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Mendelevian search for materials with optimal properties

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21th USPEX workshop on Modern Trends on Computational Materials Discovery.

November 21, 2022

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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).

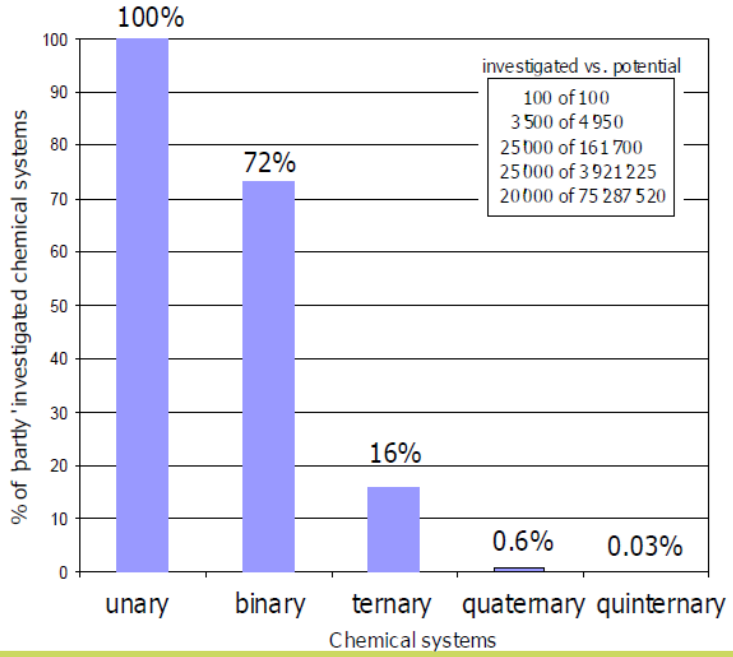
Outline

Search for materials with optimal target properties:

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

- Coevolutionary algorithm.
- A well-designed 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

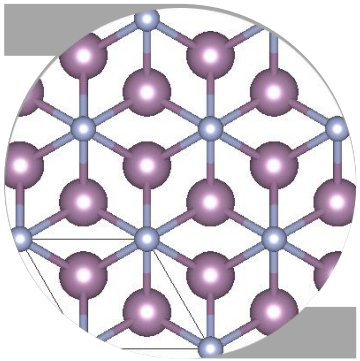
Most optimal phases and structures of X

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 what is the difference of evolutionary algorithm and coevolutionary algorithm?

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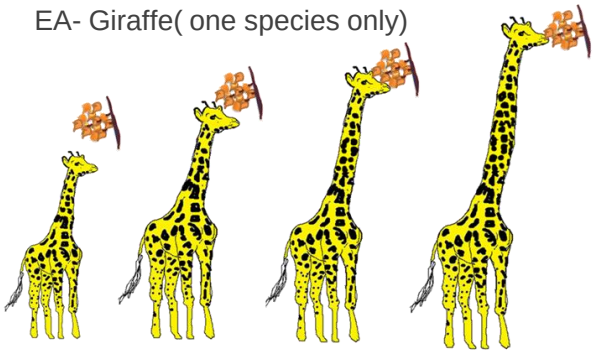
EA- Giraffe(one species only)



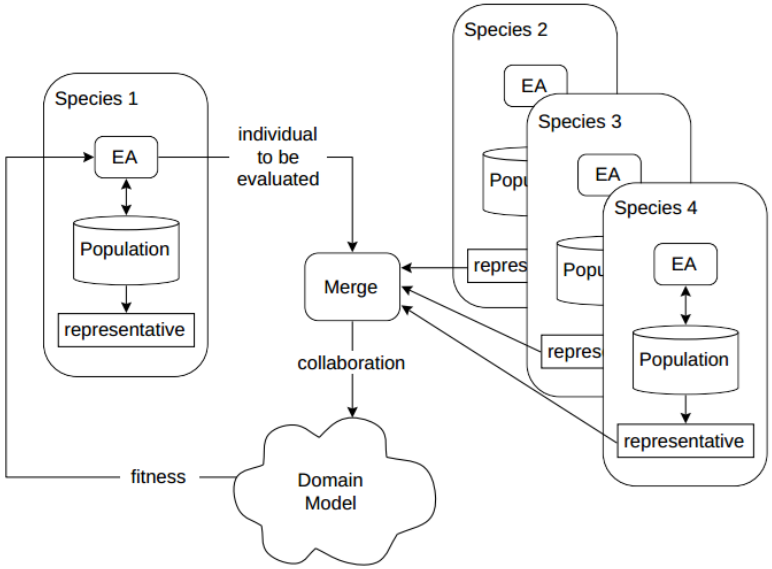


Figure 1: Cooperative coevolutionary architecture from the perspective of species number one.

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

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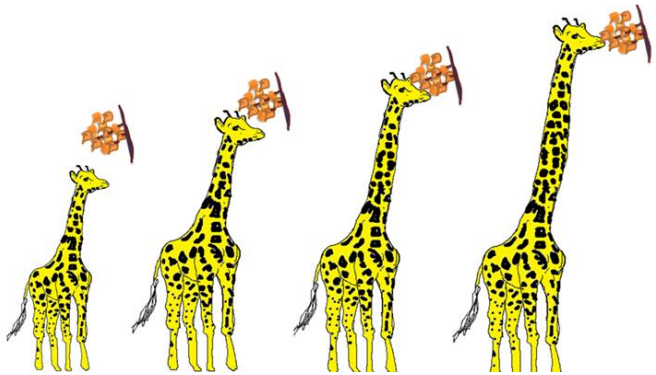
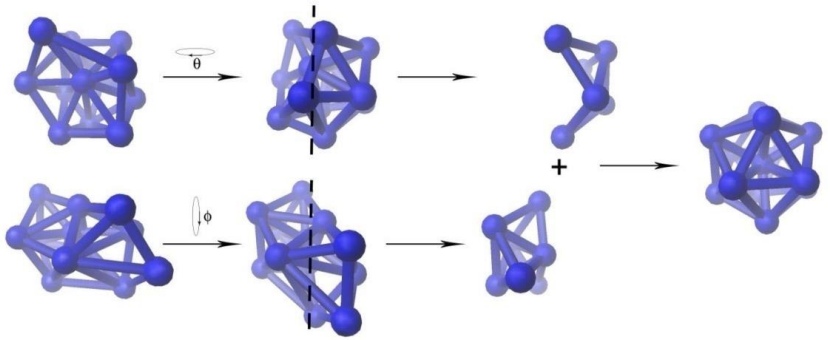


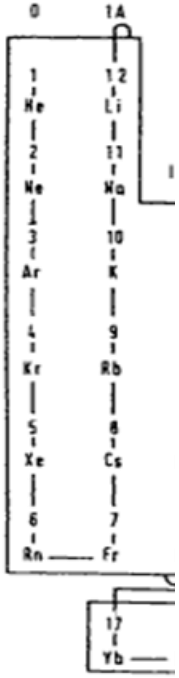


What is the child of

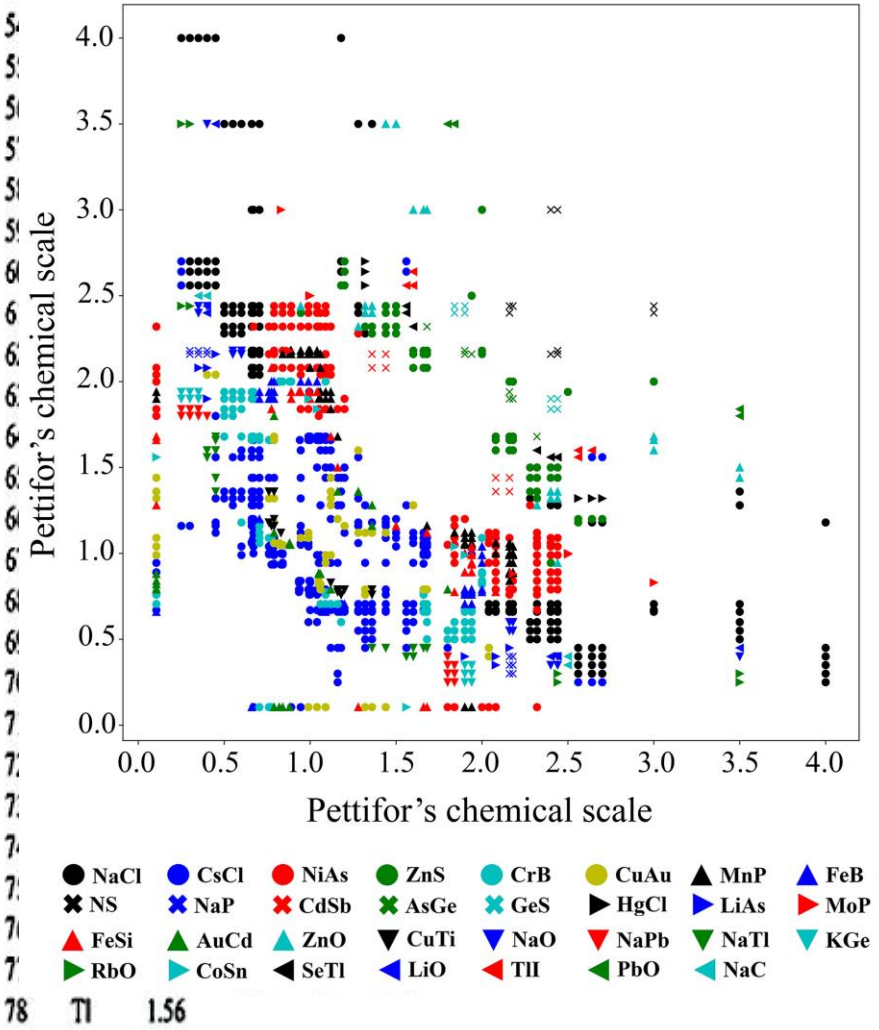
C-Fe & P-W

?

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].

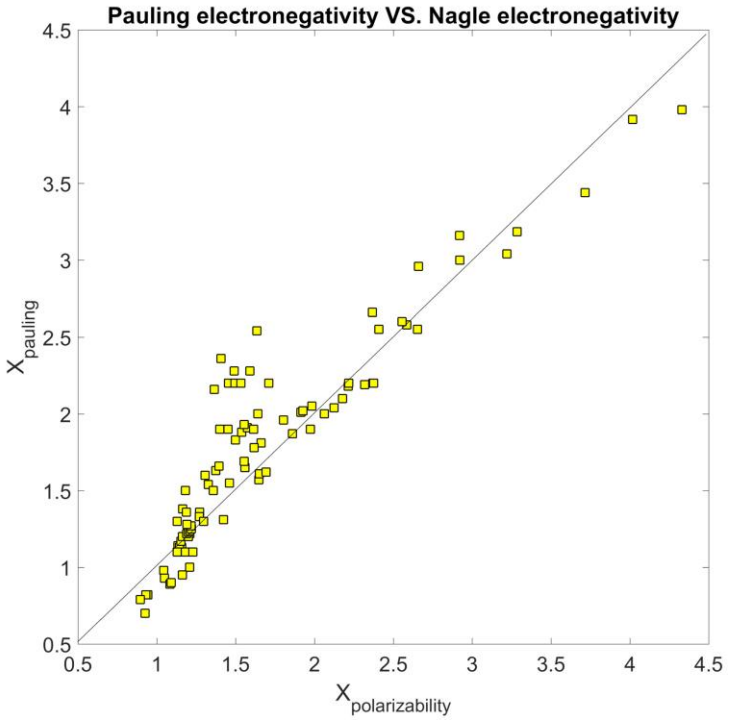
- 1955 - Ringwood modification: **electronegativity** is as another important parameter for determining the crystal structure [3].

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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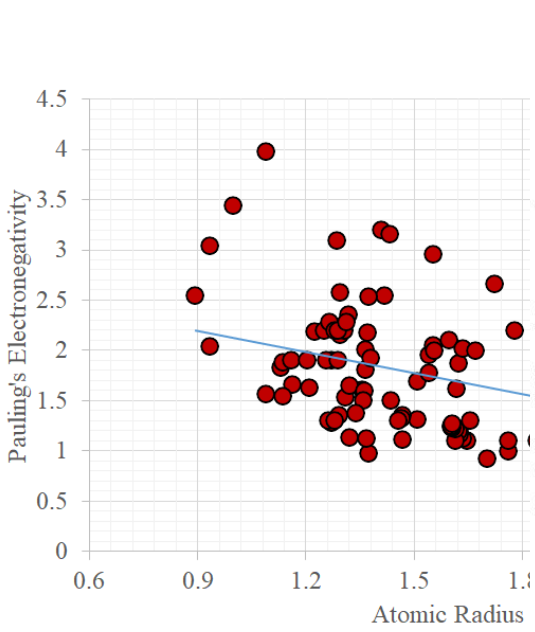

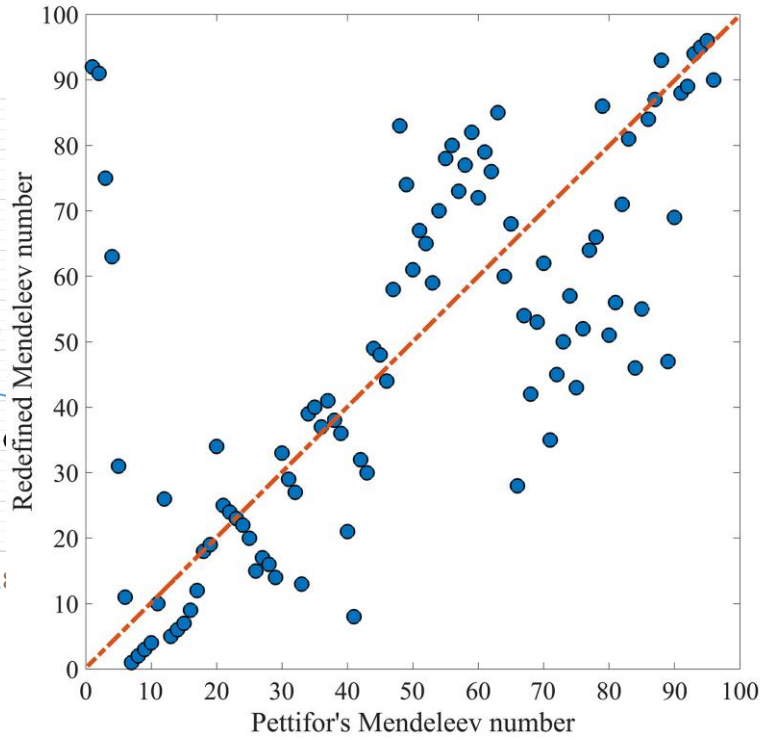

Pettifor maps of Lyakhov-Oganov model of hardness for different MNs

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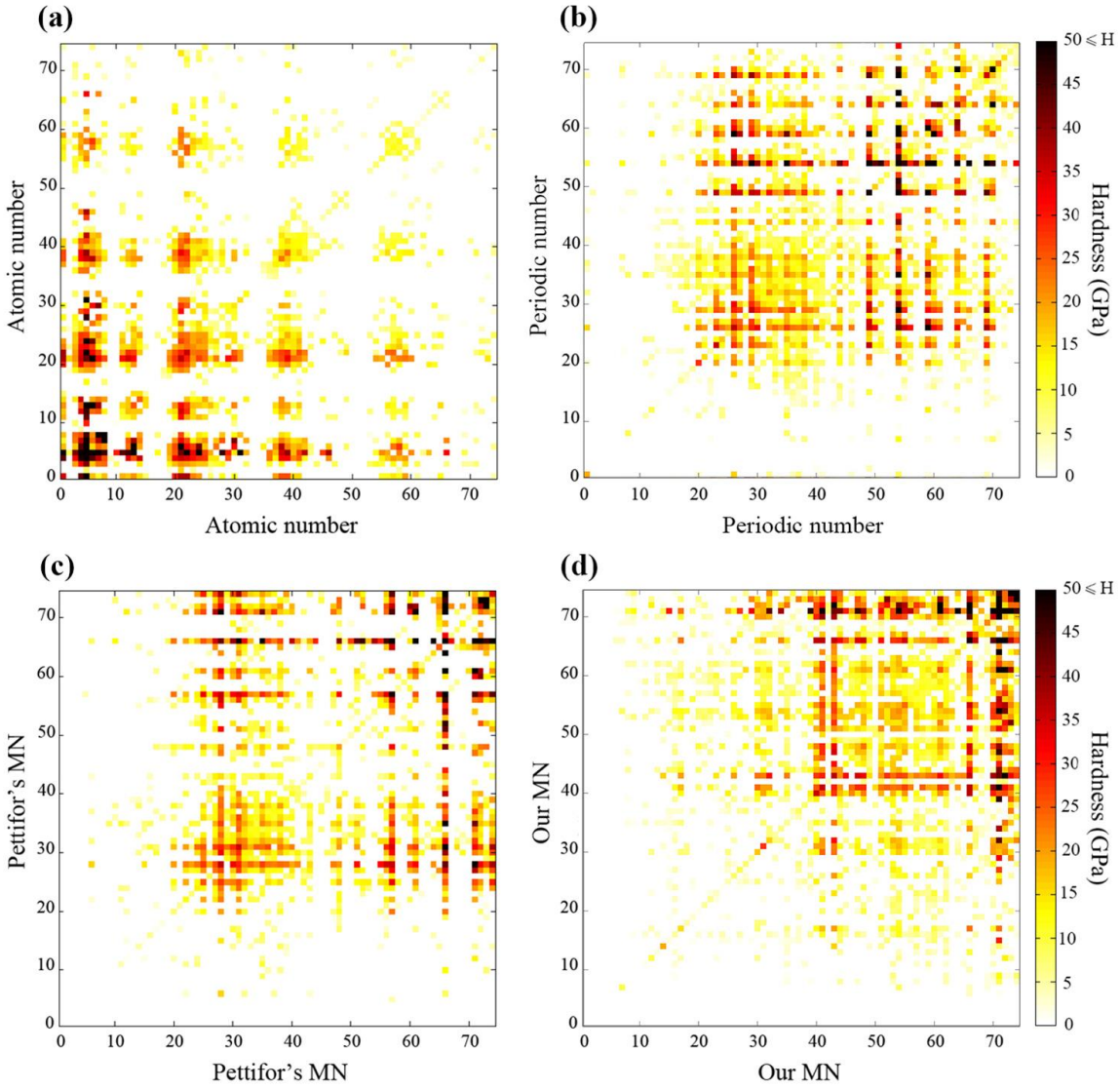

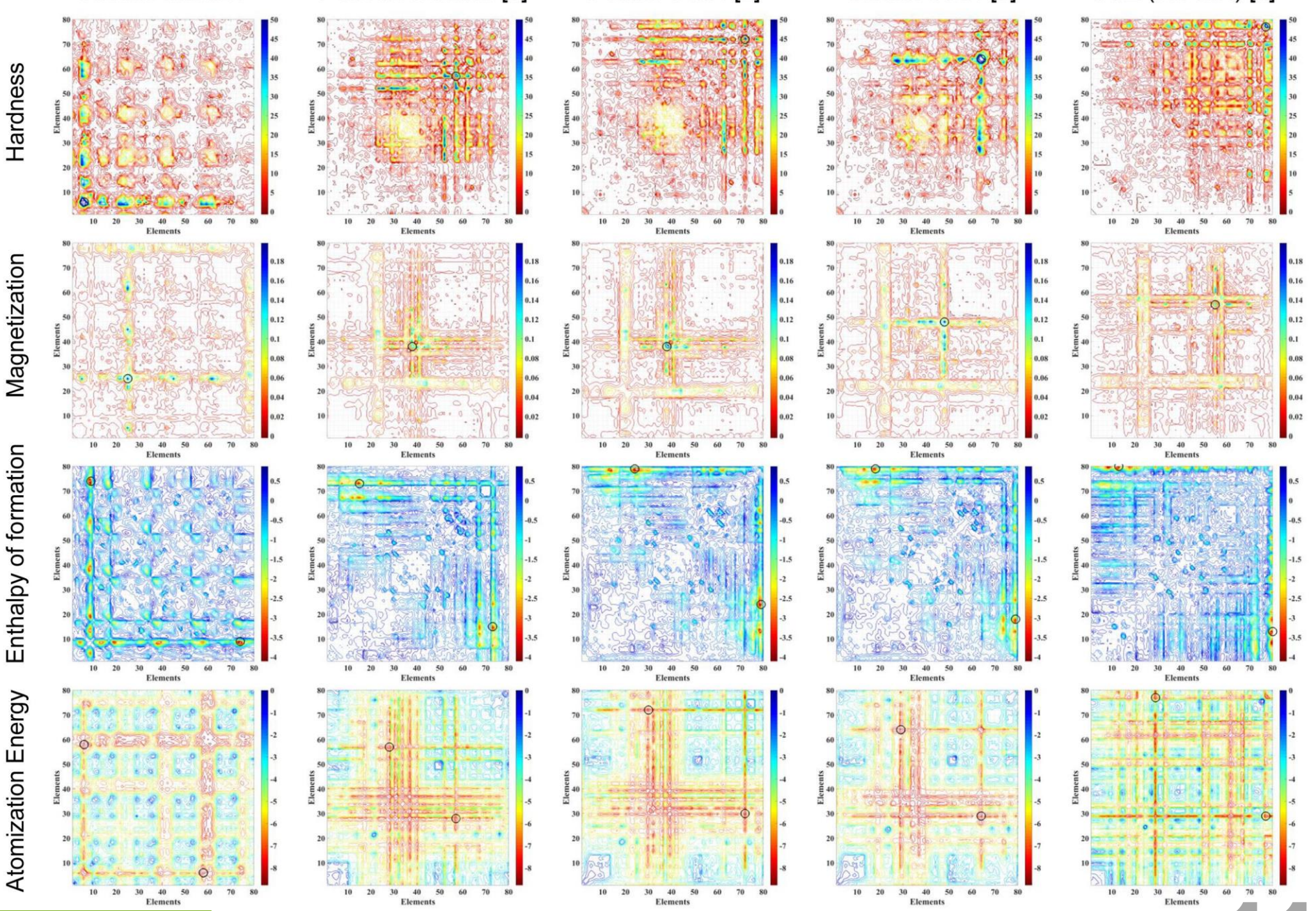

[1] *Chem. Met. Alloy*. **1**, 1-23 (2008).
[2] *J. Phys. C Solid State Phys.* **19**, 285 – 313 (1986).
[3] *New J. Phys.* **18**, 093011 (2016).
[4] *J. Phys. Chem. C,124*, 43, 23867–23878 (2020).

Clustering using density peaks

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Science, **344**, 1492 – 1496 (2014).

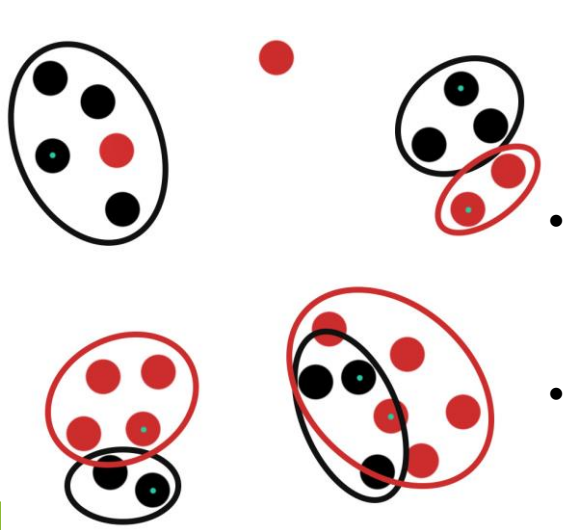
Local density(ρ_i) $\rho_i = \sum_j \chi(dij - dc)$

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}).



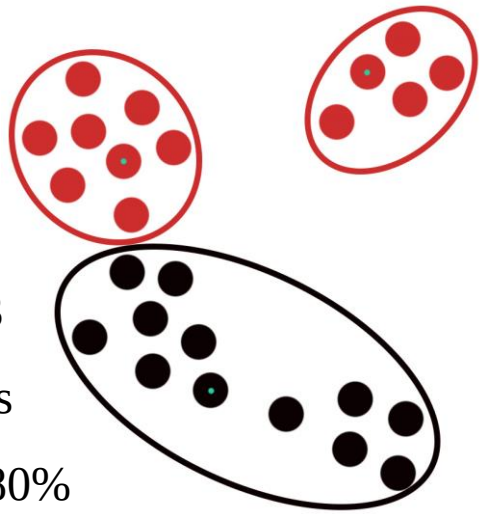
$N_{min} = 2$

- 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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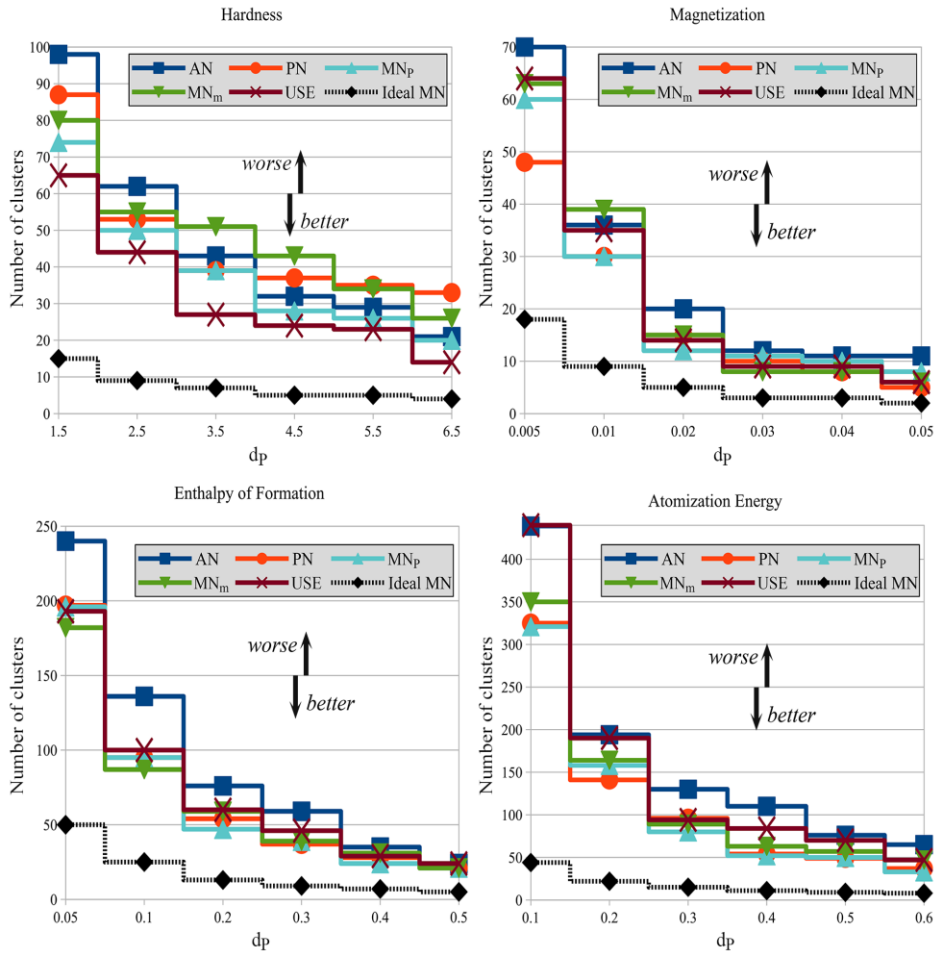
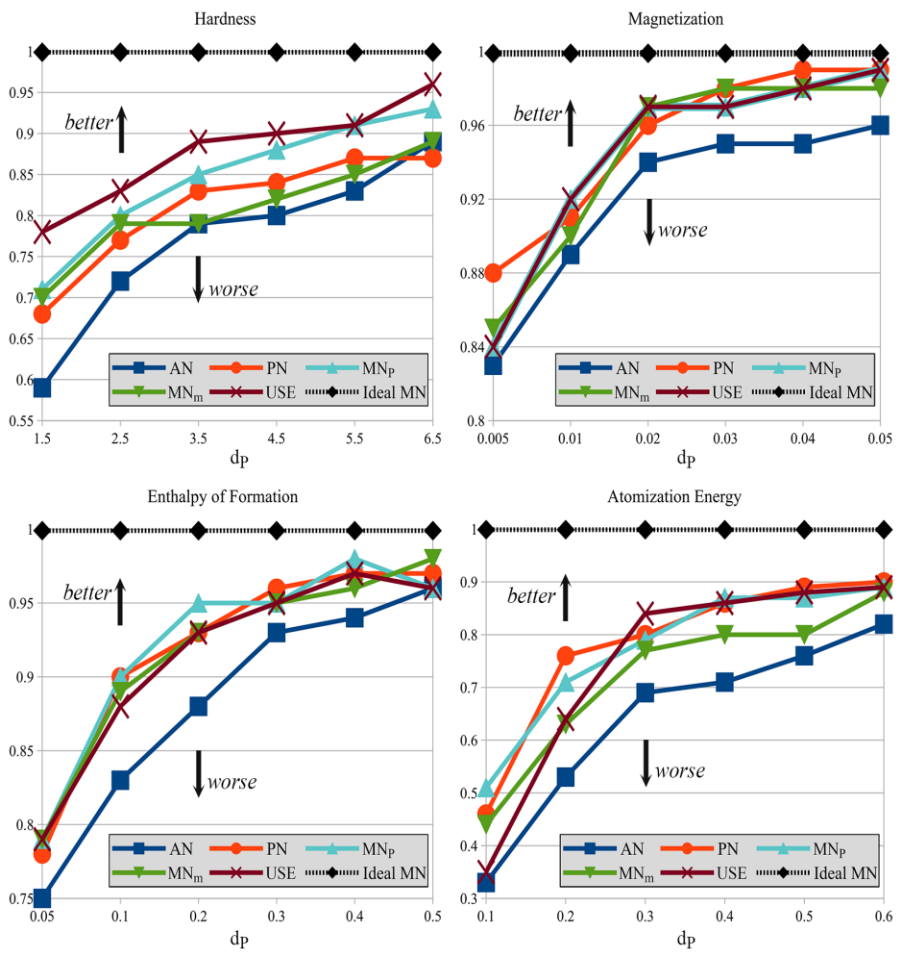
Number of clusters



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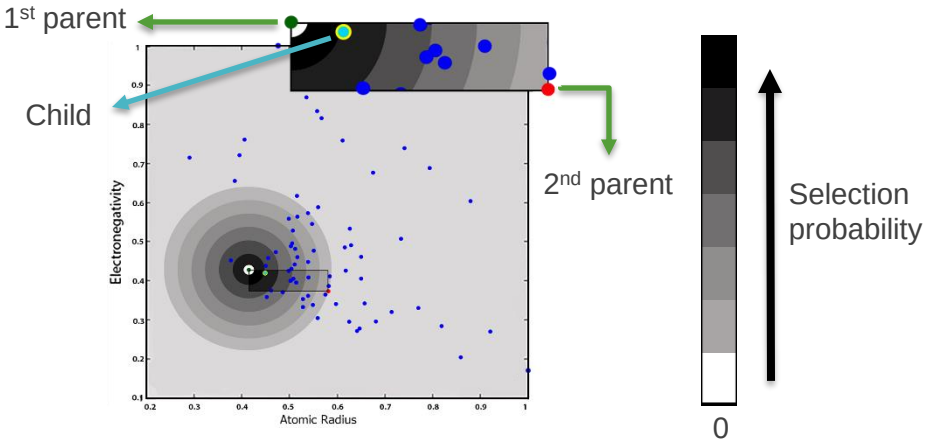
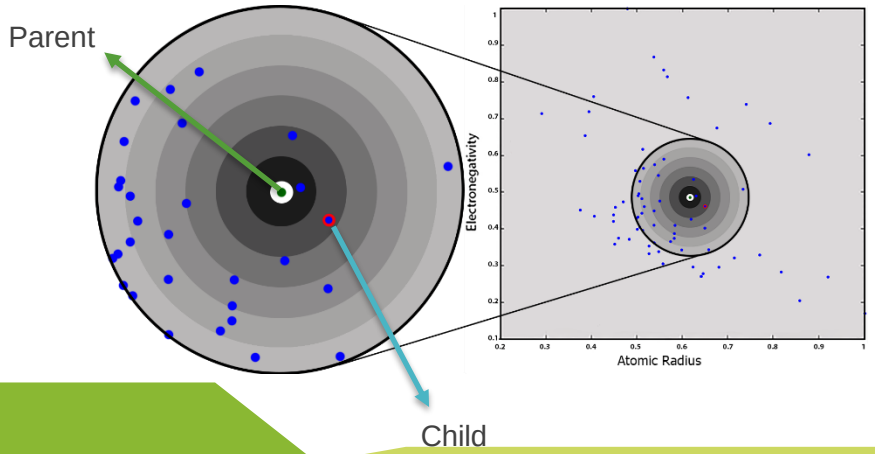
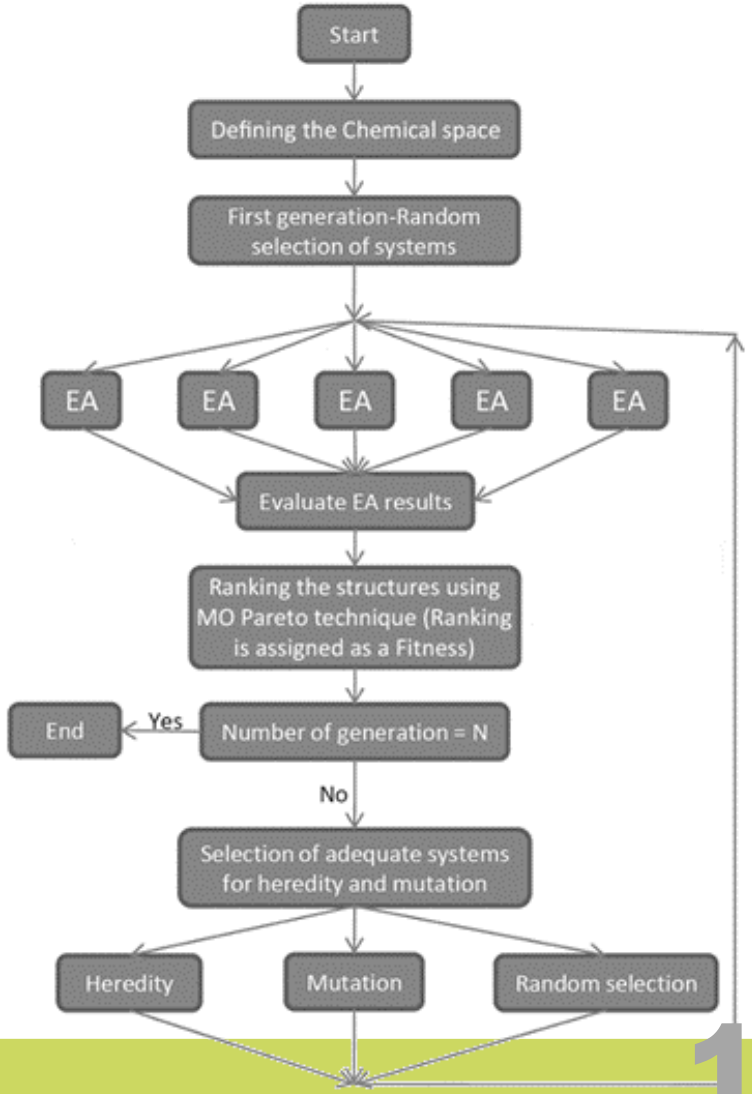
Finding the child of two parent red and green using **chemical heredity operator.**



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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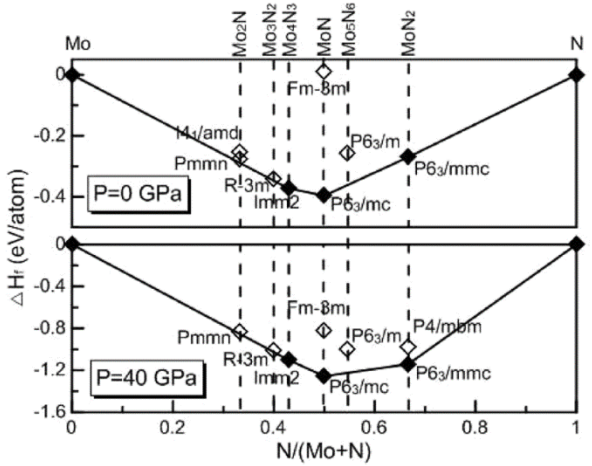
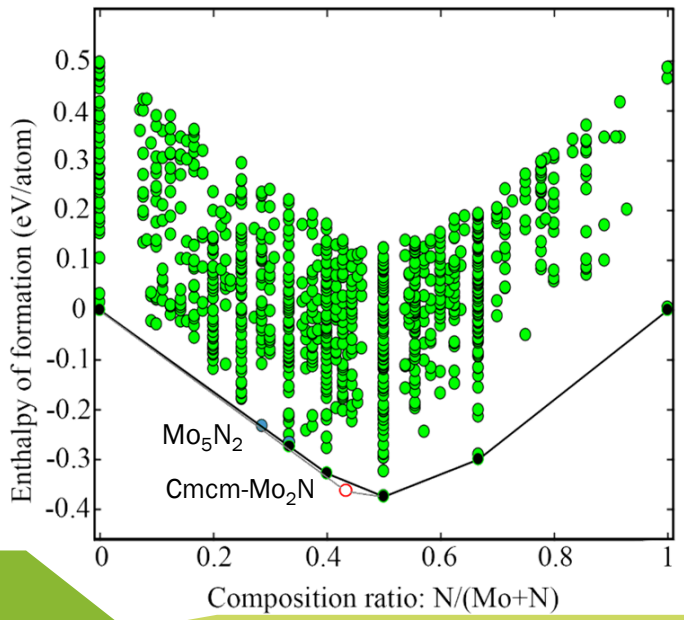
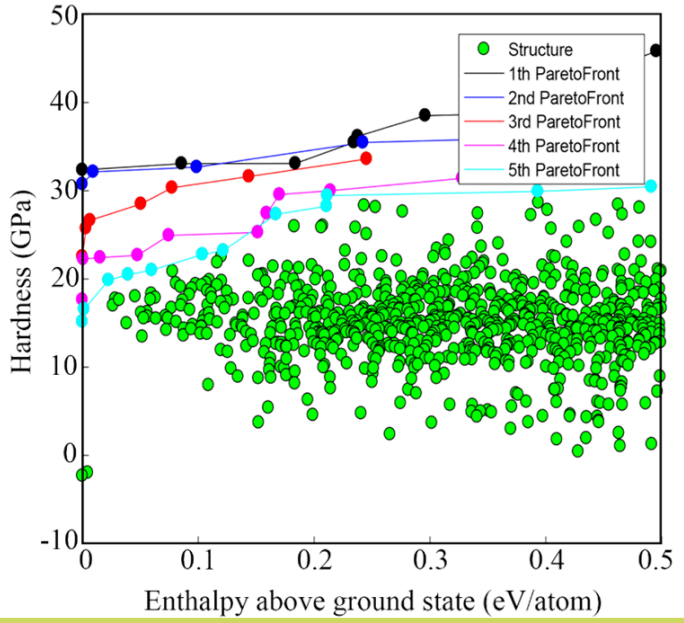
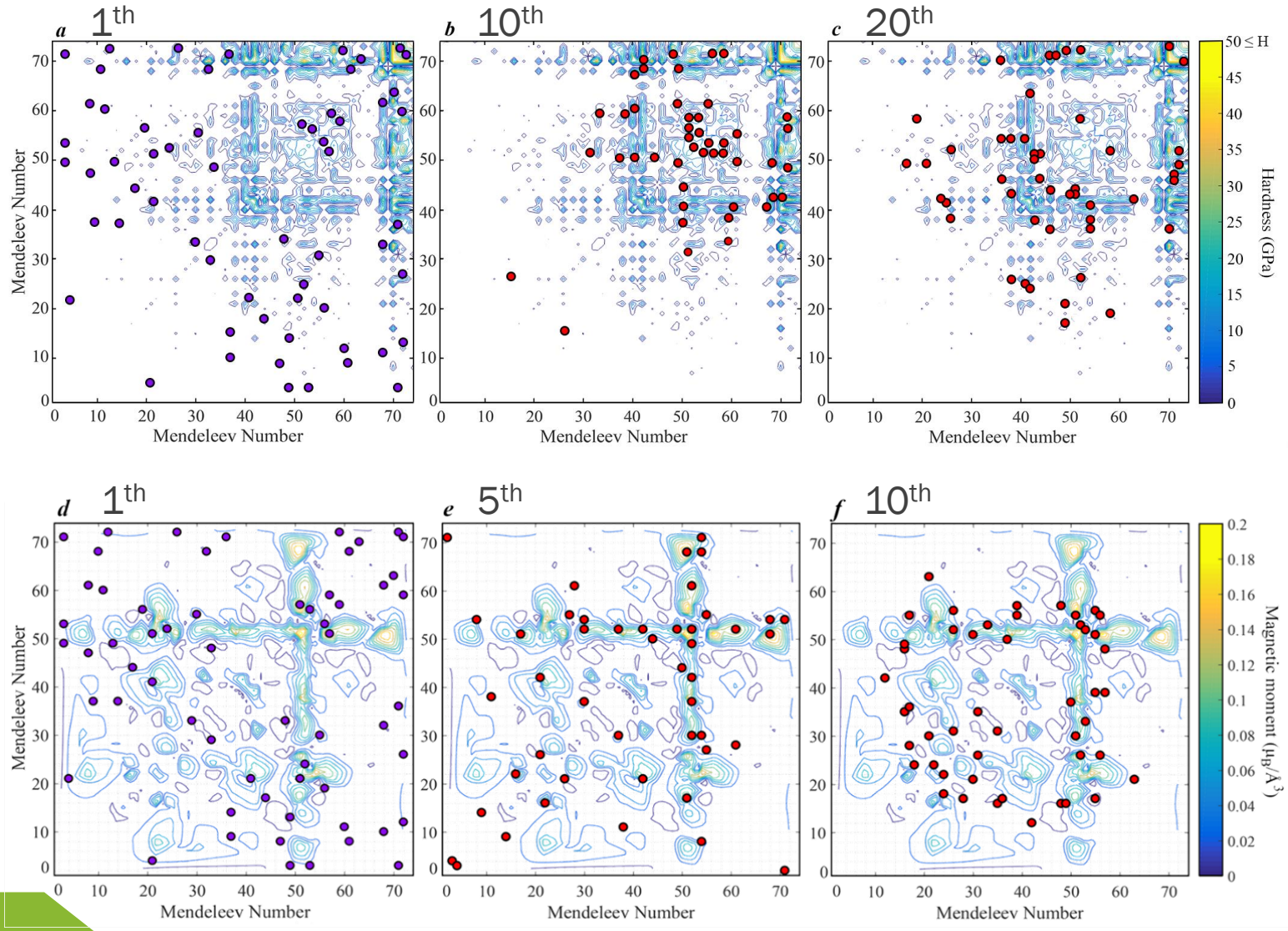


J. Phys. Chem. C, *120* (20), pp 11060–11067 **(2016)**.






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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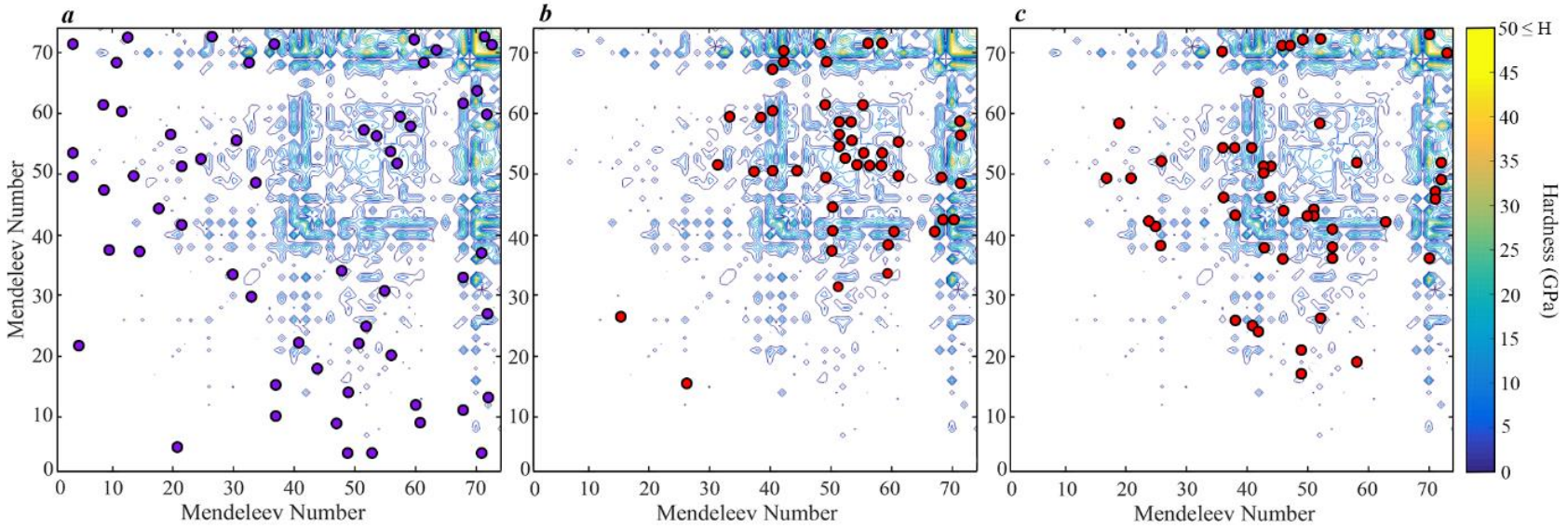

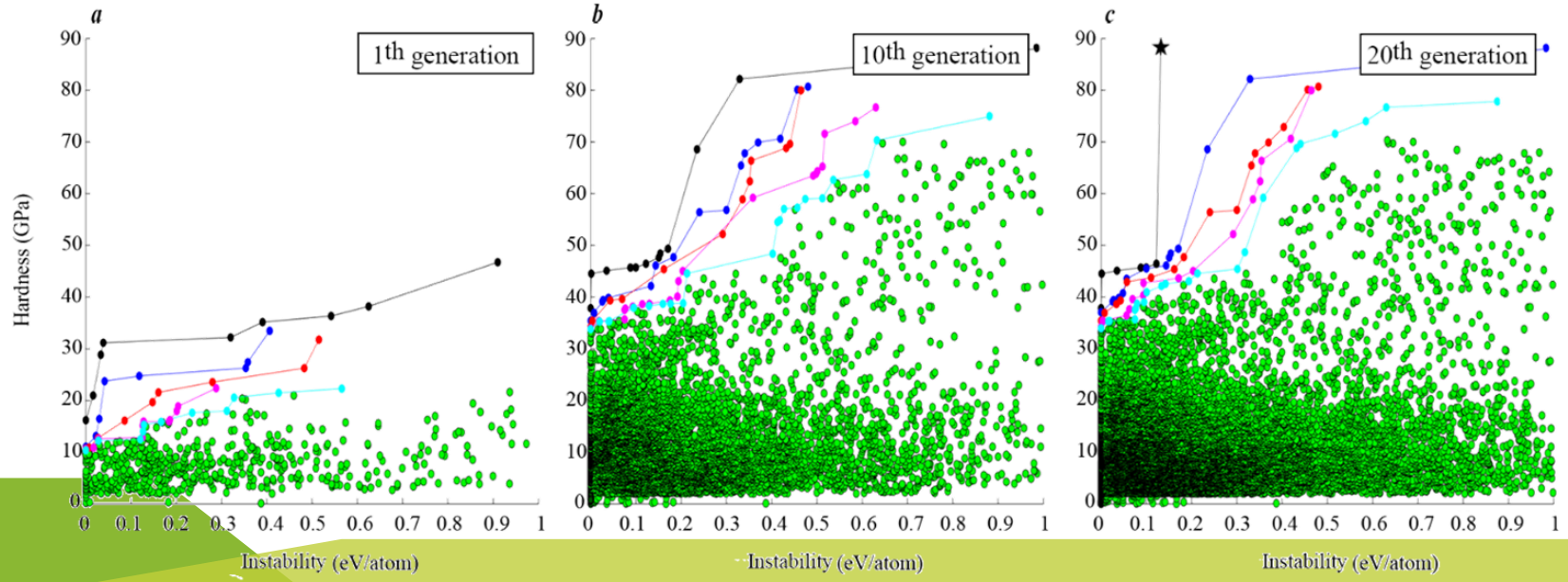

	Compounds	H_v (GPa)	K_{1C} (MPa.m$^{1/2}$)	Instability (eV/atom)	Space group		Compounds	H_v (GPa)	K_{1C} (MPa.m$^{1/2}$)	Instability (eV/atom)	Space group
Carbon	C	**92.7, (93.6), [96]**	6.33	0.13	$Fd\bar{3}m$	**Boron**	B	38.9, (39), [27-34]	2.87	0	$R\bar{3}m$
	C	**93.6**	6.36	0.139	$P6_3/mmc$		B	**44.8**	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	$F\bar{4}3m$
Mo-B	MoB_2	28.5,(33.1)[44],[24.2][53]	3.76	0	$R\bar{3}m$	**Tc-B**	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	$R\bar{3}m$
	MoB_4	35.4	3.57	0.099	$Pmmn$			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	$R\bar{3}m$
V-B	VB	39.1,(38.3)[49]	3.66	0	$Cmcm$	**Mn-H**	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	$Fm\bar{3}m$
	VB_7	39.7	3.19	0.143	$P\bar{3}m1$		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	$A2/m$
Mn-B	MnB_3	32.2	3.5	0.029	$P\bar{6}m2$	**Fe-B**	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	$Immm$
	MnB_4	38.2	3.56	0.1	$R\bar{3}m$		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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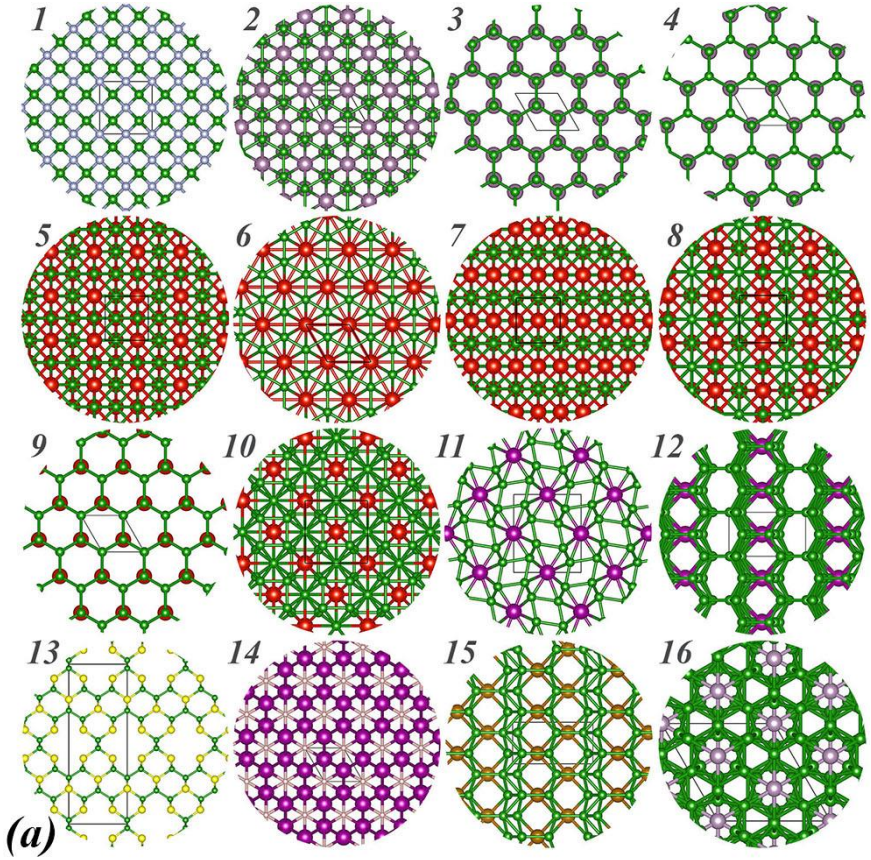
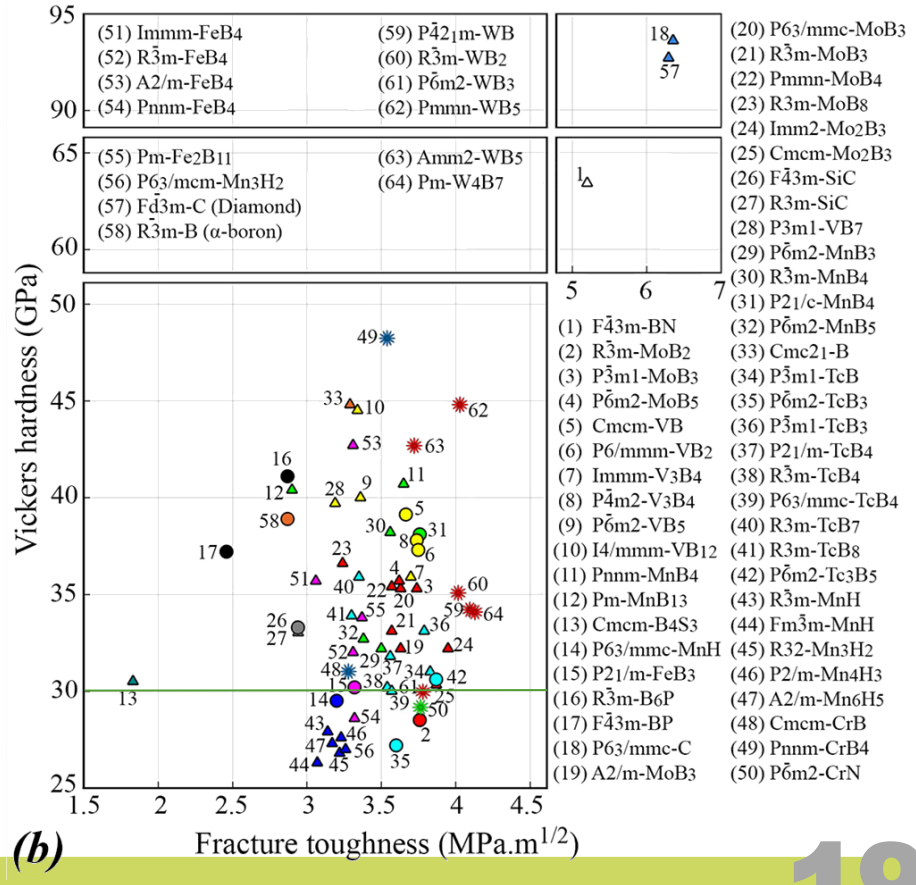
- *J. Phys. Chem. Lett.*, *8* (4), pp 755–764 **(2017)**. Cr-N, Cr-B
- *J. Phys. Chem. Lett.*, *9* (12), pp 3470–3477 **(2018)**. W-B



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?