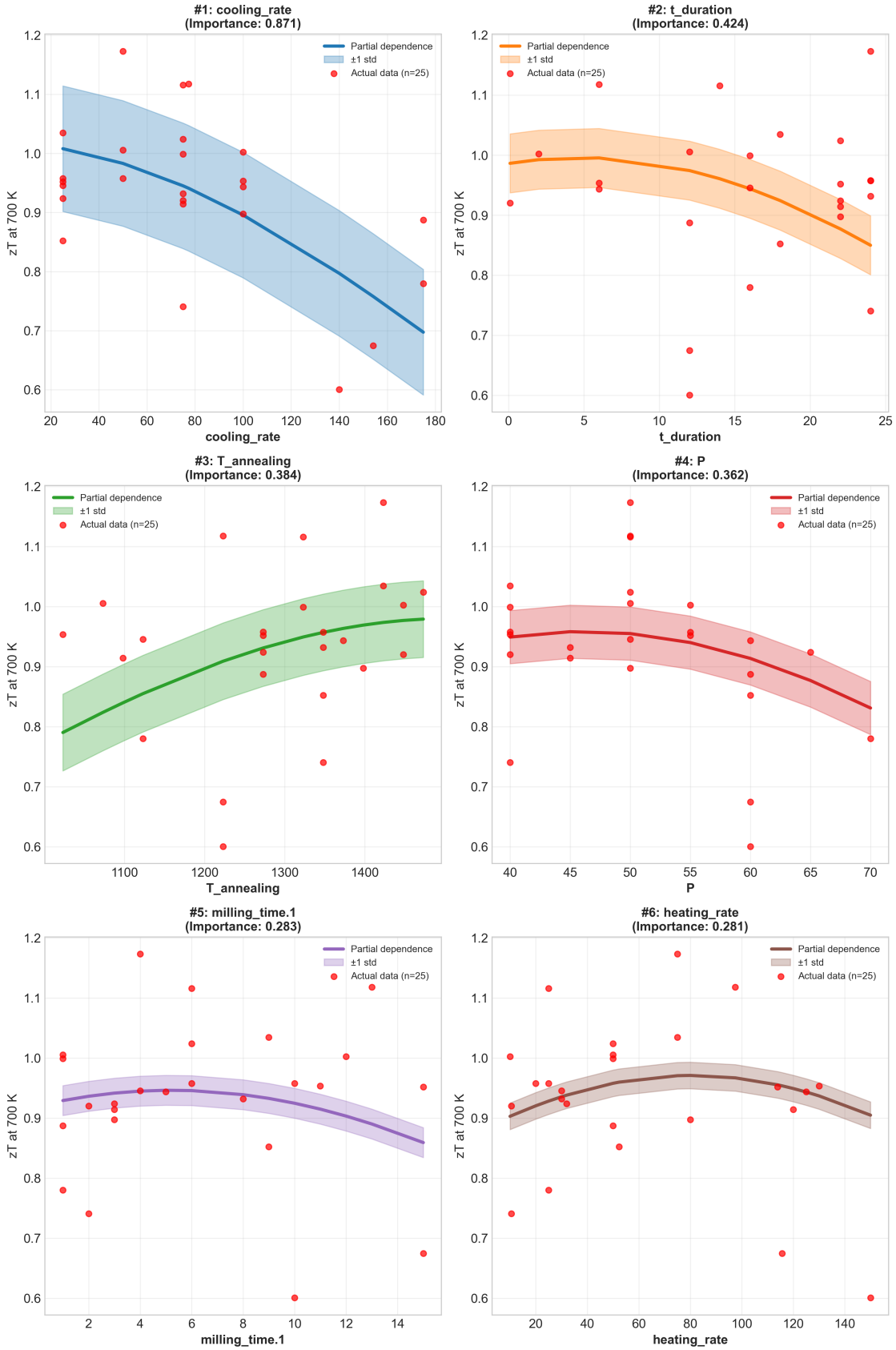

Figure S5: Partial dependence plots of the GP-fitted zT at 700 K for the six most important variables, in descending order: melt-cooling rate, dwell time, annealing temperature, SPS uniaxial pressure P, milling-time.1 and heating-rate. A ridge of high zT appears at rapid melt-quenching combined with short, slow-cooled SPS profiles.

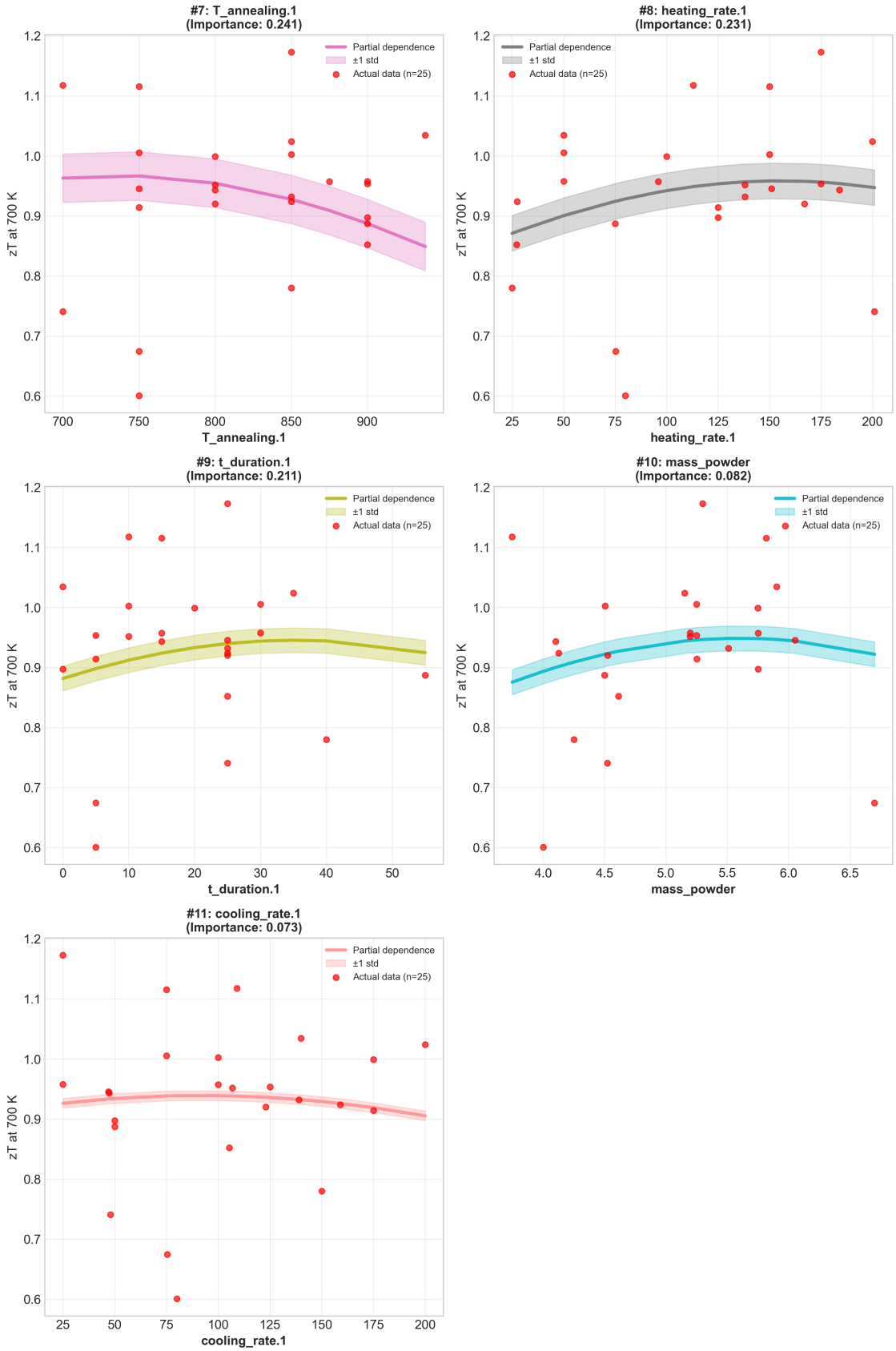

Figure S6: Partial dependence plots of the GP-fitted zT at 700 K for for the less influential variables, in descending order: annealing-temperature.1, heating-rate.1, dwell-time.1, mass-powder and cooling-rate.1. All show comparatively shallow trends, confirming their minor contribution to the overall variance in zT at 700 K.

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