Supplementary Materials

Piezoelectric Ceramic/Photopolymer Composites Curable with UV light: Ciscosity, Curing depth, and Cielectric Properties

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Abstract:

Four piezoelectric ceramic materials with varying particle sizes and geometries are added up to 30 vol.% to a photopolymer resin to form UV-curable piezoelectric composites. Such composites solidify in a few minutes, can be used in UV-curing-based 3D printing processes, and can achieve improved sensor performance. The particle dispersion with ultrasonication shows the most homogeneous particle dispersion with ethanol, while two other solvents produced similar results. The viscosities of the prepared suspensions show some dependency on the particle size. The curing depth results show a strong dependency on the ceramic particle size, the difference in refractive index, and the particle size distribution, whereby composites filled with PZT produced the worst results and composites filled with KNN produced the highest curing depths. The SEM images show a homogeneous dispersion of ceramic particles. The highest dielectric properties are also shown by KNN-filled composites, while BTO and PZT produced mixed results of dielectric constants and dielectric losses. KNN-filled composites seem to be very promising for further 3D-printable, lead-free piezoelectric composite development.

Keywords: BTO; KNN; PZT; piezoelectric composite; photopolymer resin; viscosity; cure depth; dielectric constant; dielectric loss

Contents

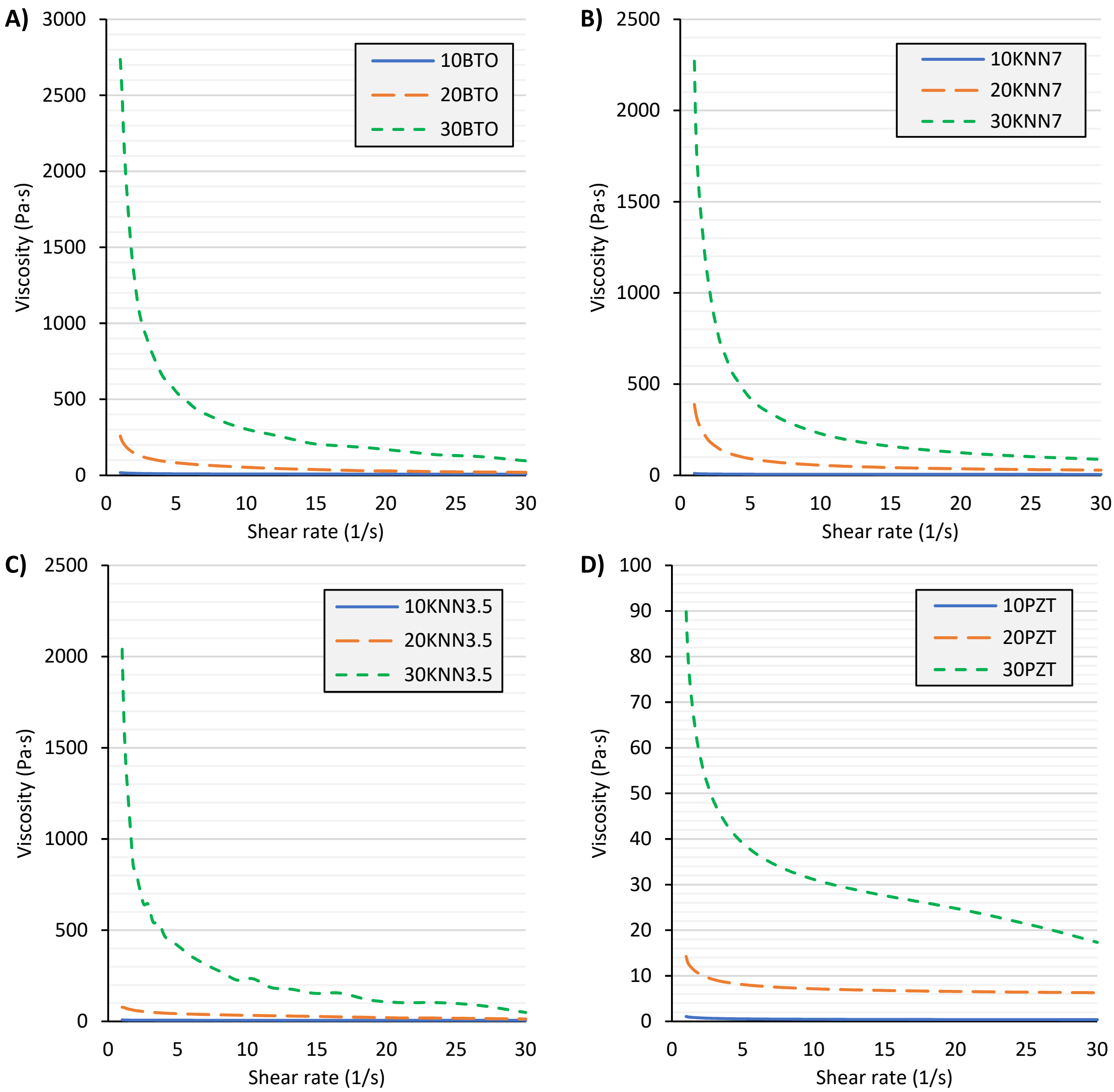

Figure S1. Viscosity of ceramic/photopolymer suspensions over the shear rate at room temperature: **(A)** BTO; **(B)** KNN7; **(C)** KNN3.5; **(D)** PZT.

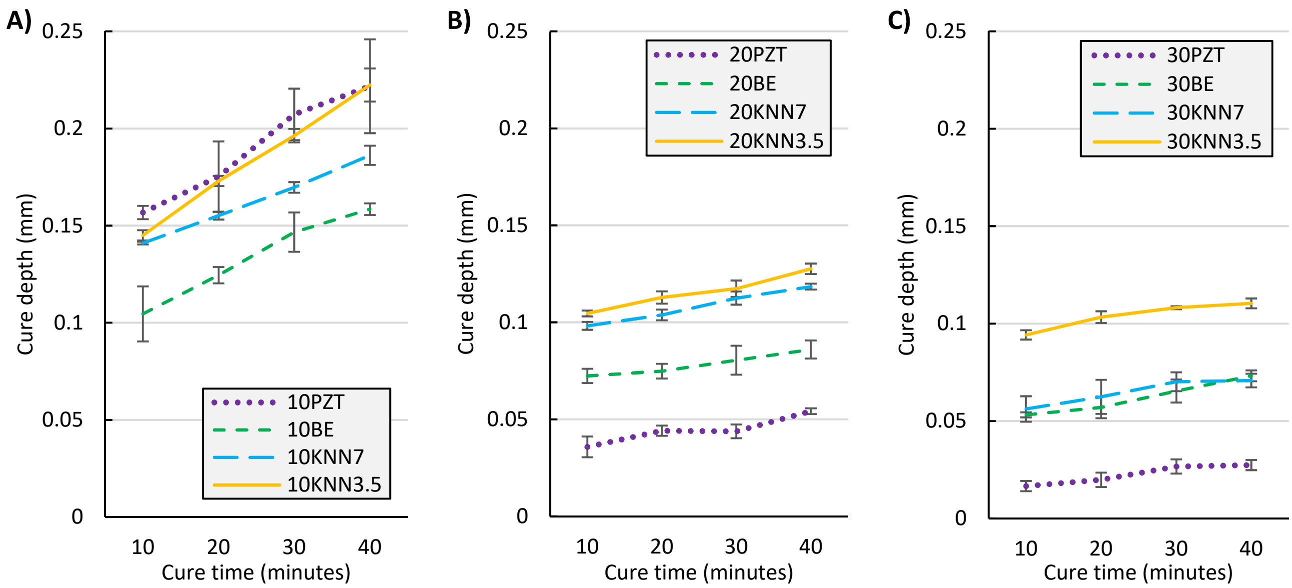

Figure S2. Cure depth of ceramic/photopolymer suspensions over time. Ceramic content: **(A)** 10 vol.%; **(B)** 20 vol.%; **(C)** 30 vol.%.

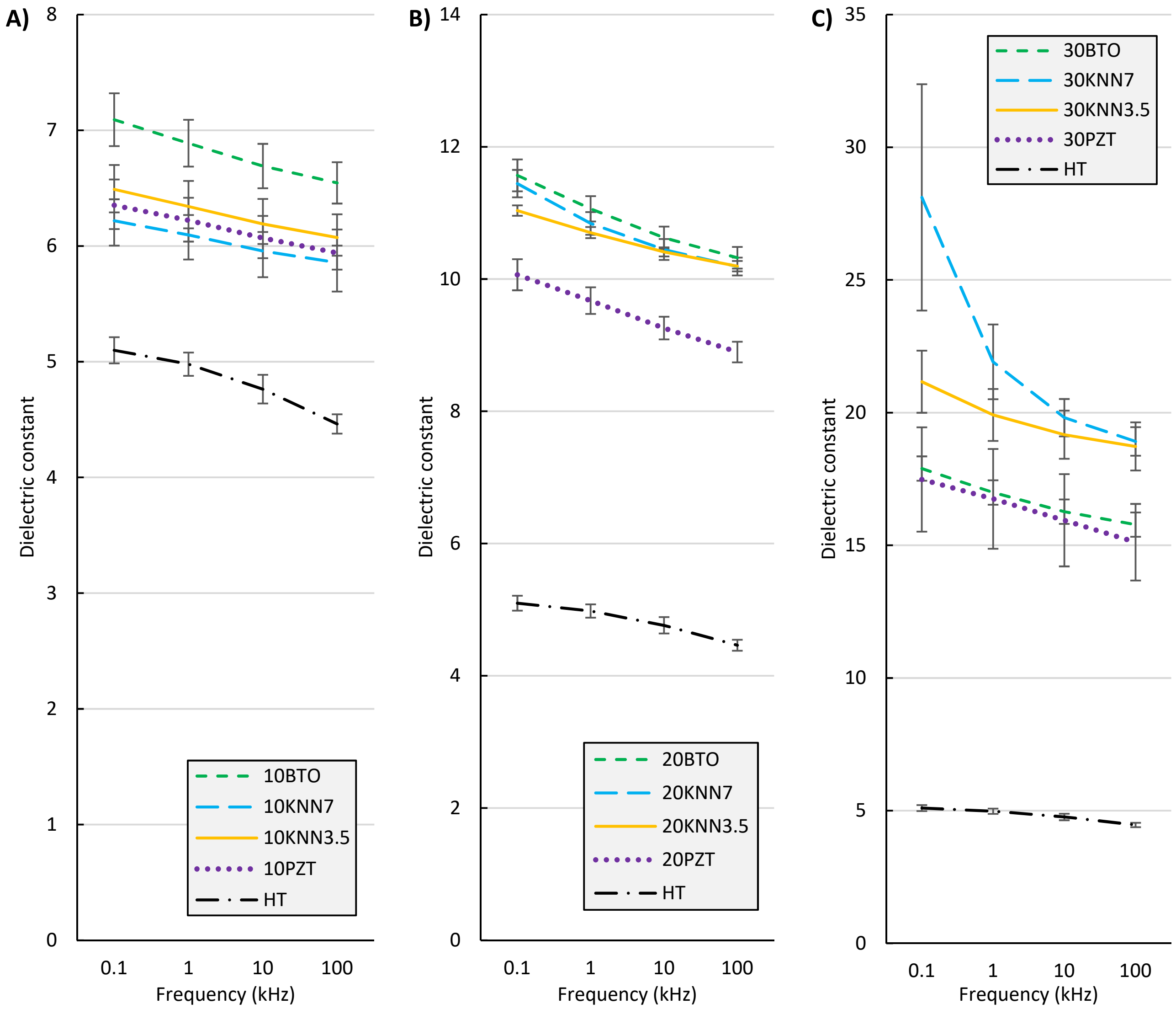

Figure S3. Dielectric constant (relative permittivity) at different frequencies at room temperature of piezoelectric composites. Ceramic content: **(A)** 10 vol.%; **(B)** 20 vol.%; **(C)** 30 vol.%. Note different Y-axis units.

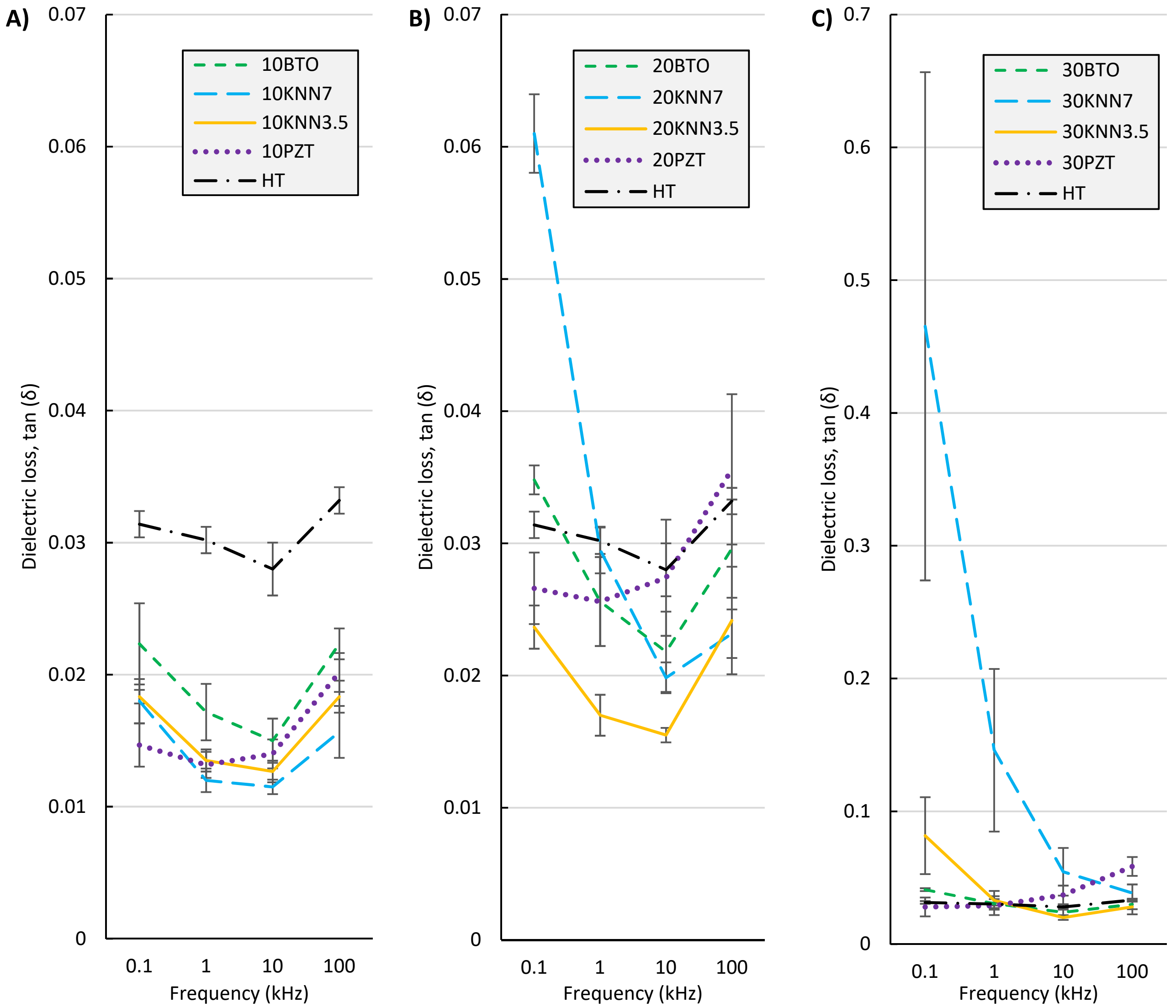

Figure S4. Dielectric loss (dissipation factor *tan (δ)*) at different frequencies at room temperature of piezoelectric composites. Ceramic content: **(A)** 10 vol.%; **(B)** 20 vol.%; **(C)** 30 vol.%. Note different Y-axis units.

Table S1. Viscosity of ceramic/photopolymer suspensions over the shear rate at room temperature.

Shear rate, 1/s	Viscosity, Pa*s														
	10BTO-A	10BTO-I	10BTO-E	HT	20BTO	30BTO	10KNN7	20KNN7	30KNN7	10KNN3.5	20KNN3.5	30KNN3.5	10PZT	20PZT	30PZT
1,00	15,38	18,44	16,96	3,231	258,36	2735,3	10,34	388,2	2270	8,2765	77,675	2040	1,0877	14,234	89,81
1,12	14,73	17,99	16,32	3,243	227,72	2523,8	9,1073	332,22	1860	7,5665	76,453	1680	0,98501	12,872	80,847
1,26	13,99	17,18	15,55	3,229	206,15	2193,5	8,652	293,63	1640	7,3433	72,317	1420	0,93046	12,217	74,661
1,42	13,28	16,34	14,82	3,193	187,3	1932,1	8,2888	263,1	1470	7,1772	68,26	1240	0,87768	11,656	69,594
1,60	12,69	15,54	14,16	3,16	171,04	1700,9	7,9691	237,79	1320	7,0331	65,122	1050	0,84427	11,137	65,135
1,80	12,14	14,77	13,56	3,158	157,16	1472,1	7,6806	214,05	1170	6,9136	61,993	854	0,80819	10,695	61,021
2,02	11,61	14,06	12,99	3,182	144,54	1288	7,4055	191,27	1050	6,8032	59,127	811	0,76494	10,274	57,682
2,27	11,07	13,45	12,43	3,19	133,26	1118,6	7,1539	172,74	925	6,7009	56,471	725	0,72959	9,8622	54,357
2,56	10,62	12,92	11,92	3,158	123,61	993,48	6,9427	157,01	818	6,6055	54,074	641	0,69444	9,5483	51,54
2,87	10,26	12,39	11,51	3,145	114,98	909,17	6,7367	140,83	720	6,5304	51,763	643	0,67029	9,2417	48,797
3,23	9,86	11,86	11,11	3,171	107,3	814,68	6,5422	128,64	652	6,4605	49,436	544	0,64066	8,9517	46,452
3,63	9,49	11,46	10,70	3,144	99,546	727,65	6,3823	116,7	572	6,3967	47,141	543	0,61435	8,687	44,246
4,08	9,21	11,07	10,39	3,135	92,966	645,39	6,2315	106,64	518	6,3385	45,213	465	0,59206	8,4528	42,191
4,59	8,88	10,70	10,06	3,143	86,863	595,43	6,0892	97,322	459	6,2917	43,569	438	0,5716	8,2293	40,31
5,16	8,65	10,38	9,80	3,117	81,047	533,33	5,9638	88,965	408	6,2487	41,923	406	0,55126	8,0275	38,625
5,81	8,39	10,08	9,53	3,115	75,626	483,92	5,8431	81,607	368	6,2116	40,313	365	0,53285	7,8462	37,04
6,53	8,19	9,80	9,31	3,111	70,523	428,57	5,7471	74,864	336	6,1775	38,813	332	0,51677	7,676	35,602
7,34	7,99	9,55	9,11	3,089	65,511	393,55	5,6543	68,807	303	6,1476	37,268	299	0,50001	7,5152	34,264
8,26	7,81	9,32	8,92	3,082	60,705	356,23	5,5714	63,331	273	6,1226	35,77	266	0,48604	7,3792	33,014
9,28	7,64	9,10	8,74	3,075	55,955	322,78	5,4969	58,351	246	6,0974	34,27	226	0,47274	7,2459	31,859
10,40	7,49	8,89	8,58	3,071	51,212	294,94	5,4304	53,905	220	6,0749	32,732	234	0,46044	7,1189	30,766
11,70	7,35	8,70	8,44	3,072	46,301	270,43	5,3691	49,822	198	6,0543	31,202	186	0,44844	7,0079	29,721
13,20	7,21	8,51	8,31	3,068	41,921	239,7	5,317	46,114	178	6,0355	29,453	176	0,43789	6,8999	28,713
14,80	7,10	8,33	8,18	3,062	38,403	207,77	5,2697	42,802	161	6,0178	27,404	154	0,42804	6,7976	27,691
16,70	6,98	8,14	8,07	3,051	33,803	193,95	5,2281	39,786	145	6,0015	24,94	155	0,41847	6,702	26,654
18,80	6,89	7,97	7,96	3,056	29,335	180,66	5,1902	37,06	131	5,9848	21,988	117	0,41017	6,6123	25,5
21,10	6,80	7,79	7,86	3,054	28,046	159,63	5,1564	34,602	118	5,9694	18,869	103	0,40251	6,5272	24,103
23,70	6,71	7,60	7,76	3,051	23,74	135,16	5,1252	32,341	107	5,9537	18,314	103	0,39499	6,4459	22,321
26,70	6,62	7,40	7,67	3,046	21,559	123,28	5,0961	30,3	96,585	5,9375	15,853	87,419	0,38841	6,3712	20,108
30,00	6,55	7,21	7,58	3,045	19,681	95,414	5,0685	28,424	87,562	5,9219	13,305	48,688	0,38235	6,3001	17,346

Table S2. Cure depth of ceramic/photopolymer suspensions over time.

	Cure depth, mm			
Material	10 min	20 min	30 min	40 min
10BTO-A	0,138	0,165	0,195	0,197
10BTO-I	0,157	0,196	0,238	0,262
10BTO-E	0,105	0,125	0,147	0,159
20BTO	0,072	0,075	0,081	0,086
30BTO	0,053	0,057	0,065	0,073
10KNN7	0,141	0,155	0,170	0,186
20KNN7	0,098	0,104	0,113	0,118
30KNN7	0,056	0,062	0,070	0,071
10KNN3,5	0,145	0,173	0,196	0,222
20KNN3,5	0,105	0,113	0,117	0,128
30KNN3,5	0,094	0,103	0,108	0,110
10PZT	0,157	0,175	0,207	0,222
20PZT	0,036	0,044	0,044	0,054
30PZT	0,017	0,020	0,027	0,027

Table S3. Dielectric constant (relative permittivity) and dielectric loss (dissipation factor *tan* (δ)) at different frequencies at room temperature of piezoelectric composites.

Dielectric constant (relative permittivity)					Dielectric loss (dissipation factor tan (δ))				
Material	0,1 kHz	1 kHz	10 kHz	100 kHz	Material	0,1 kHz	1 kHz	10 kHz	100 kHz
HT	5,099	4,979	4,763	4,462	HT	0,0314	0,0302	0,0280	0,0332
10BTO-A	7,506	7,324	7,088	6,942	10BTO-A	0,0214	0,0174	0,0148	0,0214
10BTO-I	8,017	7,743	7,484	7,300	10BTO-I	0,0276	0,0210	0,0182	0,0276
10BTO-E	7,091	6,888	6,691	6,545	10BTO-E	0,0223	0,0172	0,0150	0,0223
20BTO	11,568	11,062	10,624	10,322	20BTO	0,0348	0,0256	0,0218	0,0296
30BTO	17,892	16,990	16,267	15,779	30BTO	0,0410	0,0305	0,0240	0,0300
10KNN7	6,218	6,095	5,956	5,856	10KNN7	0,0180	0,0120	0,0115	0,0157
20KNN7	11,443	10,841	10,446	10,189	20KNN7	0,0610	0,0295	0,0198	0,0232
30KNN7	28,110	21,914	19,809	18,914	30KNN7	0,4653	0,1460	0,0545	0,0385
10KNN3.5	6,489	6,342	6,190	6,073	10KNN3.5	0,0183	0,0135	0,0127	0,0183
20KNN3.5	11,036	10,703	10,411	10,194	20KNN3.5	0,0237	0,0170	0,0155	0,0242
30KNN3.5	21,164	19,912	19,167	18,726	30KNN3.5	0,0817	0,0330	0,0200	0,0280
10PZT	6,352	6,223	6,068	5,940	10PZT	0,0147	0,0132	0,0140	0,0202
20PZT	10,065	9,674	9,259	8,896	20PZT	0,0266	0,0256	0,0274	0,0356
30PZT	17,480	16,751	15,943	15,117	30PZT	0,0280	0,0290	0,0370	0,0585