Russian Science Foundation

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Multi-objective optimization and Coevolutionary search for materials with optimal properties

Zahed Allahyari

19th USPEX workshop on crystal structure prediction with USPEX code.

November 11, 2020

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And is based on the following papers:

- Z. Allahyari, A. R. Oganov. Nonempirical Definition of the Mendeleev Numbers: Organizing the Chemical Space. ***J. Phys. Chem. C***,124, 43, 23867–23878 (2020).
- Z. Allahyari, A. R. Oganov. Coevolutionary search for optimal materials in the space of all possible compounds. ***NPJ Comput Mater***, 6, 55 (2020).
- Z. Allahyari, A. R. Oganov. Multi-objective Optimization as a Tool for Material Design. In: Andreoni W., Yip S. (eds) Handbook of Materials Modeling. Springer, Cham (2020).

Objectives

1. To solve the challenging and the main problem of material science: predicting optimal materials in any desired properties, let's say in the space of all binary systems.

2. To make sure, that the predicted materials have a high chance to be synthesized. We don't need unrealistic materials which are only perfect in computers.

3. Such searches are huge. To reduce the time and computational cost of the searches, without significant loss in the results.

4. To have an automatic and unbiased search for hard/superhard materials. Is diamond the hardest material in nature? Are there any new super(hard) materials?

Outline

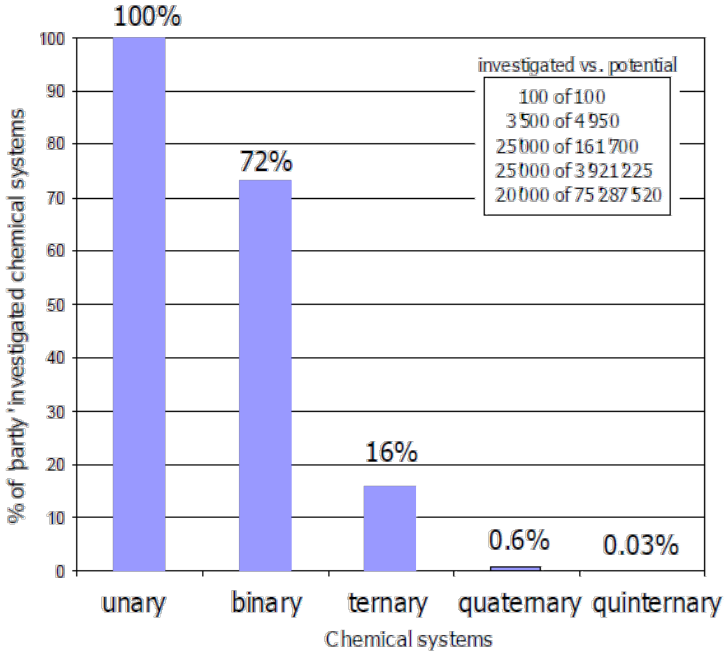
Search for materials with optimal target properties:

Mendelevian **S**erach-**MendS** (algorithm, method)[1][2].

Defining a good chemical space.

Multi-objective Pareto technique.

Results on searching (using MendS) for hard/superhard and low energy binary materials under zero pressure in the entire chemical space.



[1] Patent No: WO2018009090A1

[2] *NPJ Comput. Mater.* **6**, 55 (2020).

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Desired properties

Most optimal Materials

MendS

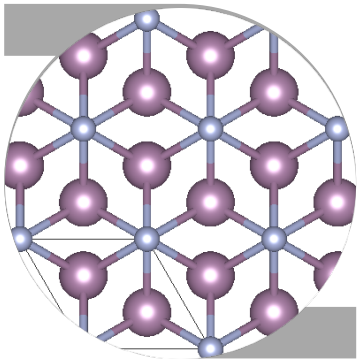
Most optimal phases and structures of X

USPEX

Which Materials ? (i.e. X), & properties?

Structure search methods

Search for the most optimal phases of a **specific** material.



Calculation of **Energy** and **Properties**. (i.e. magnetic, electronic, mechanical etc).

Ab initio codes

Energy and Properties

What is coevolution and the difference of evolutionary algorithm and coevolutionary algorithm?

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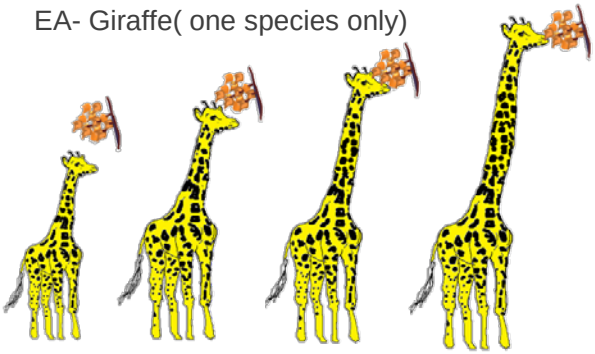
EA- Giraffe(one species only)



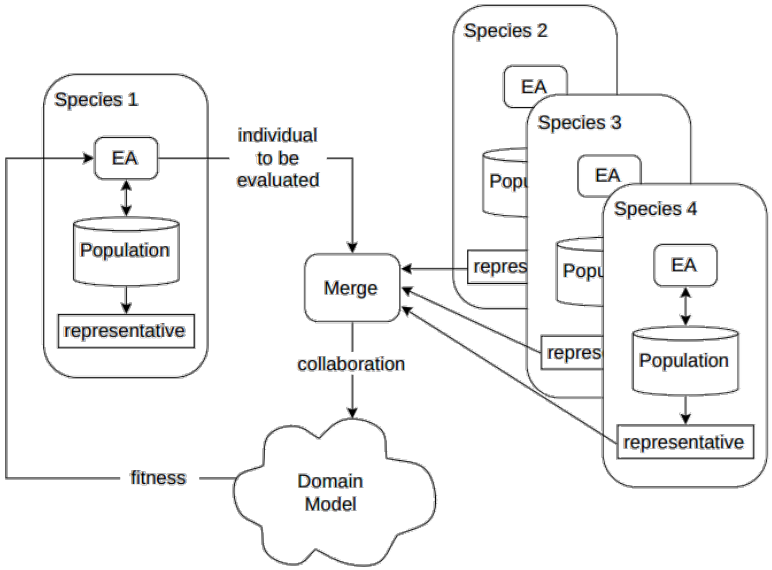


Figure 1: Cooperative coevolutionary architecture from the perspective of species number one.

Child making in evolutionary algorithm is so obvious, but what about coevolutionary algorithm?

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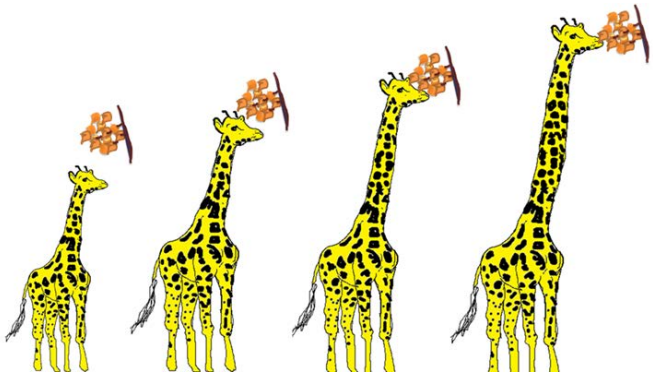
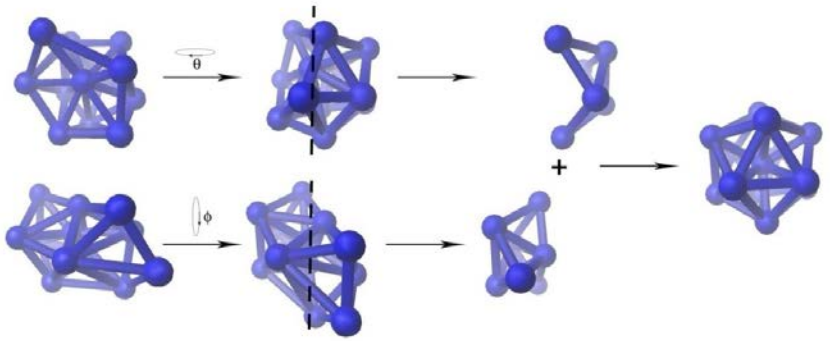


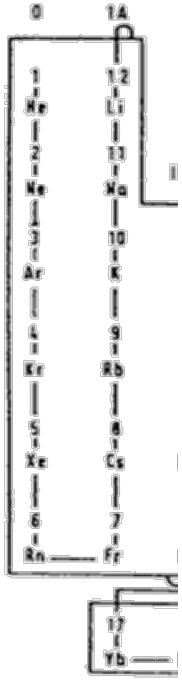


What is the child of

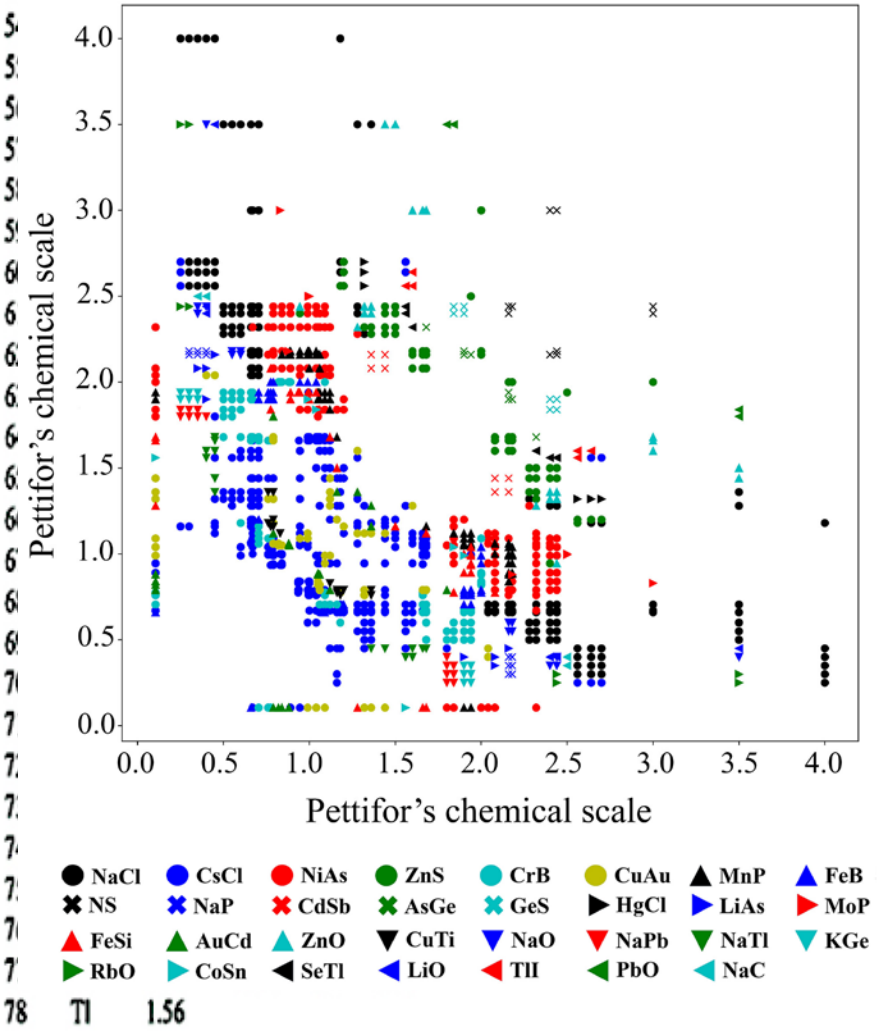
C-Fe & P-W

?

Chemical scale suggested by Pettifor (1984)



1	He	0.00	27	Gd	0.69	5
2	Ne	0.04	28	Sm	0.6925	5
3	Ar	0.08	29	Pm	0.695	5
4	Kr	0.12	30	Nd	0.6975	5
5	Xe	0.16	31	Pr	0.70	5
6	Rn	0.20	32	Ce	0.7025	5
7	Fr	0.23	33	La	0.705	5
8	Cs	0.25	34	Lw	0.7075	6
9	Rb	0.30	35	No	0.71	6
10	K	0.35	36	Md	0.725	6
11	Na	0.40	37	Fm	0.715	6
12	Li	0.45	38	Es	0.7175	6
13	Ra	0.48	39	Cf	0.72	6
14	Ba	0.50	40	Bk	0.7225	6
15	Sr	0.55	41	Cm	0.725	6
16	Ca	0.60	42	Am	0.7275	6
17	Yb	0.645	43	Pu	0.73	6
18	Eu	0.655	44	Np	0.7325	7
19	Sc	0.66	45	U	0.735	7
20	Lu	0.67	46	Pa	0.7375	7
21	Tm	0.675	47	Th	0.74	7
22	Er	0.6775	48	Ac	0.7425	7
23	Ho	0.68	49	Zr	0.76	7
24	Dy	0.6825	50	Hf	0.775	7
25	Y	0.685	51	Ti	0.79	7
26	Tb	0.6875	52	Ta	0.82	78



- *Solid. State. Commun.*, **51**, 31-34 (1984).
- *Journal of Physics C*: Solid State Physics, **19**, 3 (1986).
- *New J. Phys.*, **18**, 093011 (2016).
- *J. Alloys. Compd.*, **317**, 26-38 (2001).

- 1929 - Goldschmidt's law of crystal chemistry: the crystal structure is determined by stoichiometry, **atomic size**, and **polarizability** of atoms/ions [1].

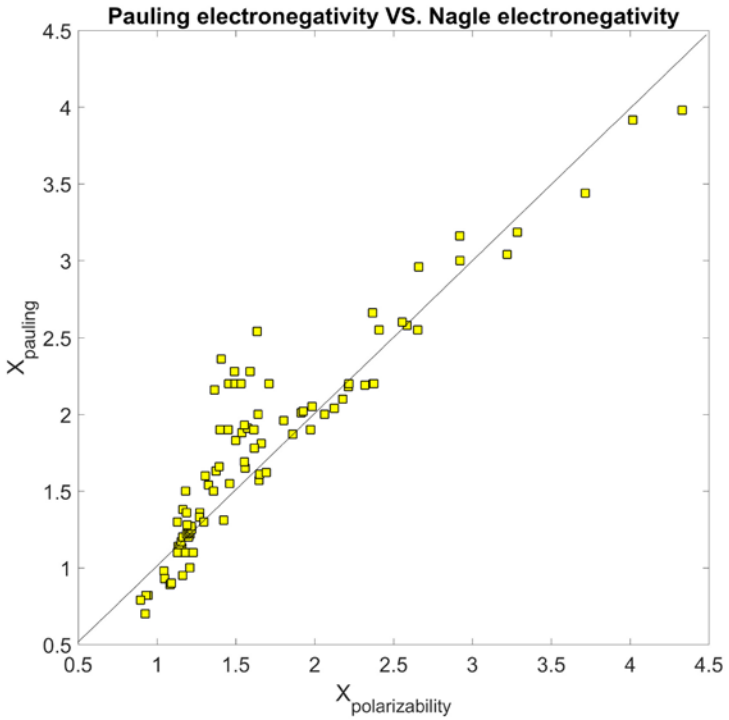
- 1932 - Linus Pauling introduced the concept of **electronegativity** [2].

- 1955 - Ringwood modification: **electronegativity** is as another important parameter for determining the crystal structure [3]. Organizing the chemica

- 1990 - Nagle: **electronegativity** and **polarizability** are strongly correlated [4].

$$\chi_\alpha = 1.66(n/\alpha)^{1/3} + 0.37$$

χ_α = electronegativity, α = polarizability & n = valence.



[1] Trans. Faraday Soc. 25, 253 (1929).
[2] J. Am. Chem. Soc. 54, 3570–3582 (1932)
[3] Geochim. Cosmochim. Acta 7, 189–202 (1955).
[4] Solid State Commun. 51, 31–34 (1984).

Our redefined MN, and it's comparison with the Pettifor's MN.

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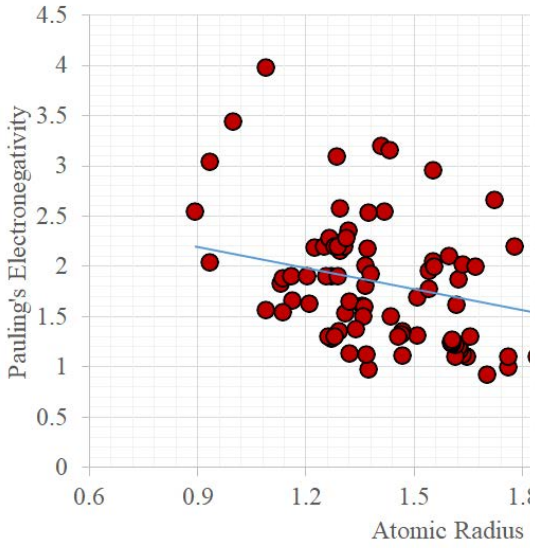

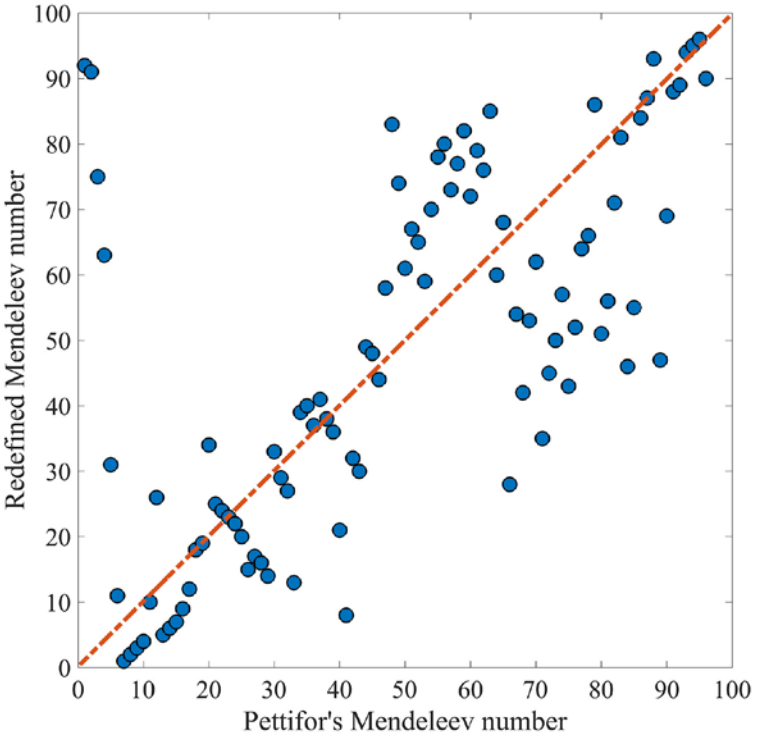

Pettifor maps of Lyakhov-Oganov model of hardness for different MNs

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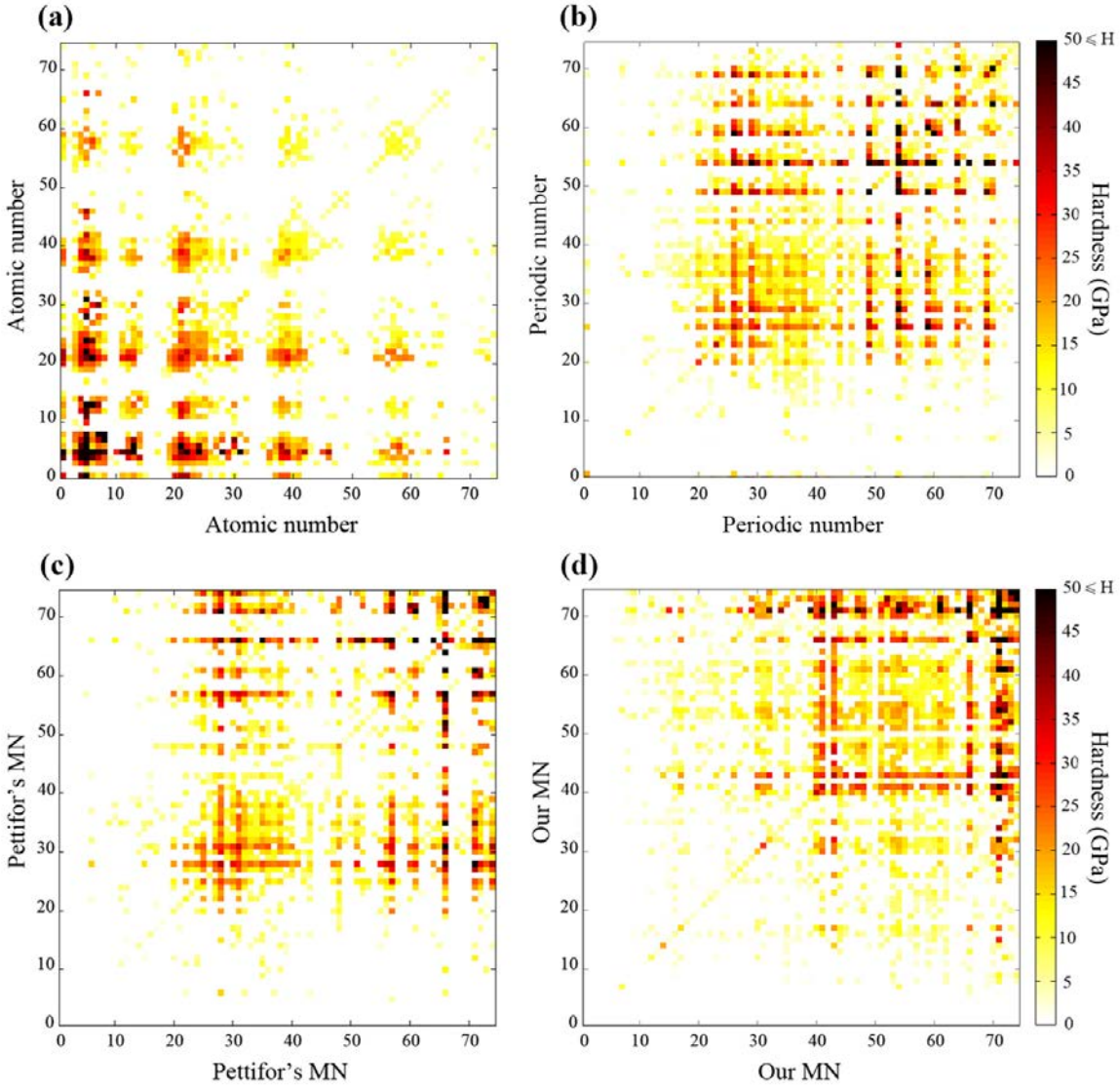

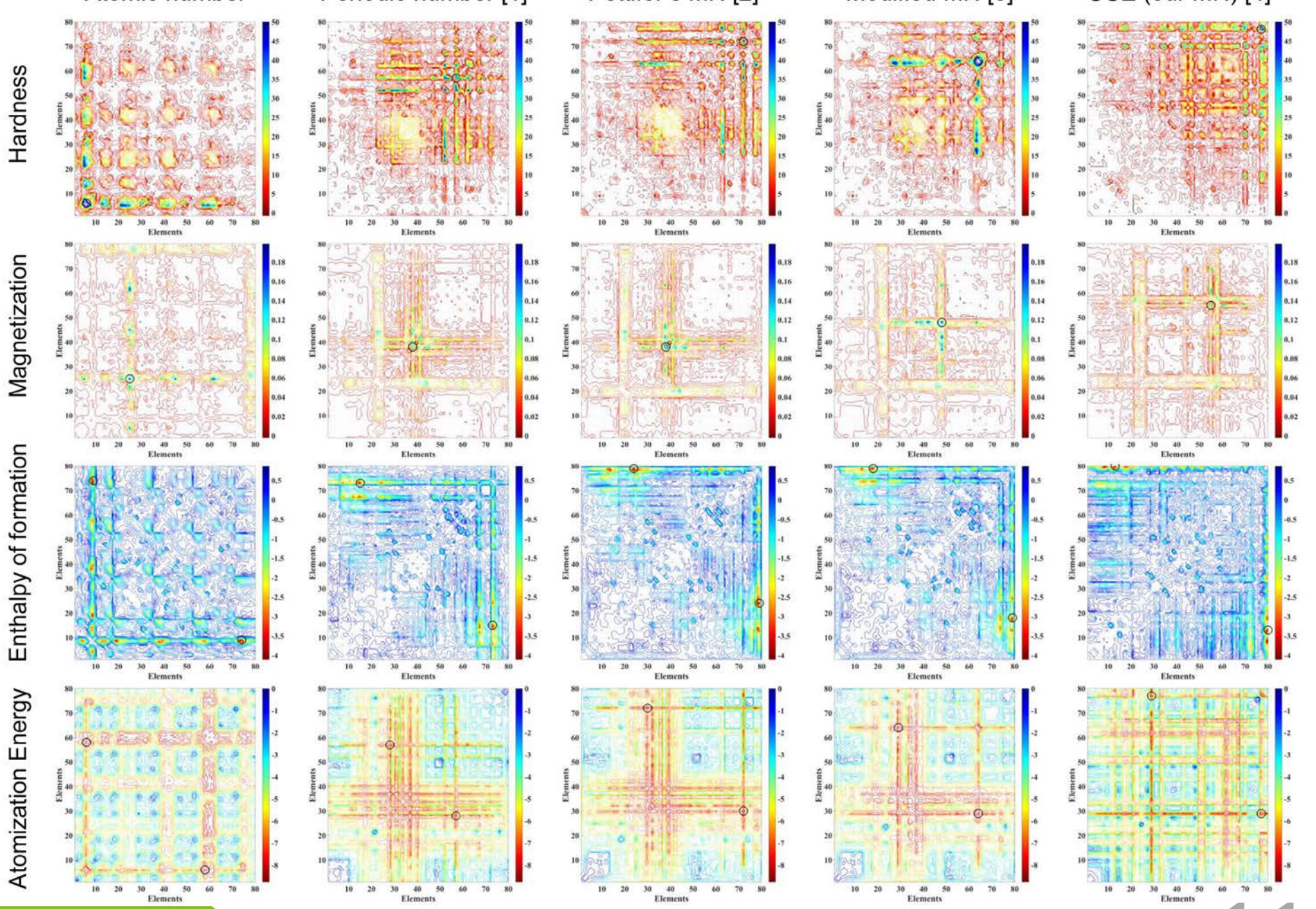

[1] *Chem. Met. Alloy*. **1**, 1-23 (2008).
[2] *J. Phys. C Solid State Phys.* **19**, 285 – 313 (1986).
[3] *New J. Phys.* **18**, 093011 (2016).
[4] *J. Phys. Chem. C,124*, 43, 23867–23878 (2020).

Clustering using density peaks

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Science, **344**, 1492 – 1496 (2014).

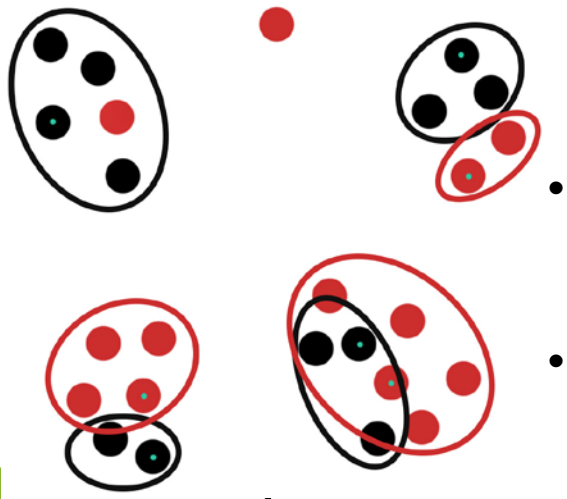
Local density(ρ_i) $\rho_i = \sum_j \chi(dij - dc)$

d_c = cutoff distance
$\chi(x) = 0 \ \ if \ x < 0$
$\chi(x) = 1 \ \ if \ x > 0$

Cluster centers are points with highest local density.

d_p = property difference cutoff between a cluster member and the cluster center.

Ideal MN = an imaginary MN, that covers all systems in minimum number of clusters (N_{min}).



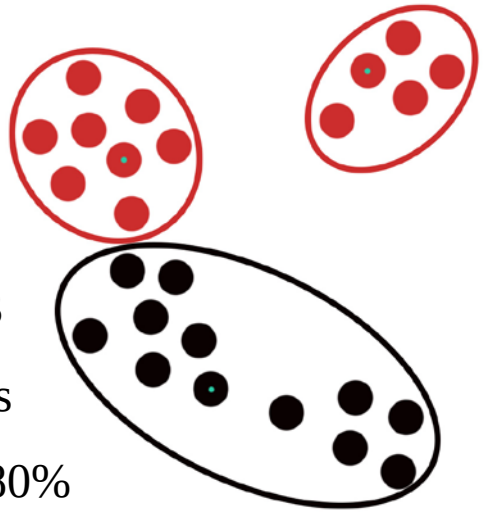
$N_{min} = 2$

- Number of clusters to cover all systems

  Example 1 = 9 Example 2 = 3

- Covered systems by biggest N_{min} (2) clusters

  Example 1 = 36% Example 2 = 80%



1

2

Evaluation of the MNs using clustering method

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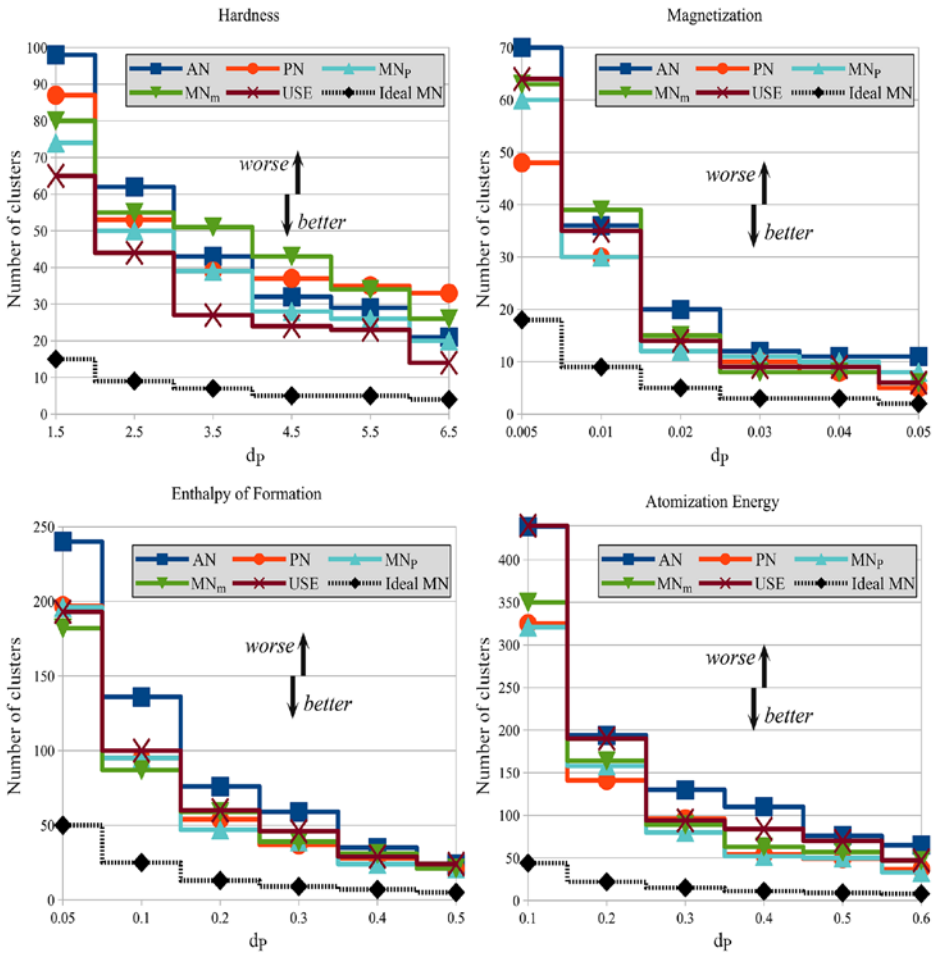
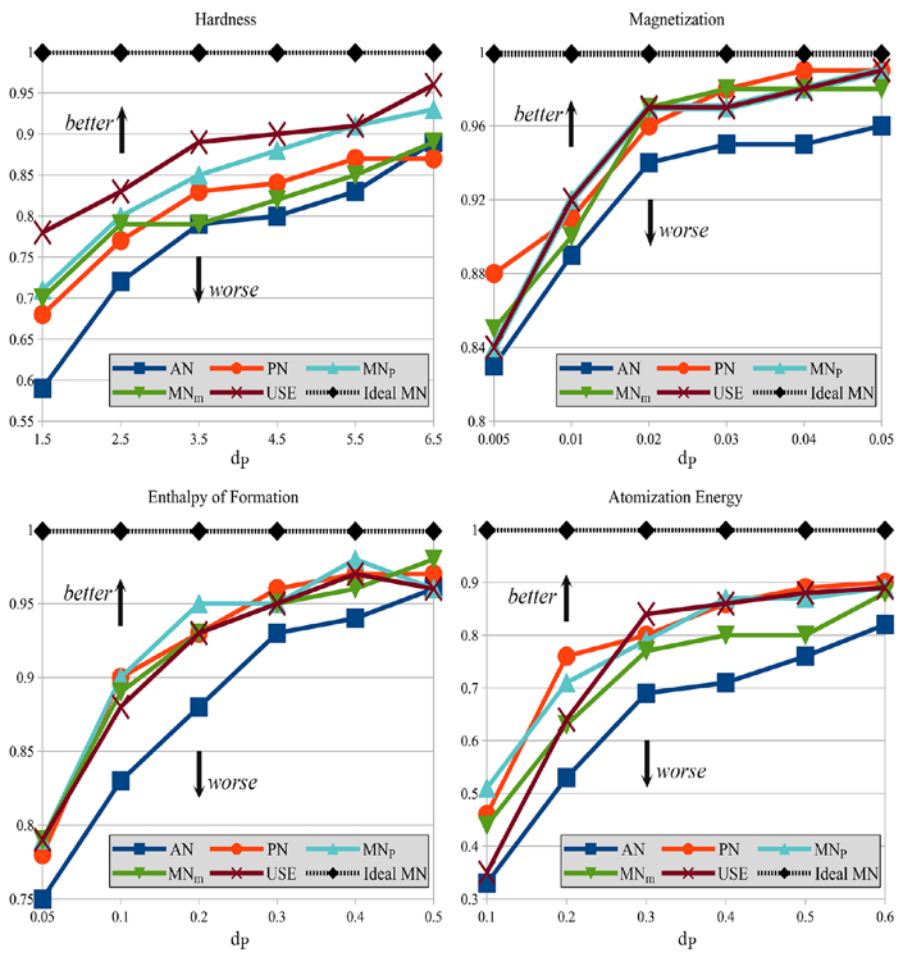
Number of clusters



Fraction of systems that are covered by N_{min} number of clusters



AN: atomic number ; PN: Periodic number ; MN_P: Pettifor's MN ; MN_m: modified MN ; USE: MN in this work.

MendS algorithm and its important Variation operators

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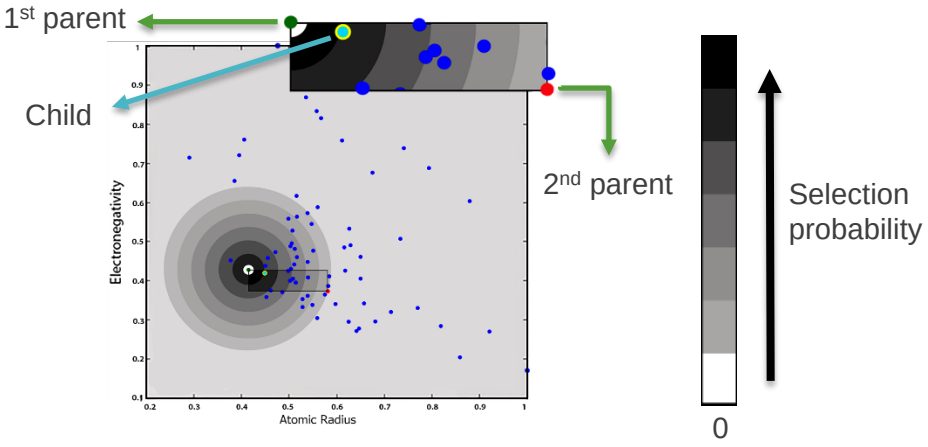
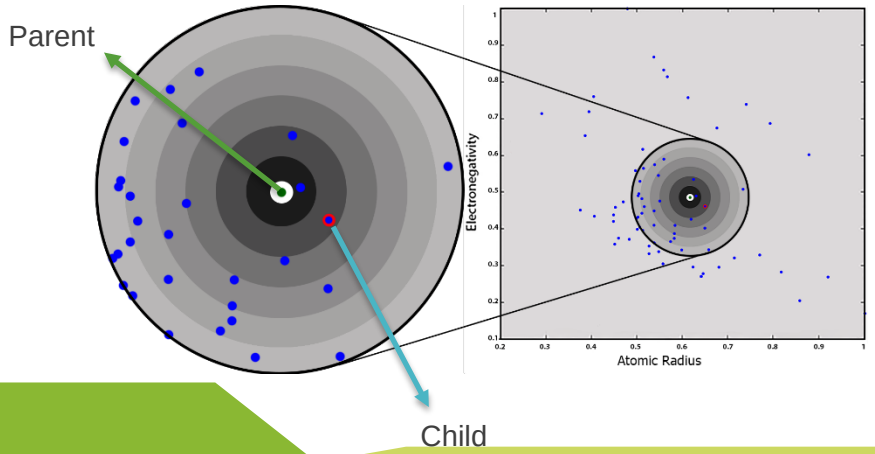
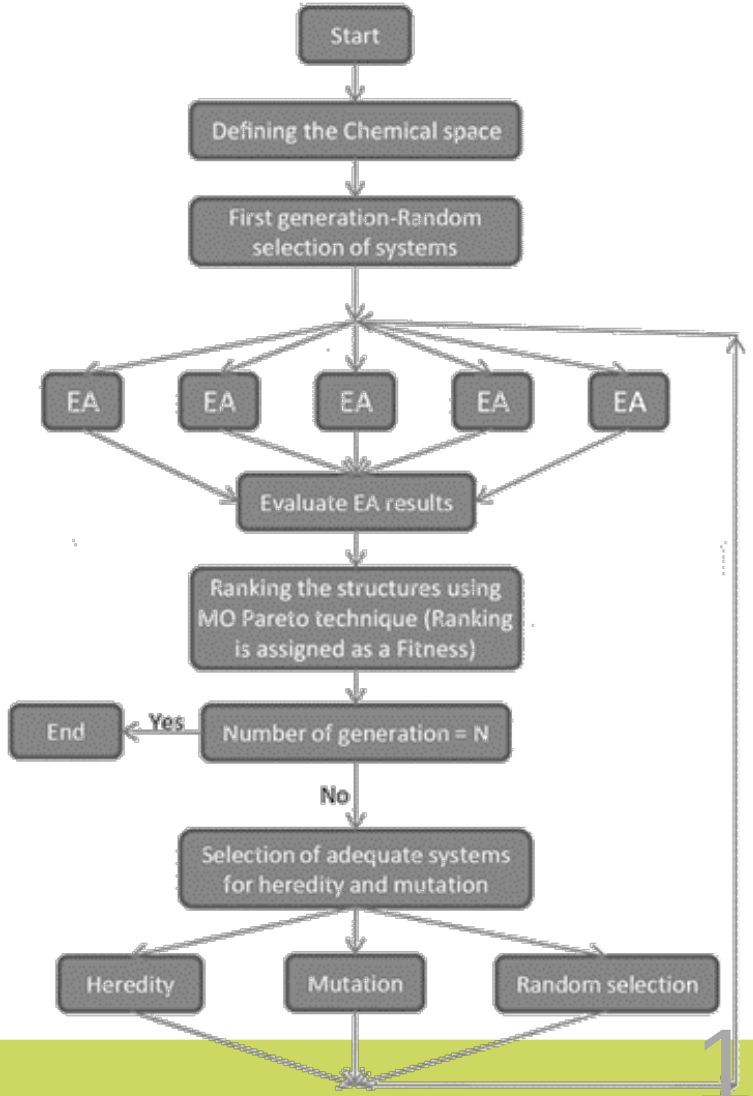
Finding the child of two parent red and green using **chemical heredity operator.**



Using **chemical mutation operator** we mutate an atom to find



Reactive heredity operator:
A-B & C-D ➔ A-C , A-D, B-C or B-D



Implementation of multi-objective Pareto method and its test on Mo_xN_y

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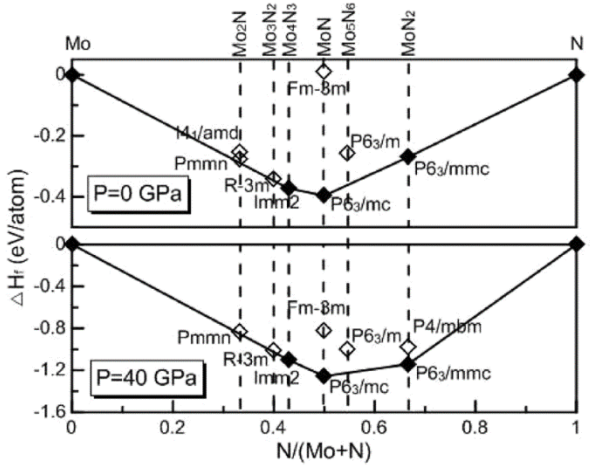
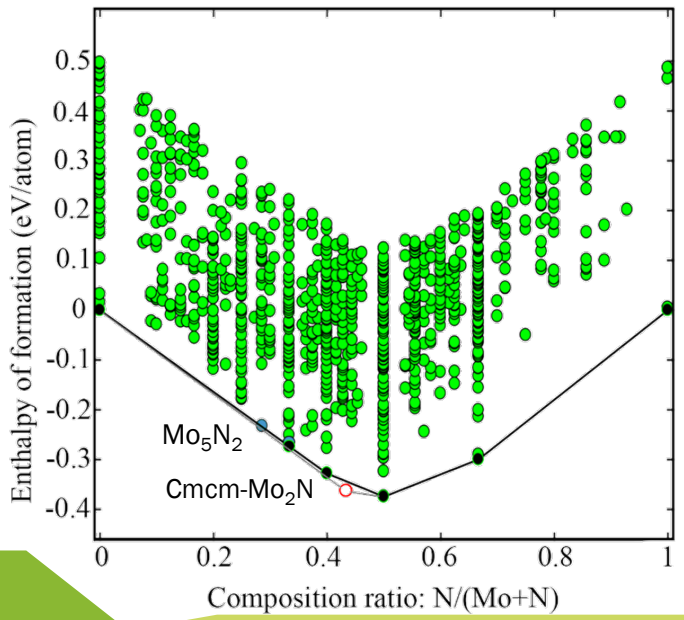
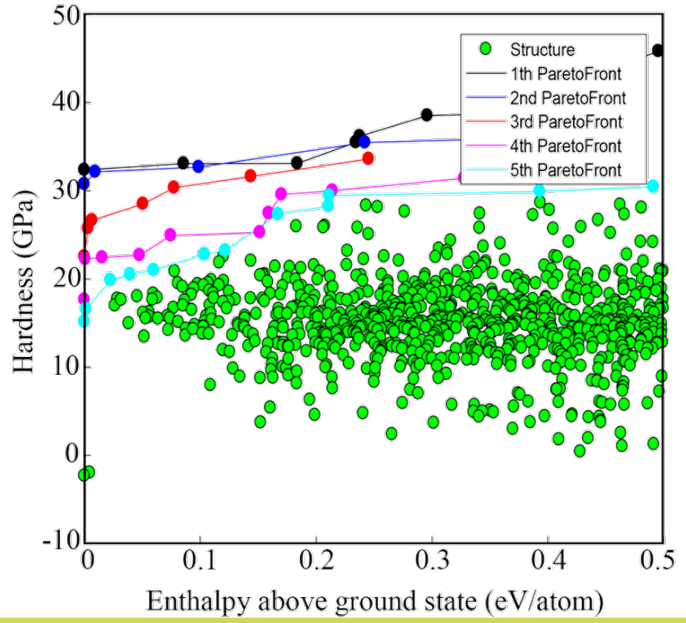
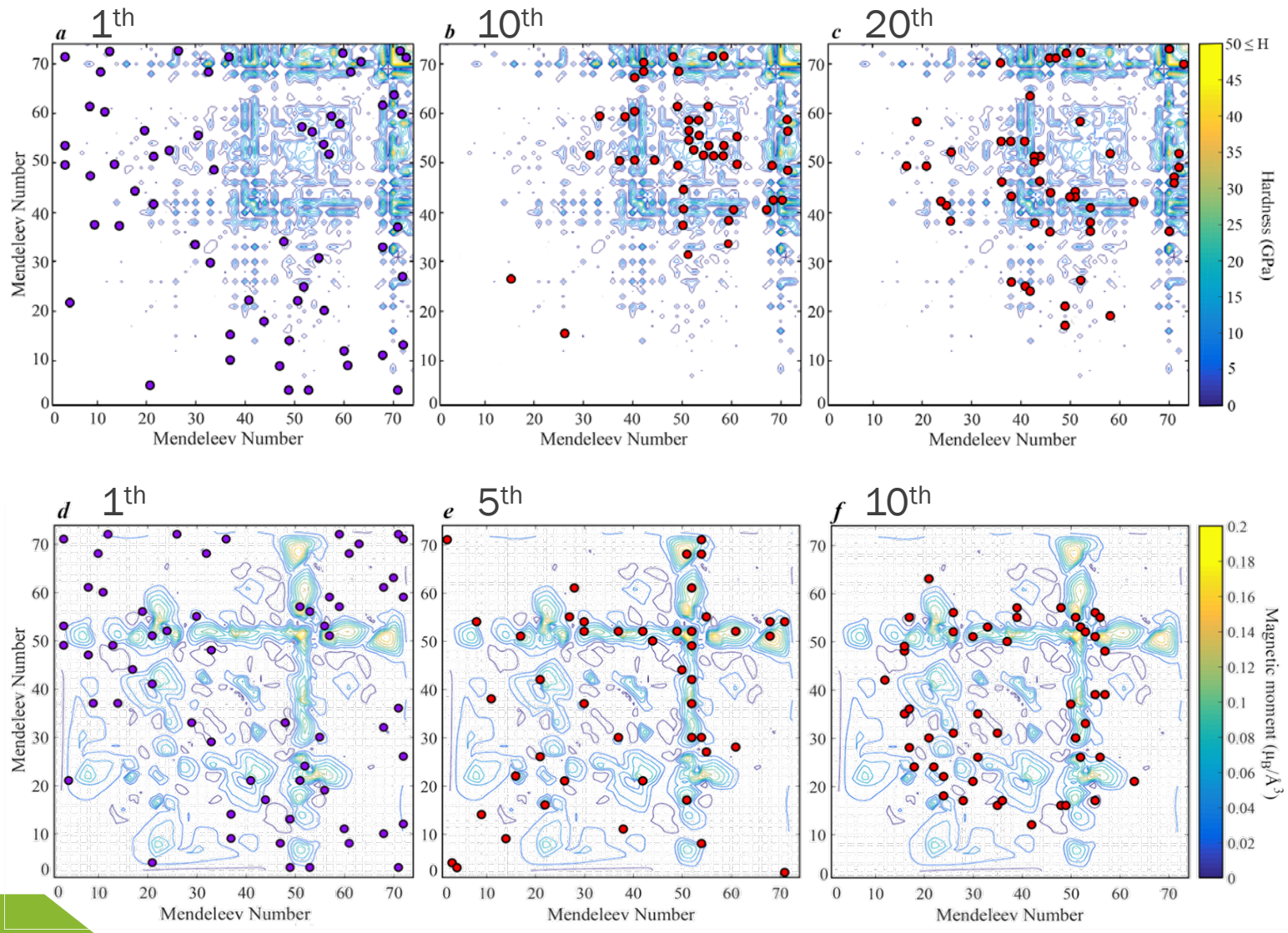


J. Phys. Chem. C, *120* (20), pp 11060–11067 **(2016)**.






a 1th
b 10th
c 20th
d 1th
e 5th
f 10th
Mendeleev Number
Hardness (GPa)
50 ≤ H
Magnetic moment (μB/Å3)

Efficiency of the algorithm in the system selection – improving the Pareto front

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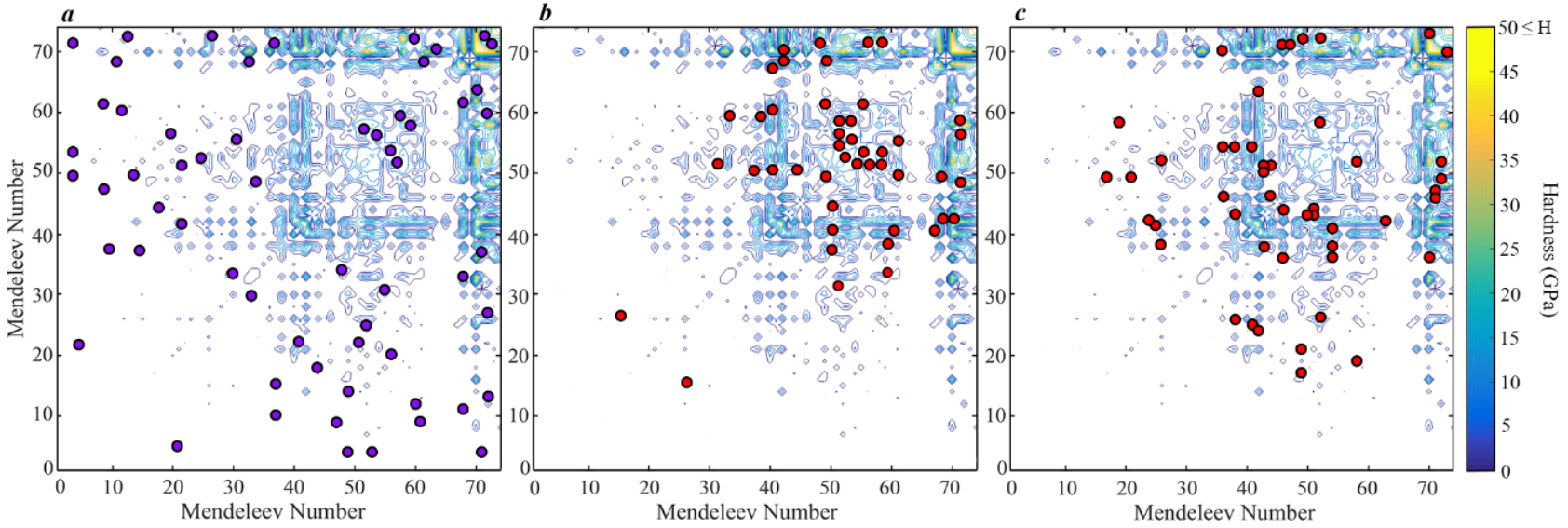

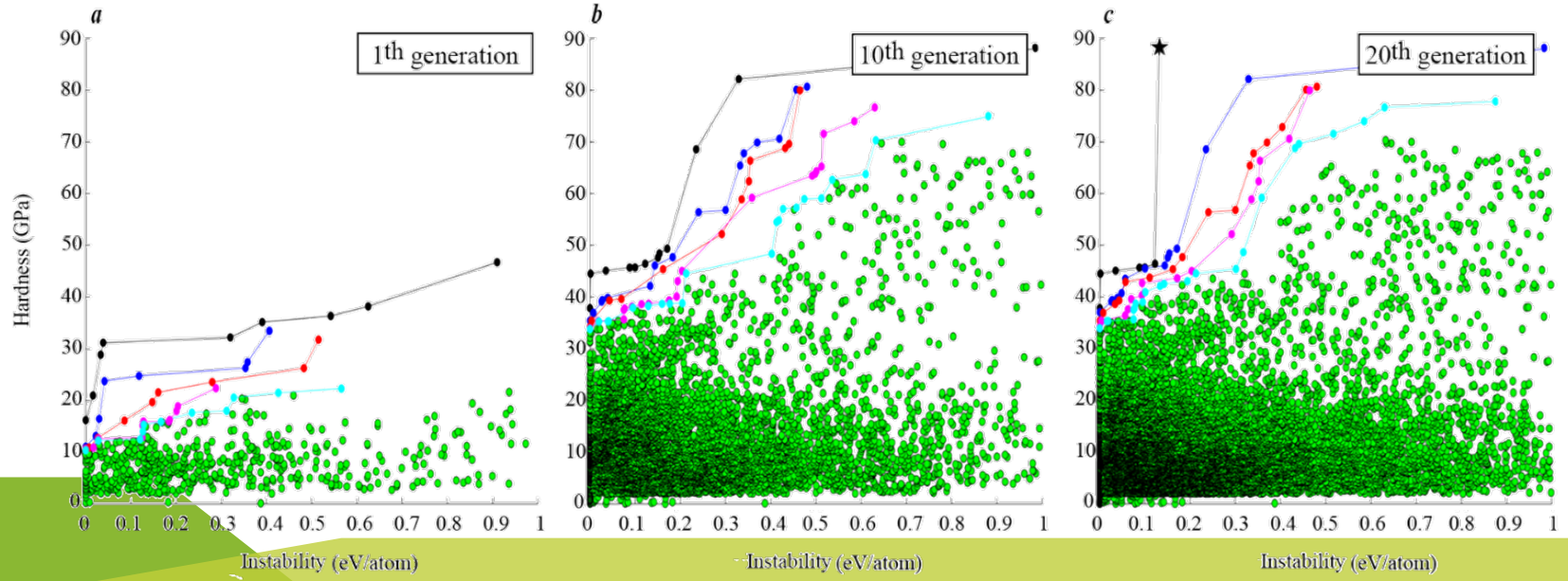

	Compounds	H_v (GPa)	K_{1C} (MPa.m$^{1/2}$)	Instability (eV/atom)	Space group		Compounds	H_v (GPa)	K_{1C} (MPa.m$^{1/2}$)	Instability (eV/atom)	Space group
Carbon	C	**92.7**	6.33	0.13	$Fd\bar{3}m$	**Boron**	B	38.9	2.87	0	$R\bar{3}m$
	C	**93.6**	6.36	0.139	$P6_3/mmc$		B	**44.8**	3.29	0.136	$Cmc2_1$
B-S	B_4S_3	30.5	1.83	0.102	$Cmcm$	**B-N**	BN	**63.4**,(**62.8**)[25],[**46-80**][20]	5.1	0.075	$F\bar{4}3m$
	MoB_2	28.5,(33.1)[44],[24.2][53]	3.76	0	$R\bar{3}m$		TcB	31,(30.3)[54]	3.83	0.013	$P\bar{3}m1$
	MoB_3	35.3	3.74	0.035	$P\bar{3}m1$		TcB_3	27.2,(29)[55]	3.6	0	$P\bar{6}m2$
	MoB_3	32.2	3.63	0.077	$A2/m$		TcB_3	33.1	3.79	0.003	$P\bar{3}m1$
		35.3,(37.3)[44]	3.63	0.017	$P6_3/mmc$		TcB_4	31.8	3.56	0.069	$P2_1/m$
		33.1,(31.8)[43]	3.57	0.011	$R\bar{3}m$		TcB_4	30.2	3.54	0.069	$R\bar{3}m$
Mo-B	MoB_4	35.4	3.57	0.099	$Pmmn$	**Tc-B**		30,(32)[55]	3.57	0.027	$P6_3/mmc$
	MoB_5	35.7	3.62	0.054	$P\bar{6}m2$		TcB_7	35.9	3.35	0.084	$R3m$
	MoB_8	36.6	3.24	0.118	$R3m$		TcB_8	33.9	3.3	0.113	$R3m$
	Mo_2B_3	32.2	3.95	0.029	$Imm2$		Tc_3B_5	30.6	3.87	0	$P\bar{6}m2$
	Mo_2B_3	30.4	3.87	0.043	$Cmcm$						
Si-C	SiC	33.3,(33.1)[31],[28][31]	2.94	0	$F\bar{4}3m$	**B-P**	BP	37.2,(29.3)[31],[33][31]	2.46	0	$F\bar{4}3m$
	SiC	33.1	2.94	0.001	$R3m$		B_6P	**41.1**	2.87	0	$R\bar{3}m$
	VB	39.1,(38.3)[49]	3.66	0	$Cmcm$		MnH	29.5	3.2	0	$P6_3/mmc$
	VB_2	37.3,(39.5)[49],[27.2][50]	3.75	0	$P6/mmm$		MnH	27.9	3.14	0.013	$R\bar{3}m$
	VB_5	**40**	3.36	0.158	$P\bar{6}m2$		MnH	26.3	3.07	0.044	$Fm\bar{3}m$
V-B	VB_7	39.7	3.19	0.143	$P\bar{3}m1$	**Mn-H**	Mn_3H_2	26.8	3.22	0.017	$R32$
	VB_{12}	**44.5**	3.34	0.125	$I4/mmm$		Mn_3H_2	27	3.26	0.019	$P6_3/mcm$
	V_3B_4	37.8	3.74	0	$P\bar{4}m2$		Mn_4H_3	27.6	3.23	0.002	$P2/m$
	V_3B_4	35.9,(38.2)[49]	3.7	0.006	$Immm$		Mn_6H_5	27.3	3.17	0.011	$A2/m$
	MnB_3	32.2	3.5	0.029	$P\bar{6}m2$		FeB_3	30.2	3.32	0	$P2_1/m$
	MnB_4†	**40.7**	3.65	0.009	$Pnnm$		FeB_4	35.7	3.06	0.021	$Immm$
Mn-B	MnB_4	38.2	3.56	0.1	$R\bar{3}m$	**Fe-B**	FeB_4‡	32	3.31	0.039	$R\bar{3}m$
		38.1,(**40.5**)[51],[37.4][52]	3.76	0	$P2_1/c$		FeB_4	**42.7**	3.31	0.063	$A2/m$
	MnB_5	32.7	3.38	0.097	$P\bar{6}m2$			28.6,(24.4)[61][**62**][59]	3.32	0.002	$Pnnm$
	MnB_{13}	**40.4**	2.9	0.181	Pm		Fe_2B_{11}	33.8	3.37	0.081	Pm

Ashby plot of hardness vs. toughness for predicted phases

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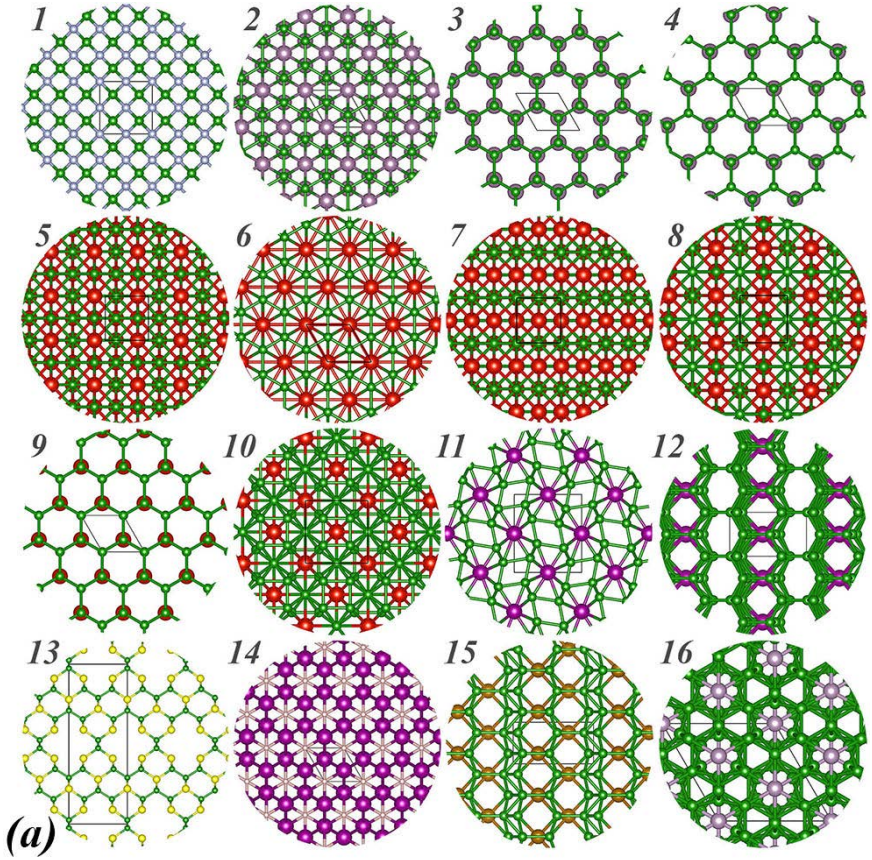
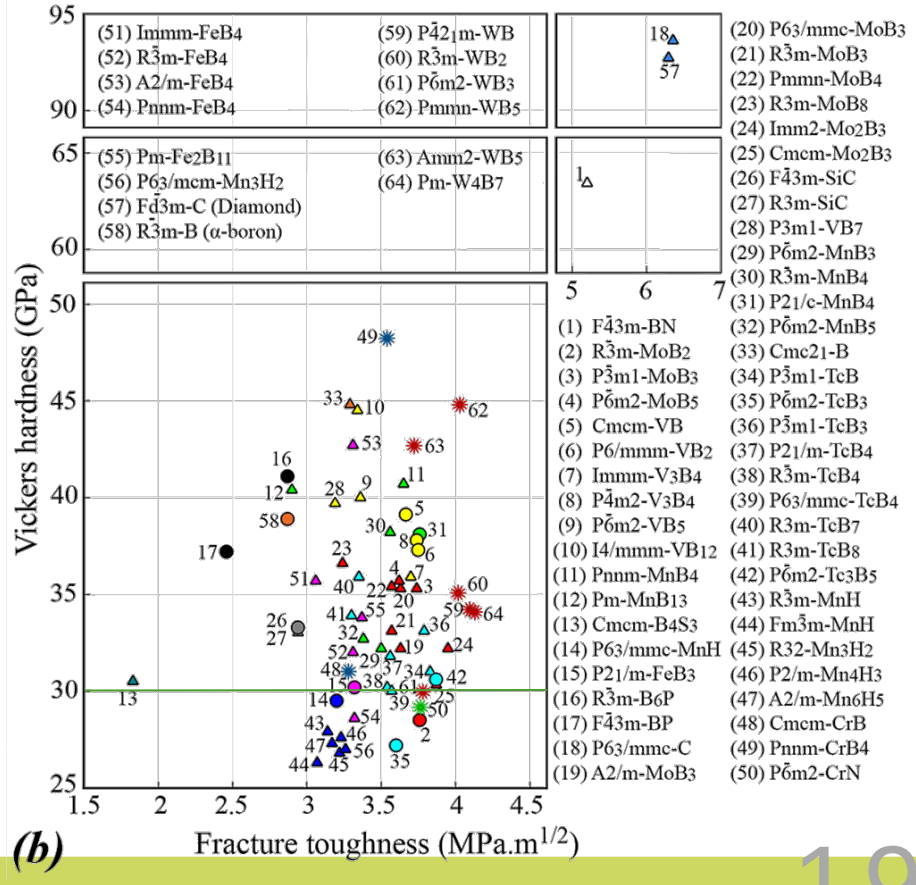
- *J. Phys. Chem. Lett.*, *8* (4), pp 755–764 **(2017)**. Cr-N, Cr-B
- *J. Phys. Chem. Lett.*, *9* (12), pp 3470–3477 **(2018)**. W-B



Tungsten carbide WC
Hardness 30 GPa



Conclusion

- A well-defined chemical space can be obtained using the most significant properties of elements (electronegativity and atomic size and binary systems with similar properties are nearby in this space.
- Multi-objective Pareto technique makes it possible to search for materials optimal in more than one property, and works efficiently at least for two properties.
- Combination of coevolutionary algorithm, MO Pareto technique and well-structured chemical space - MendS – works efficiently in searching for materials optimal in multiple target properties.
- Our Mendelevian search suggests that diamond is the hardest material (among binaries) in nature – harder cannot be found.
- MendS works at arbitrary pressure and for binary, ternary systems.

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Thank you for your attention!

Any questions?