Final Report

AI Guided Alloy Design

Prepared for:

Paulette Clancy, pclancy3@jhu.edu
Department of Chemical and Biomolecular Engineering, 124 Latrobe Hall
John Hopkins University, Whiting School of Engineering
3400 North Charles Street
Baltimore, MD 21218-2682

Submitted by:

Eddie Gienger, Project Manager
443-280-0946
Eddie.Gienger@jhuapl.edu

Morgan Trexler, Program Manager
Science of Extreme and Multifunctional Materials
240-228-9817
Morgana.Trexler@jhuapl.edu

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The Johns Hopkins University Applied Physics Laboratory (JHU/APL)
11100 Johns Hopkins Road, Laurel, MD 20723-6099

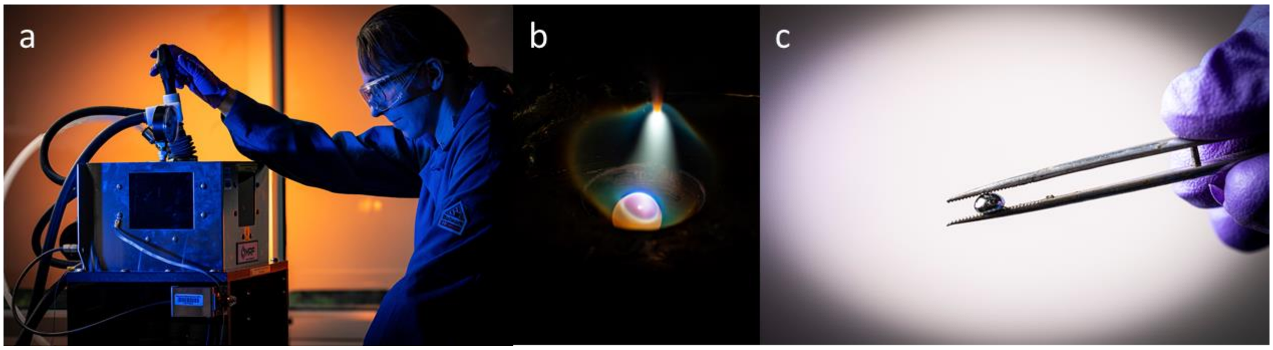

Figure 1: An overview of the arc melting process including (a) the MRF arc melting furnace (b) a close up of a molybendum alloy being melted and (c) the final form factor of arc melted ingots

I. Project Overview

Multiple principal element alloys (MPEAs) are a class of alloy that frequently have desirable properties such as high strength, hardness, and temperature stability. By mixing roughly equi-atomic amounts of four or more elements, targeted FCC and BCC structures are formed, theoretically enabled by the high entropy of mixing multiple base metals. While these materials have shown promise, the potential compositional variations are massive, making machine learning guided exploration of the space promising. However, this is complicated by the relatively limited availability of materials properties data.

The Clancy Group at Johns Hopkins University Whiting School of Engineering has successfully demonstrated the use of the Physical analytics pipeline, PAL 2.0 model, to efficiently navigate complex compositional spaces compared to Edisonian approaches. PAL 2.0 combines a physics-based surrogate model with Bayesian optimization. PAL 2.0 uses a physics-based hypothesis to make predictions using neural networks. Prior work has generated predictions of high hardness MPEAs, however, these predictions are largely unverified with synthesis and experimental characterization.

The project had two main objectives:

1. Fabricate samples of compositions predicted by the Clancy Group. Predictions of high hardness (greater than 1000 Vickers hardness) MPEA compositions were made using the PAL 2.0 framework from existing data sets. JHU/APL arc melted small MPEA ingots. Raw material forms (powders, foils) were mixed in mass percentages predicted by the models. These materials were then melted to form a single alloy ingot using an MRF SA-200 arc melter.
2. Validate the PAL 2.0 model for high hardness MPEAs by measuring and comparing to experimental data. In this closed-loop discovery process, improvements are made by informing the model with actual experimental data. The impact of this process is measured by its ability to predict experimentally validated material properties. In this effort, MPEAs were characterized with Vickers hardness testing.

II. Technical Accomplishments

Task 1: Experimental synthesis of proposed MPEAs

A total of 20 samples was fabricated from the recommendations list provided by the Clancy Group (Attachment 1). Samples were prioritized for synthesis based on availability of elements (no materials were purchased on this project) and perceived ease of synthesis. For example, samples containing Manganese tend to vaporize during melting making synthesis of a targeted compositions difficult.

Table **1** contains a list of samples that were synthesized, the starting weight of the raw elements, and the final ingot weight.

Some of the samples lost significant mass during synthesis, particularly samples that contain a mixture of light elements (aluminum and manganese) with refractory elements (molybdenum and tungsten). This is expected given that aluminum boils at 2,470 ^{0}C and tungsten doesn't boil until temperatures above 3,000 ^{0}C. In the future, alloying strategies can be used to mitigate these losses, but these invesitgations were outside the scope of this project.

Table 1: Synthesized samples including model-recommended elemental weight percentage. The target mass for each sample was 700 mg.

Sample #	Recommended Elemental Comp (wt%)			Sample Masses (mg)					Post Melt Weight (mg)
	Fe	Mo	Si		Fe	Mo	Si	Total	
170	31	55	14	Target:	218	382	100	700	
				Measured:	217	382	102	701	700
	Al	Fe	Mo		Al	Fe	Mo	Total	
118	15	29	55	Target:	108	206	386	700	
				Measured:	110	207	385	702	702
	Al	Mn	Mo		Al	Mn	Mo	Total	
283	16	32	51	Target:	114	226	360	700	
				Measured:	115	225	362	702	494
	Co	Si	Ta		Co	Si	Ta	Total	
42	23	11	67	Target:	158	74	468	700	
				Measured:	161	72	468	702	695
	Co	Fe	Si		Co	Fe	Si	Total	
114	39	41	20	Target:	271	285	143	700	
				Measured:	271	287	146	704	702
	Co	Si	Ta		Co	Si	Ta	Total	
53	22	11	67	Target:	153	75	472	700	
				Measured:	155	78	472	705	701
	Al	Mn	Mo		Al	Mn	Mo	Total	

283 Redo	16	32	51				Target:	114	226	360				700	
							Measured:	114	255	363				732	580
	Co	Fe	Si	Ti	W			Co	Fe	Si	Ti	W		Total	
3	5	6	4	17	68		Target:	38	42	27	119	475		700	
							Measured:	41	43	29	121	475		708	656
	Al	Co	Fe	Mo	Si	Ta		Al	Co	Fe	Mo	Si	Ta	Total	
89	5	3	6	28	3	55	Target:	36	22	45	195	18	383	700	
							Measured:	36	26	48	198	18	383	708	702
	Al	Co	Mo	V				Al	Co	Mo	V			Total	
954	17	38	29	15			Target:	121	269	202	108			700	
							Measured:	120	269	203	109			701	711

	Al	Cr	Mo					Al	Cr	Mo				Total	
780	15	31	54				Target:	107	214	379				700	
							Measured:	107	214	379				699	668
	Fe	Si	V	W				Fe	Si	V	W			Total	
72	13	3	19	65			Target:	90	23	134	453			700	
							Measured:	89	23	138	456			705	644
	Al	Co	Ni	Si				Al	Co	Ni	Si			Total	
9	22	31	33	14			Target:	154	215	230	101			700	
							Measured:	156	218	231	101			706	432
	Cr	Mn	Mo					Cr	Mn	Mo				Total	
420	26	27	47				Target:	184	186	329				700	
							Measured:	186	202	332				720	530
	Al	Co	V					Al	Co	V				Total	
461	20	44	36				Target:	138	310	253				700	
							Measured:	135	311	253				699	646
	Al	Co	Ti					Al	Co	Ti				Total	
233	20	45	35				Target:	140	316	244				700	
							Measured:	140	318	244				702	698
	Al	Co	Mo	Ti				Al	Co	Mo	Ti			Total	
430	17	40	24	18			Target:	121	280	171	127			700	
							Measured:	123	284	175	126			706	701
	Al	Co	V					Al	Co	V				Total	
489	20	45	36				Target:	137	313	251				700	

		Measured:	137	310	251	699	675
	Cr Hf Si		Cr	Hf	Si	Total	
149	20 70 10	Target:	138	491	71	700	
		Measured:	136	492	72	700	639
	Al V W		Al	V	W	Total	
130	10 20 70	Target:	67	141	493	700	
		Measured:	68	143	490	701	300

Task 2: Characterization and analysis of synthesized alloys

A total of seven samples were chosen for testing with Vickers hardness testing to compare to the predictions. Samples were prioritized from Table 1 based on how close the final mass to the target mass, thus ensuring the composition of the sample is close to that predicted by the model. Samples that lost significant mass were not tested because their final composition was unkown. Between three and five indents were taken on each sample. Indents were performed at either 300 or 500 grams of indent force. In previous efforts all samples were tested at 500 grams. However, some of the very high hardness samples experienced brittle cracking during indentation that can be mitigated by decreasing indentation load. The results from these tests are listed in Table 2.

Table 2: Measured and predicted hardness values for synthesized materials

Sample #	Indent	Force (g)	Measured VHN	Predicted VHN
42	1	300	1185	
	2	300	1101	
	3	300	1093	
	4	300	1175	
	Average	300	**1137**	**931**
53	1	500	1170	
	2	500	1245	
	3	500	1363	
	4	500	1309	
	Average	500	**1269**	**894**
72	1	300	880	
	2	300	992	
	3	300	872	
	4	300	864	
	Average	300	**900**	**862**
114	1	500	974	
	2	500	952	
	3	500	940	

	4	500	933	
	Average	500	**950**	**894**
118	1	300	750	
	2	300	728	
	3	300	711	
	4	300	653	
	Average	300	**709**	**1032**
170	1	500	1297	
	2	500	1323	
	3	500	1387	
	4	500	1081	
	Average	500	**1263**	**1042**
489	1	300	475	
	2	300	479	
	3	300	455	
	4	300	494	
	Average	300	**476**	**839**

III. Important Findings and Conclusions

- The PAL 2.0 model is able to predict high hardness materials in the MPEA design space. Through three rounds of predictions (the first two happening on previous efforts), the hardness values for synthesized materials has increased. Multiple materials had Vickers hardness values (VHN) over 1200 VHN, exceeding anything in the initial training data set (Figure 1).
- Additional considerations need to be paid to the idea of "synthesizability". Several interesting compositions predicted by the model contain elements of very different melting temperatures making JHU/APL's rapid arc melting approach difficult.

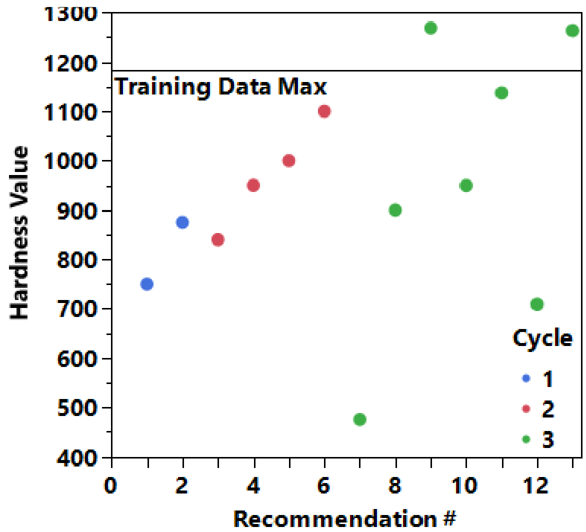

Figure 2: Experimentally measured hardness values for each recommended composition. Plot includes prior data from cycles 1 and 2. Within each cycle, samples are ordered by the model predicted hardness from low to high

IV. Implications for Further Research

1. Two proposed approaches can be taken to more accurately synthesize some of the predicted compostions. The first is to incorporate some of the physics behind synthesizability (i.e., melting temperature) into the model; the second is to explore additional processing routes (quartz ampule melting, multiple alloying steps) into the synthesis approach.
2. Given the model has predicted high hardness materials that are relatively cheap, light, and straightforward to synthesize, effort should be taken to scale up the material from the small ingot used for *discovery* to a *useful material system* in an application like hard coating for wear resistance.
3. There are several other metallic microstructures like B2 phases for strengthening that potentially possess similiarities to the design framework used for this project. These materials have demonstrated high temperature strength and would be useful for several applications where room temperature ductility is also needed. Taking the lessons learned from this project and applying it to a more complex, high impact material class is worth investigating.