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Supplementary Materials for

Accelerated discovery of 3D printing materials using data-driven multiobjective optimization

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Supplementary Text

Stopping Criteria Considerations

Many different stopping criteria can be used when deciding when to stop the optimization process. The optimization process stopping criteria could be reached once the performance improvement has not exceeded a given threshold over several consecutive iterations. Another method is a fixed amount of time or cost that can be spent in total for experimentation. The time or cost limit is often the constraining factor in many different applications. As shown in this work, a stopping criterion could also be given with a fixed budget in the number of experiments, driven by time or other resource limitations. We decided to do a fixed number of iterations (30) since the fixed budget is the most commonly used criterion in practice.

Batching Experiments in Parallel

Batching the number of experiments conducted at once reduces the total amount of time needed for experimentation. However, if the total number of experiments is limited with a fixed budget, having less iterations with larger batch size could decrease the algorithm's ability to accurately predict potentially high-performing designs due to the surrogate model being updated less over the entire optimization process. In circumstances like ours, physical constraints limit the total number of samples it was feasible to produce at once. When selecting the number of experiments that can be batched together, these trade-offs and constraints must be considered.

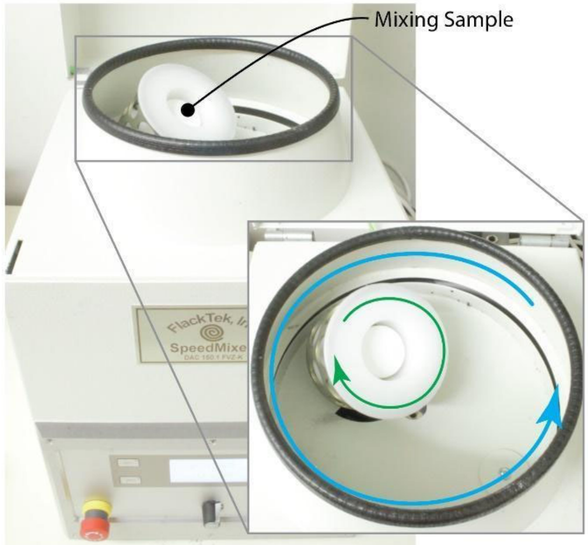

Fig. S1.
Formulation mixer. Dual asymmetric centrifuge mixer utilized for sample fabrication pipeline. The arrows depict the rotation of the sample during operation. (Photo Credit: Michael Foshey, Massachusetts Institute of Technology)

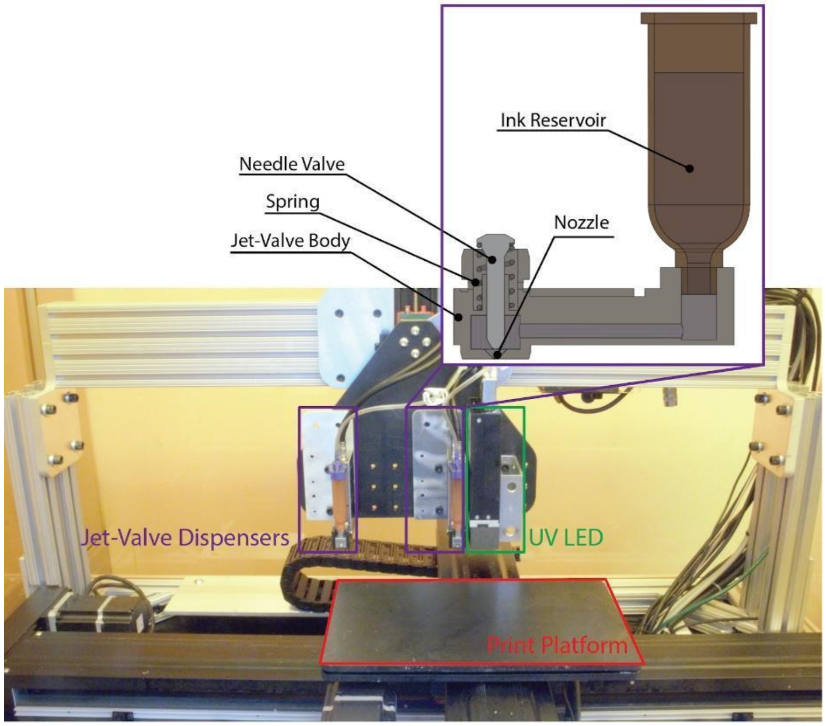

Fig. S2.
Jet-valve 3D printer utilized in the sample fabrication pipeline. Circled in purple are the 2 jet-valve dispensers used for dispensing the material in parallel. Circled in green is the UV LED used for curing the dispensed material. Circled in red is the print platform, the area where the samples are dispensed onto and cured. (Photo Credit: Michael Foshey, Massachusetts Institute of Technology)

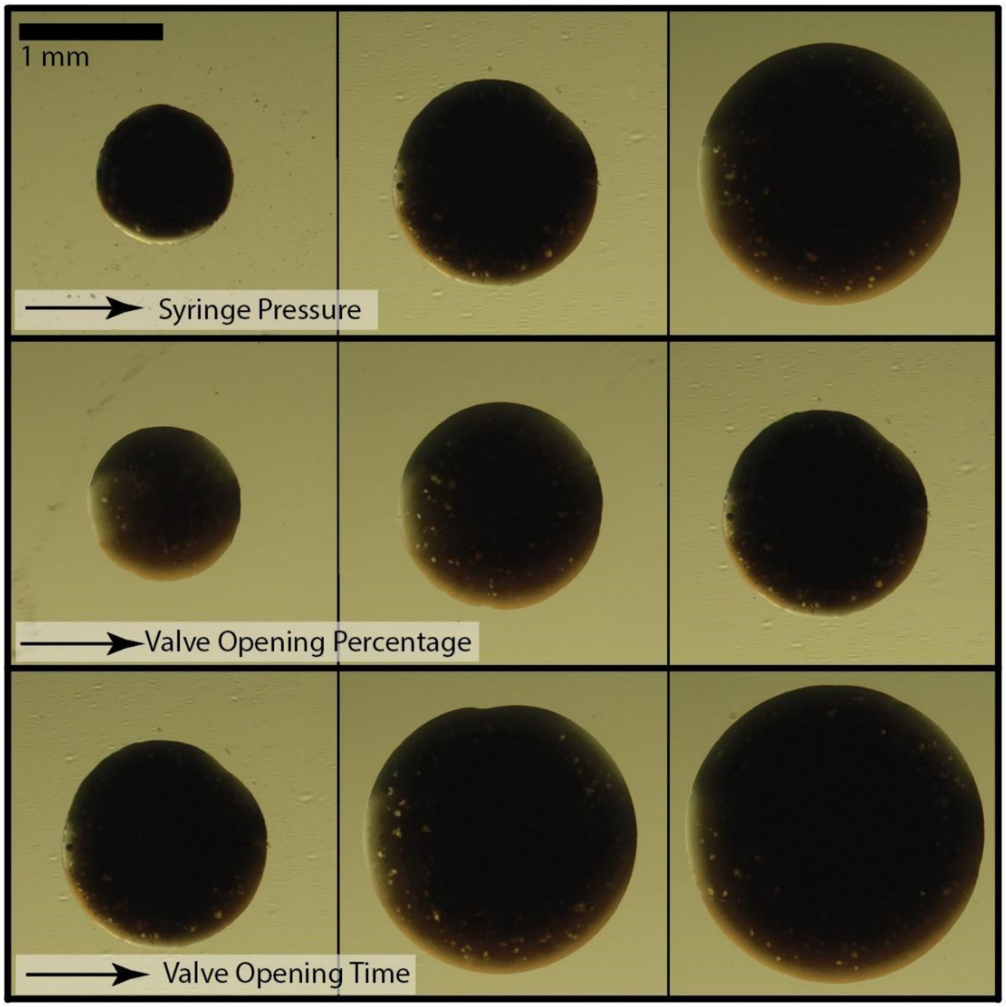

Fig. S3.
Optical microscopy images of jet-valve dispensing droplets. Droplet variations in Syringe pressure (top) (parameters from left to right: 15 psi, 35 psi, 50 psi, all 90 % opening percentage and 1 ms opening time), valve opening pressure (middle) (parameters from left to right: 70 %, 80 %, 90 %, all 35 psi syringe pressure and 1 ms opening time), and valve opening time (bottom) (parameters from left to right: 1 ms, 1.5 ms, 2 ms, all 35 psi syringe pressure and 90 % opening percentage).

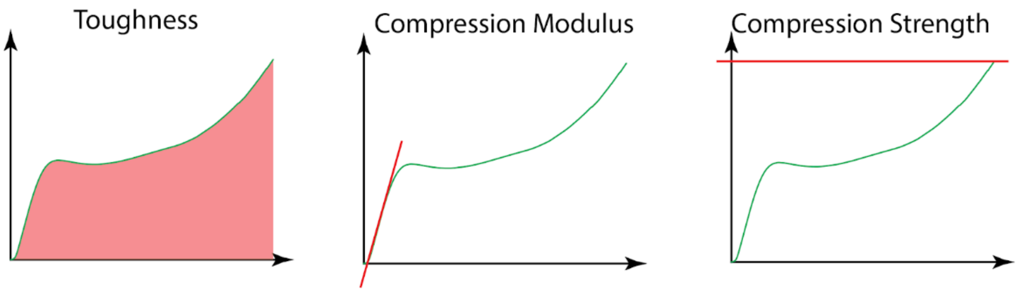

Fig. S4.
The 3 performance objectives that formulations are optimized for. Performance objectives are parsed from stress-strain data that is resultant of the compression testing.

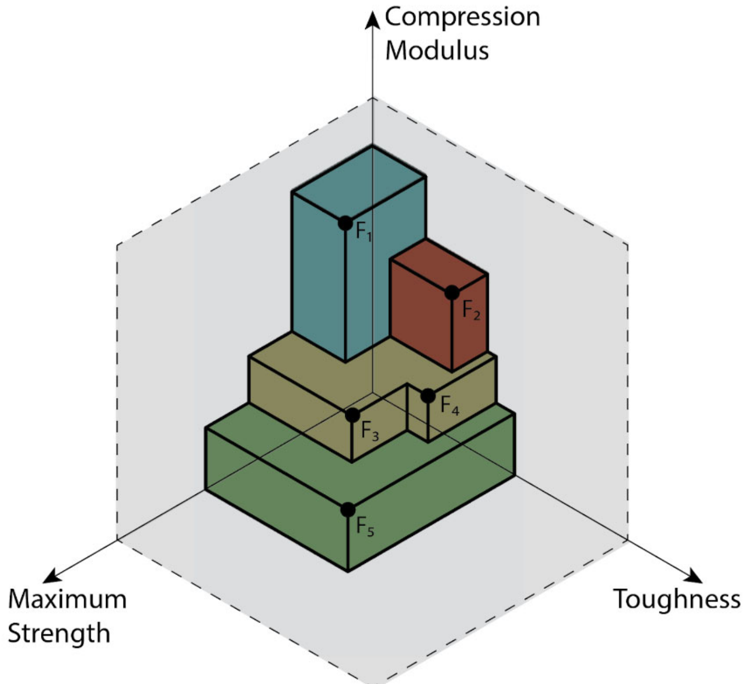

Fig. S5. Hypervolume calculation. A hypervolume that is calculated on Pareto optimal points F_1, F_2, F_3, F_4, and F_5, with an origin as a reference point.

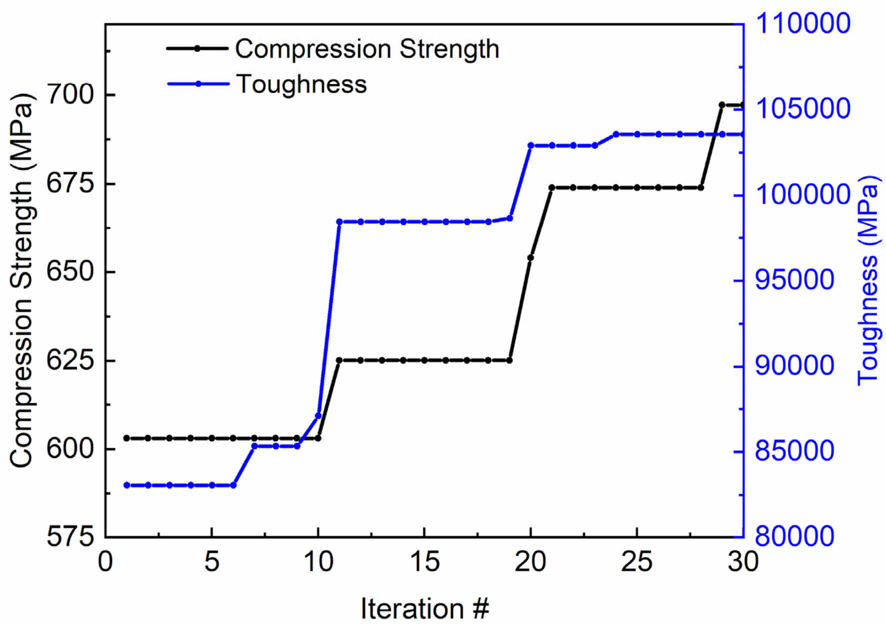

Fig. S6.

Performance objective improvement. The influence of iteration number on the maximum attained compression strength and toughness over the entire optimization process. The maximum attained compression modulus did not increase over the entire optimization process (not shown in this figure).

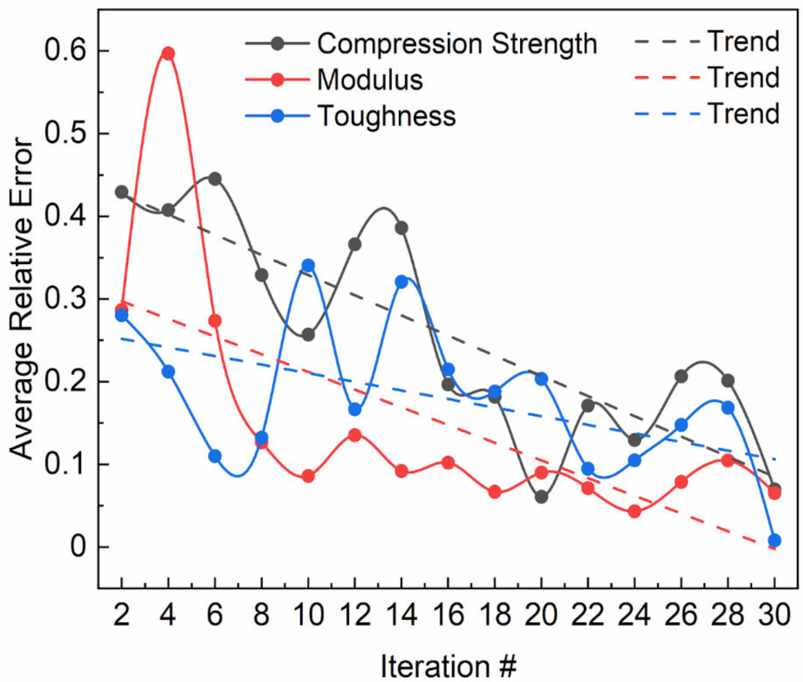

Fig. S7.

Gaussian process performance evolution. The influence of iteration number on the relative model prediction error averaged over four samples in each iteration. The relative average error is calculated by (predicted performance-experimental performance)/(experimental performance) for every formulation in each iteration and then leveraging them. As indicated by the trendlines, with the number of iterations increases and more data points are added to the model, the model prediction accuracy increases. In circumstances where only sparse data is available for model training, the model's prediction capability is relatively low and is not recommended for prediction; however, they are still helpful for optimization problems.

Table S1.
The average duration and standard deviation of each time processing step within the sample fabrication pipeline.

	Dispensing	Mixing	Syringe Loading	3D Printing	Post-Processing	Testing	Total
Average Time	157.7 s	64.7 s	45.7 s	1975 s	4354 s	199.7 s	**6797 s**
Standard Deviation	3 s	0.6 s	3.2 s	190.9 s	13.54 s	30 s	**241 s**

Table S2.
Formulations of the 6 primary inks (weight percentage).

Primary Formulation	Oligomer	Diluent	Photoinitiator	Surfactant
A	59% Modified Urethane Di-Acrylate	39.5% Acrylic Acid Amide	0.5% Bis(2,4,6-trimethylbenzoyl)-phenylphosphineoxide	1% Tween-20
B	69% Aliphatic Urethane Di-Acrylate	30% Acrylic Acid Ester	0.5% Bis(2,4,6-trimethylbenzoyl)-phenylphosphineoxide	0.5% Tween-20
C	69% Aliphatic Urethane Hexa-Acrylate	30% Acrylic Acid Ester	0.5% Bis(2,4,6-trimethylbenzoyl)-phenylphosphineoxide	0.5% Tween-20
D	69% Aliphatic Urethane Di-Acrylate	30% Polyoxyethyleneamine	0.5% Bis(2,4,6-trimethylbenzoyl)-phenylphosphineoxide	0.5% Tween-20
E	59% Modified Urethane Di-Acrylate	40% Acrylic Acid EsterRahn Genomer 1121	0.5% Bis(2,4,6-trimethylbenzoyl)-phenylphosphineoxide	0.5% Tween-20
F	89.5% Urethane Dimethacrylate	10% Acrylic Acid Ester	0.5% Bis(2,4,6-trimethylbenzoyl)-phenylphosphineoxide	

Table S3.

Viscosity of the 6 primary inks measured with a rotary viscometer (Brookfield, Middleboro, MA, USA).

Primary Formulation	Viscosity (cP)	Rotational Speed (rpm)
A	640	1
B	180	1
C	60	5
D	60	5
E	610	1
F	160	5

Table S4.

The composition and performance of the 150 samples that were tested during the optimization process. Formulations with no mechanical performance values could not be tested due to being too brittle to manufacture or test. The formulations are listed in order of when each sample was tested during the optimization process.

Mechanical Performance			Formulation					
Compression Strength [MPa]	Compression Modulus [MPa]	Toughness [GPa]	A [wt%]	B [wt%]	C [wt%]	D [wt%]	E [wt%]	F [wt%]
455	1.37	68113	0	18	21	28	20	13
280	2.11	51731	0	2	38	4	29	27
470	1.76	77877	19	30	10	6	7	28
440	1.96	76655	8	16	14	4	19	39
366	1.23	62118	34	6	23	16	19	2
411	1.12	66050	14	10	7	20	27	22
136	0.90	22269	7	7	32	21	3	30
369	1.31	61644	2	1	27	13	49	8
534	0.90	68785	9	32	3	13	37	6
443	0.73	59416	8	21	2	0	61	8
507	0.98	66651	9	32	4	21	17	17
444	1.12	65323	14	30	7	10	25	14
490	1.33	72484	4	17	7	22	24	26
359	1.73	60280	17	15	28	29	0	11

485	0.89	66192	10	14	14	26	27	9
229	0.73	49588	100	0	0	0	0	0
436	1.17	60782	0	100	0	0	0	0
368	0.04	29840	0	0	0	100	0	0
558	1.01	72324	60	40	0	0	0	0
545	1.01	76481	55	45	0	0	0	0
596	1.03	80252	50	50	0	0	0	0
578	1.03	78300	45	55	0	0	0	0
603	1.05	79495	40	60	0	0	0	0
536	1.34	83057	27	44	9	4	8	8
381	1.58	63939	4	19	36	20	12	9
488	1.69	74453	0	59	0	8	8	25
492	2.04	77476	0	39	13	7	3	38
483	1.70	74332	1	51	0	10	2	36
461	1.53	68684	1	62	1	3	10	23
429	1.96	73565	6	30	41	1	1	21
420	1.75	70958	8	32	50	7	2	1
186	2.00	39188	15	28	25	1	6	25
308	2.93	68674	0	0	0	0	0	100

248	0.34	41639	0	0	0	0	100	0
330	1.61	58955	2	11	0	50	3	34
261	2.54	54114	1	19	0	0	0	80
374	2.13	72005	1	39	0	2	1	57
442	1.64	73645	18	12	0	36	0	34
326	2.26	65493	1	0	13	25	0	61
407	1.71	79299	32	0	12	1	0	55
281	2.09	55835	1	15	10	21	0	53
345	2.26	66935	1	0	15	25	0	59
338	1.71	65225	15	45	4	3	1	32
381	0.61	47362	5	32	0	57	6	0
424	1.31	68133	31	52	0	8	2	7
413	0.72	59056	15	4	13	67	1	0
390	1.30	60316	0	21	20	58	1	0
244	1.58	43427	0	1	28	67	1	3
-	-	-	0	0	4	0	0	96
419	1.12	61445	1	28	11	46	11	3
463	1.47	81857	49	6	0	14	2	29
-	-	-	10	2	5	0	3	80

260	1.94	59061	14	0	7	1	1	77
461	1.57	75813	3	58	18	4	10	7
435	1.97	80546	0	31	1	2	15	51
411	1.71	85333	25	1	1	4	13	56
594	1.08	82485	29	45	6	8	12	0
332	1.23	62332	2	1	16	1	50	30
462	1.57	80821	2	24	0	3	26	45
386	1.91	76224	0	1	0	1	27	71
459	1.39	78607	9	2	0	6	40	43
386	1.80	73308	0	16	0	0	25	59
421	1.90	83070	32	9	2	1	1	55
256	2.25	53258	0	8	13	0	11	68
344	1.90	72698	39	1	3	0	0	57
317	1.92	66141	26	18	2	1	1	52
406	1.94	81223	20	16	0	0	0	64
579	1.04	87101	51	31	7	1	1	9
297	2.27	61772	14	17	2	0	0	67
230	1.78	53154	0	3	2	21	32	42
296	0.89	60049	57	0	11	1	10	21

625	1.13	94709	51	25	10	2	12	0
559	1.56	98461	51	1	7	2	11	28
350	2.67	74202	0	0	0	2	0	98
484	1.58	91455	60	0	3	0	2	35
334	1.83	67470	34	0	11	1	6	48
401	1.52	75082	44	11	11	0	6	28
340	1.17	68371	66	0	8	0	0	26
620	1.32	92325	37	47	10	0	6	0
457	1.03	73400	44	30	9	4	13	0
527	1.21	80673	27	61	12	0	0	0
182	2.15	33836	1	0	0	2	8	89
-	-	-	30	2	3	1	19	44
299	1.91	62780	21	11	6	0	14	48
-	-	-	30	5	3	0	19	43
342	1.81	73191	52	1	1	0	0	46
293	2.10	64377	0	38	1	0	0	61
284	2.03	60696	0	31	1	0	0	68
419	1.51	75788	32	34	2	0	1	31
273	1.95	60035	1	41	0	0	1	57

325	2.00	65022	0	41	0	13	0	46
361	2.09	69396	0	18	0	26	1	55
310	1.92	61302	4	45	0	7	1	43
321	2.02	62666	0	25	0	25	0	50
408	1.58	76318	36	20	2	8	0	34
398	1.91	74553	21	0	0	33	1	45
438	1.81	79745	25	0	0	31	1	43
403	1.88	75747	19	0	0	32	1	48
368	1.77	65606	0	63	10	0	0	27
393	1.89	69484	0	66	9	0	0	25
317	2.20	63762	0	33	11	1	0	55
356	1.80	64450	0	68	9	0	1	22
438	2.12	87349	35	0	0	1	0	64
335	2.04	65124	1	0	0	36	0	63
514	1.54	93644	48	0	1	0	12	39
526	1.92	98664	39	0	0	2	1	58
555	1.41	91162	40	17	0	0	12	31
654	1.12	96006	44	27	0	0	17	12
613	1.35	102904	34	14	0	0	13	39

575	1.48	99855	33	12	0	0	13	42
596	1.59	97568	39	0	0	0	22	39
674	1.31	96610	39	31	0	0	22	8
571	1.57	97058	41	0	0	0	25	34
454	1.86	85375	35	0	0	0	20	45
512	1.78	93176	33	0	0	0	21	46
385	1.85	77998	31	0	0	0	20	49
536	1.71	96498	35	0	0	0	21	44
290	1.97	66399	20	0	0	0	1	79
597	1.00	92485	50	0	0	0	30	20
391	0.89	64361	53	6	0	0	32	9
430	0.80	68900	53	0	0	0	33	14
397	1.08	76450	42	0	0	0	27	31
413	1.73	80203	42	1	0	0	14	43
512	1.67	92117	39	0	0	0	13	48
627	1.59	103559	44	0	0	5	13	38
459	1.88	85164	28	0	0	11	8	53
489	1.87	89171	42	12	0	0	6	40
523	1.82	92589	42	11	0	1	7	39

484	2.01	89597	42	11	0	0	4	43
507	1.87	91083	45	8	0	0	5	42
415	1.44	76846	41	14	0	0	7	38
498	1.59	86258	41	12	0	0	6	41
387	1.52	74536	42	16	0	0	7	35
605	1.70	100895	40	13	0	0	8	39
330	2.39	72121	25	0	0	0	0	75
431	2.06	85777	27	0	0	0	0	73
379	1.24	79506	23	0	0	0	0	77
-	-	-	24	0	0	0	0	76
-	-	-	32	0	0	0	9	59
374	1.76	76874	27	0	0	0	14	59
305	1.89	67571	30	0	0	0	9	61
474	1.64	89097	32	1	0	0	9	58
697	1.10	95259	37	37	0	0	19	7
632	1.02	84624	38	41	0	1	19	1
-	-	-	39	39	0	0	22	0
558	1.15	81235	34	36	0	0	20	10
500	1.68	86776	39	23	0	0	0	38

502	1.65	86185	37	22	0	1	0	40
498	1.68	87186	41	21	0	1	0	37
465	1.95	85124	32	19	0	0	0	49