HERAWS

Highlights on Europe Raw Materials Sustainability

UNIVERSITÉ DE LORRAINE

Jožef Stefan Institute

Co-funded by the Erasmus+ Programme of the European Union

The challenge of Critical Metals Substition

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www.heraws.eu

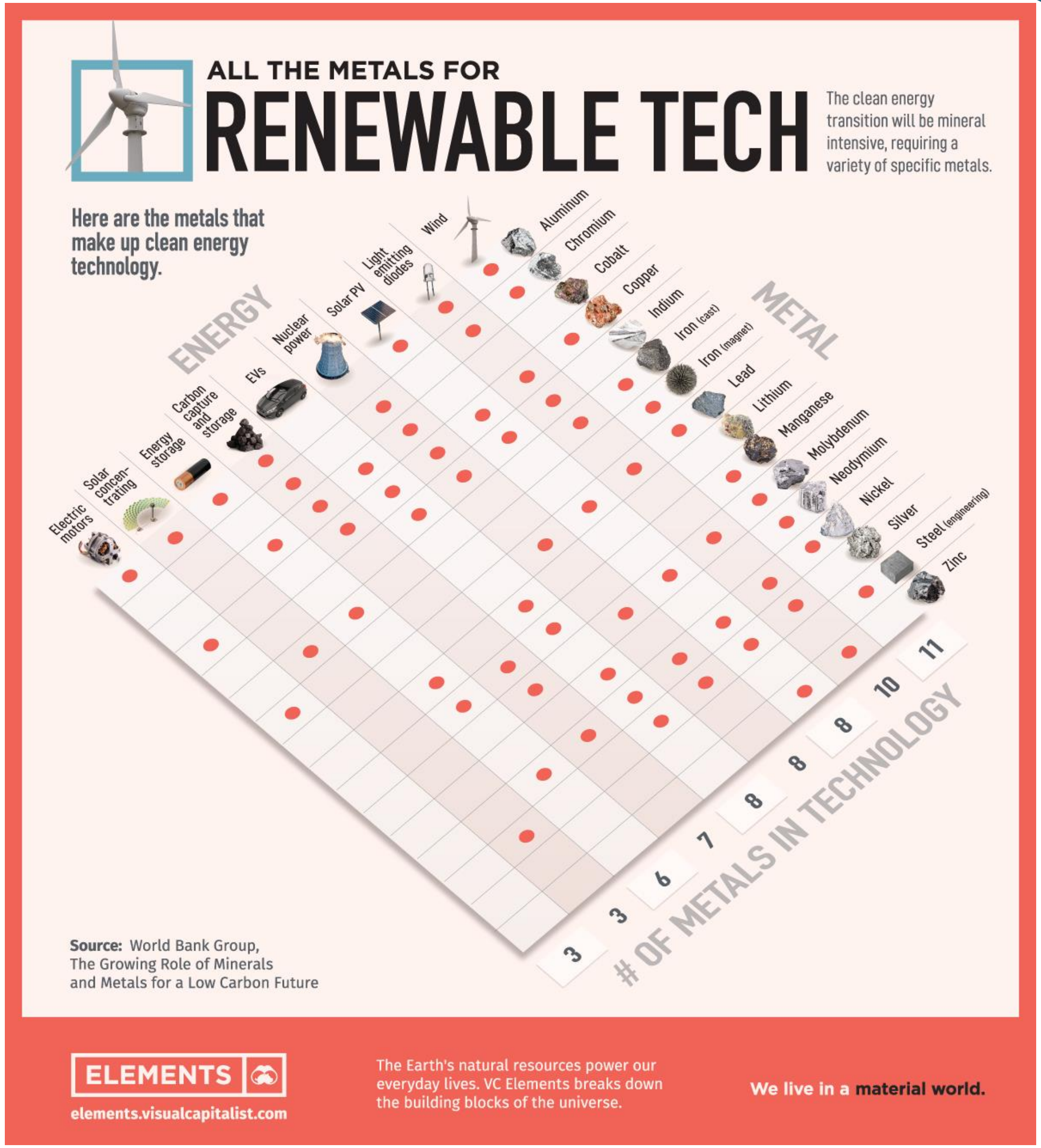
Context

20th century

Oil Era

21st century

Metal Era



Context

Metals for a low-carbon society

Olivier Vidal, Bruno Goffé and Nicholas Arndt

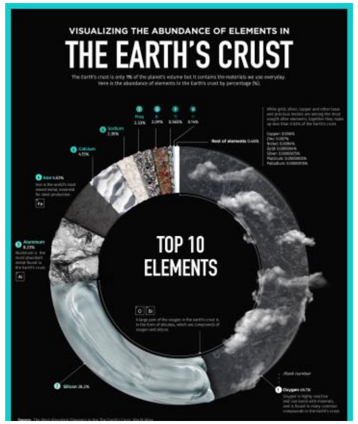
Renewable energy requires infrastructures built with metals whose extraction requires more and more energy. More mining is unavoidable, but increased recycling, substitution and careful design of new high-tech devices will help meet the growing demand.

A shift to renewable energy will replace one non-renewable resource (fossil fuel) with another (metals and minerals).

Risks

Abundance

Green transition induces a systemic change in the metal demands – possibly reaching the limit of the reserves/resources



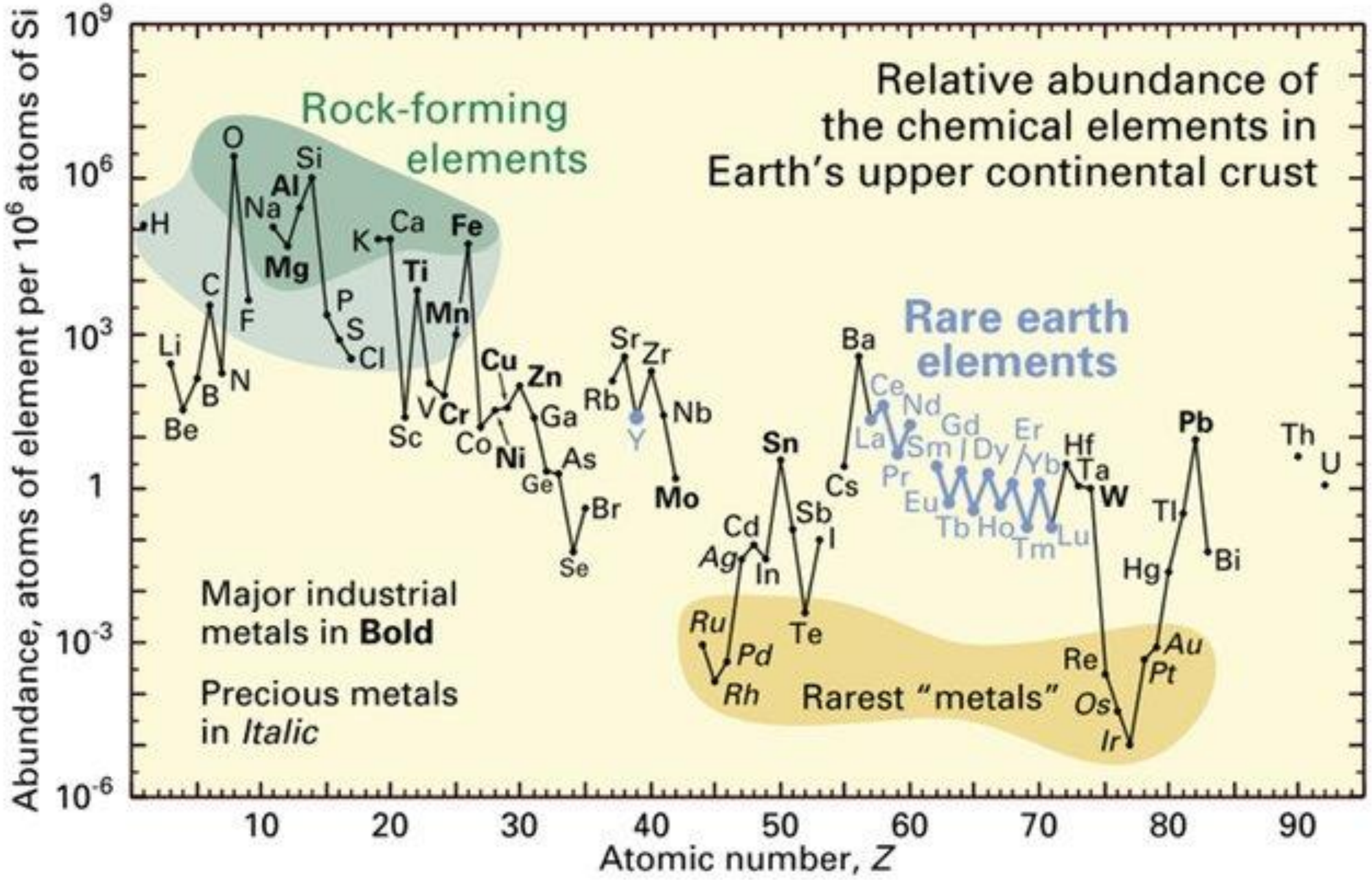
Environment

Mining – even for green application – is not an environnent/social/geopolitics neutral industry

Bypoducts

More than 50% of elements are not mined for themselves but as by products of a host metal. The (highly necessary) recycling of the host metals (Al, Cu, Fe, Ti…) may ironnically lead to shortage in the by products… necessary for the green and Digital transitions

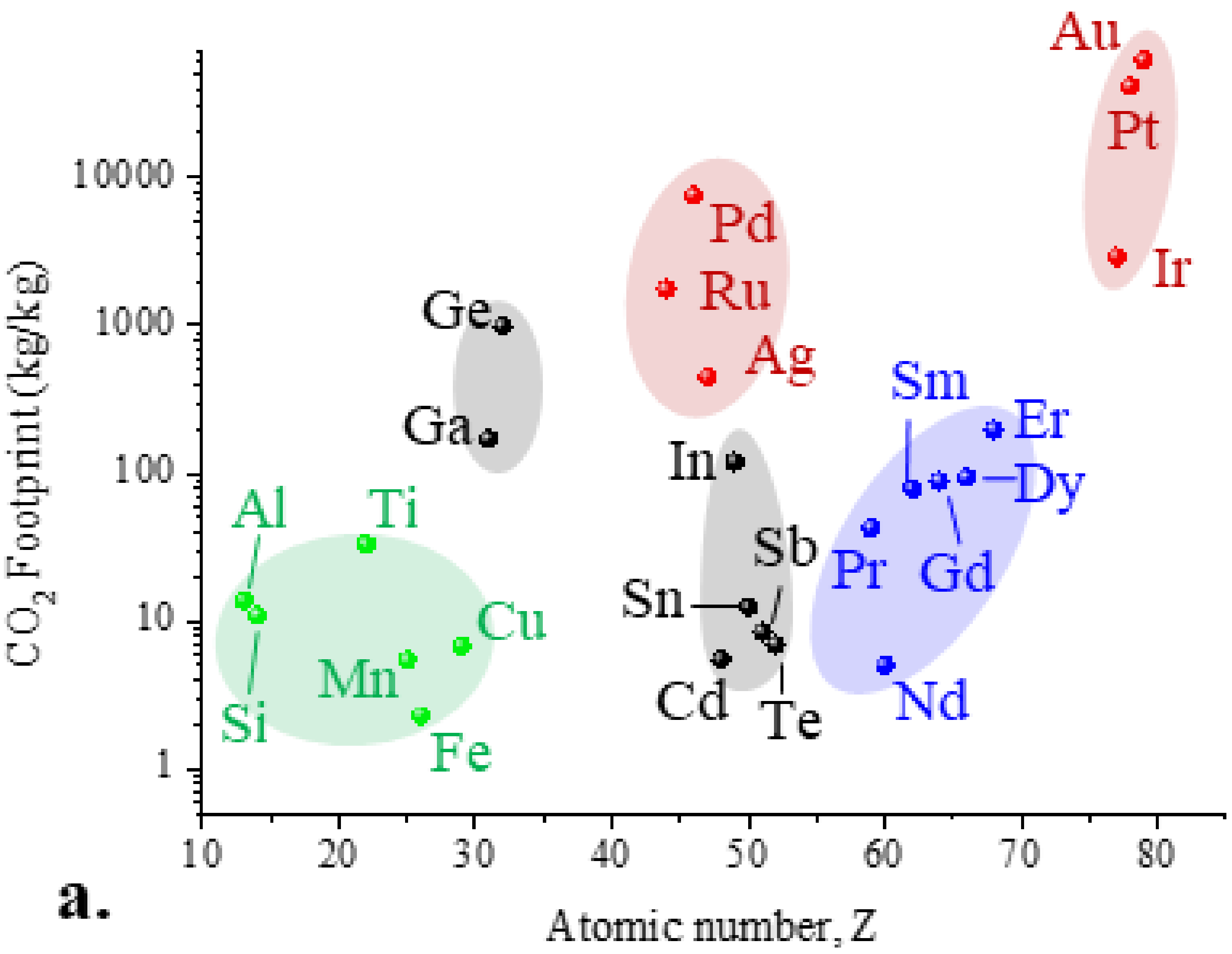
Abundance



6 – 12

orders of magnitudes between the abundance of Rock-forming elements and Platinoids

CO_2 Footprint

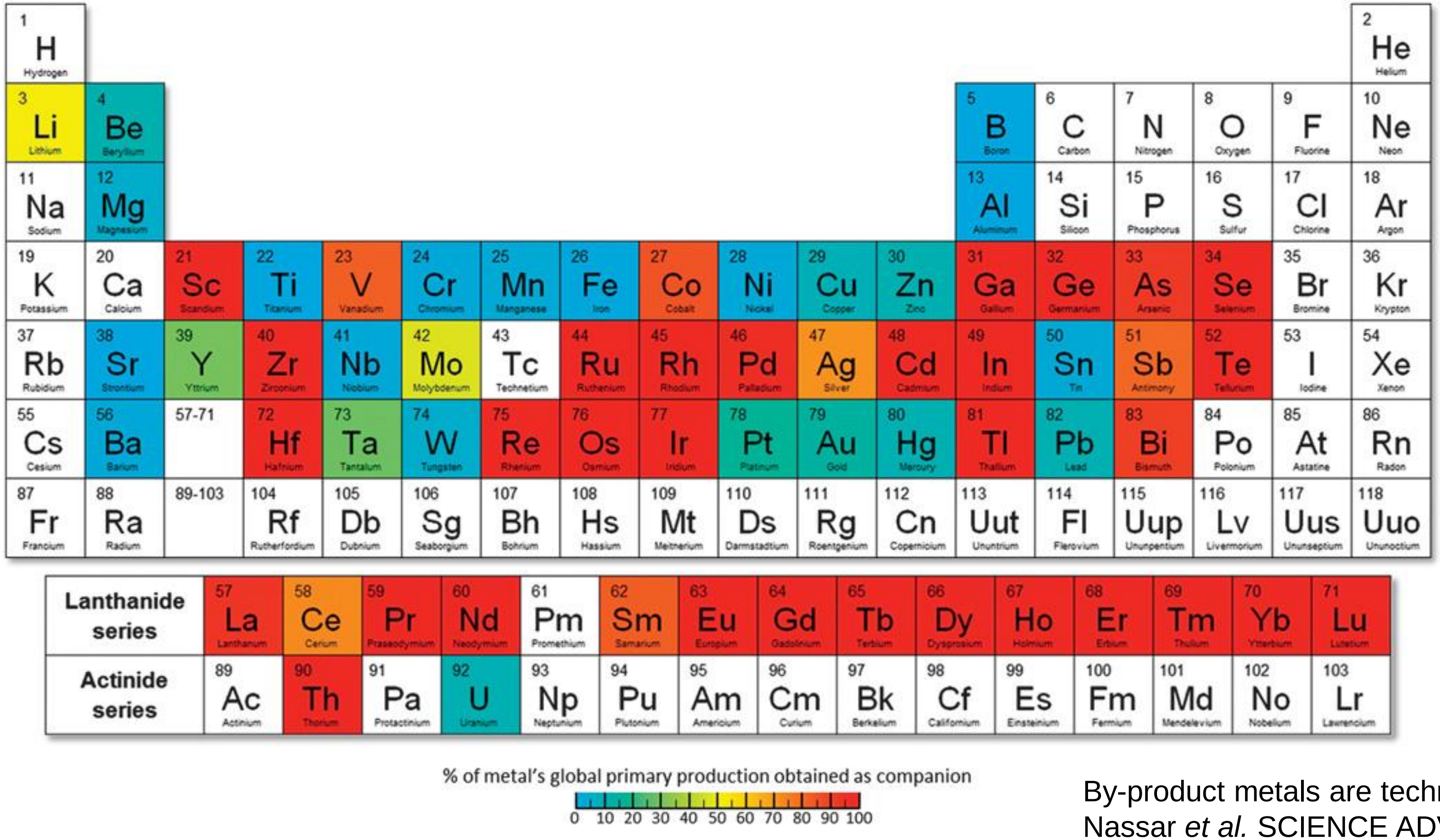


2 kg CO_2/kg of Iron
50t CO_2/kg of Gold

CO_2 Footprint

Extracting the volume of a bottle of water of Gold, requires to extract the volume of an Olympic swimming pool of rock

Byproducts



61%

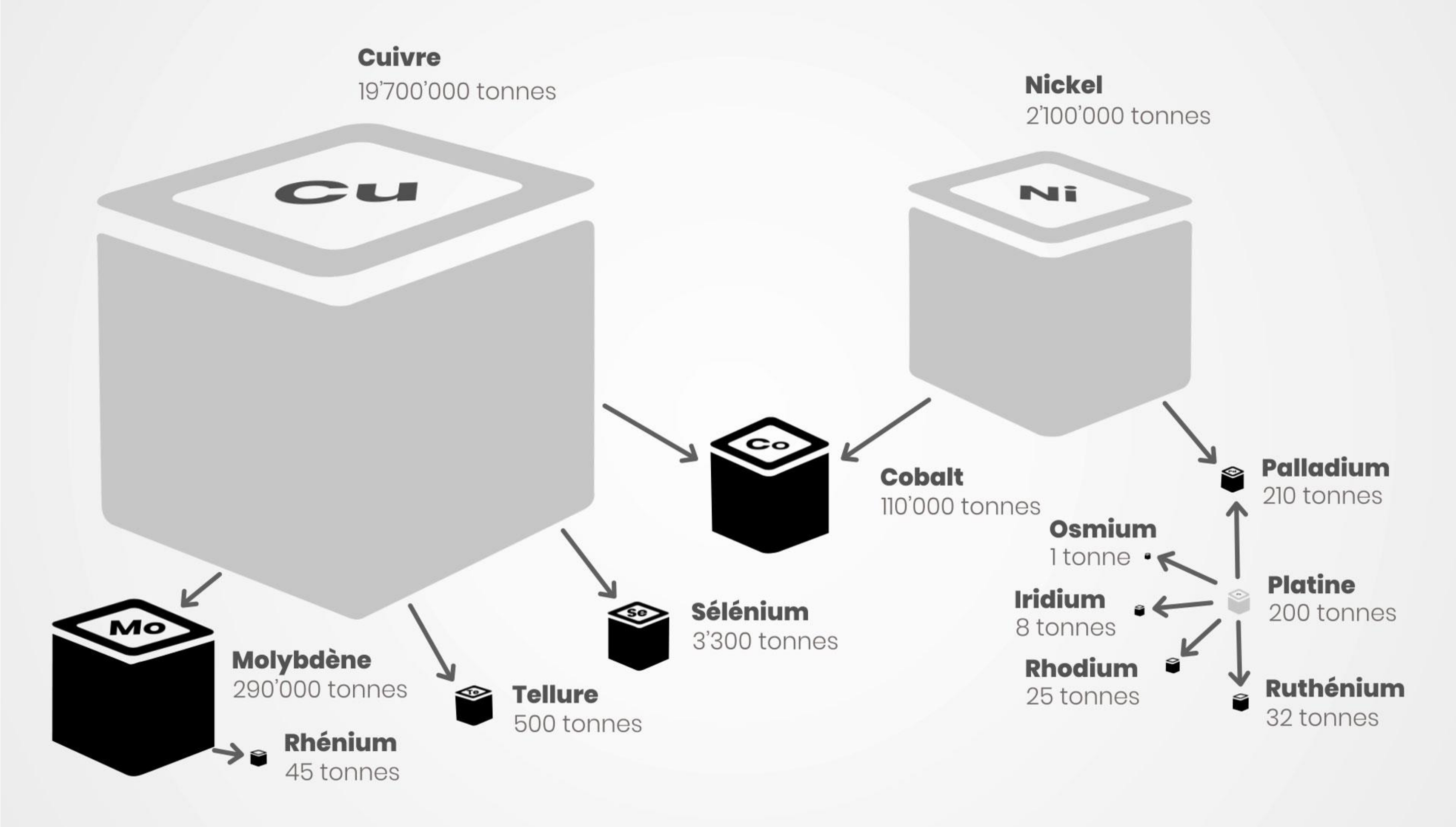
of metals and metalloids elements (38 of 62) have companionality greater than 50%

By-product metals are technologically essential but have problematic supply
Nassar *et al.* SCIENCE ADVANCES (2015)
DOI: 10.1126/sciadv.1400180

Byproducts

Price inelasticity of demand:

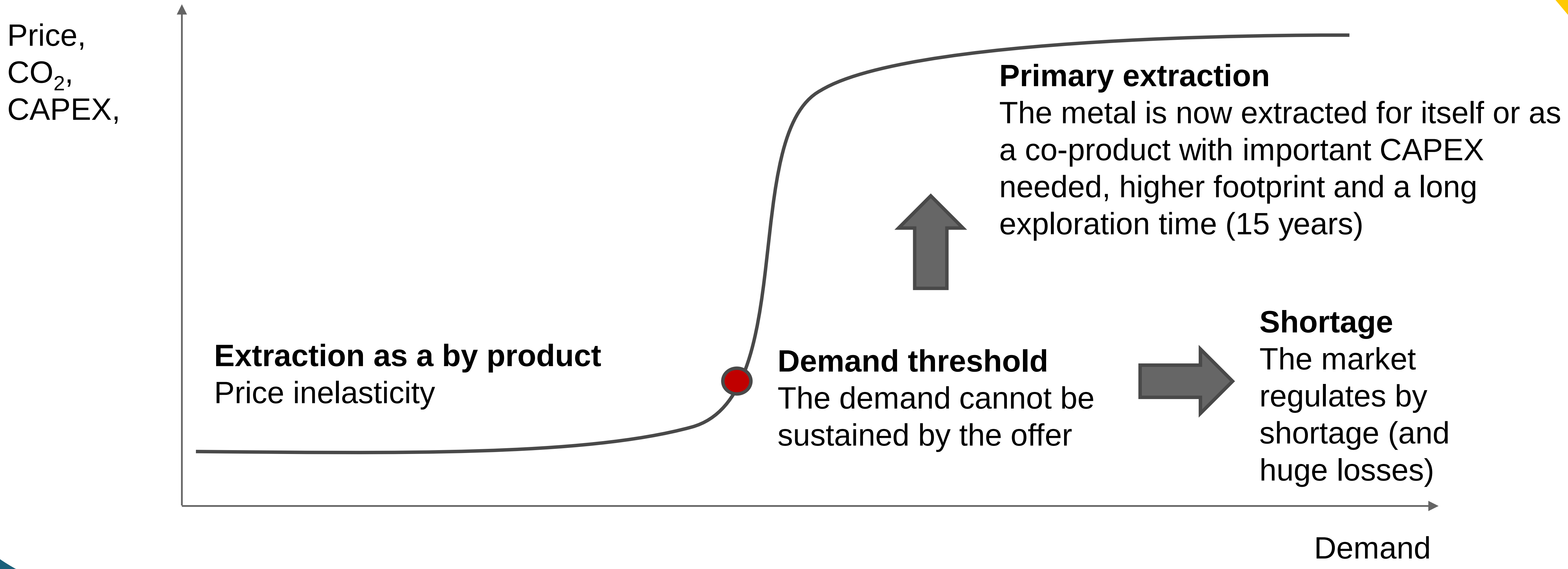
- The demand will not decrease due to a higher price
- The offer will not increase due to a higher price



https://cdmr.ch/

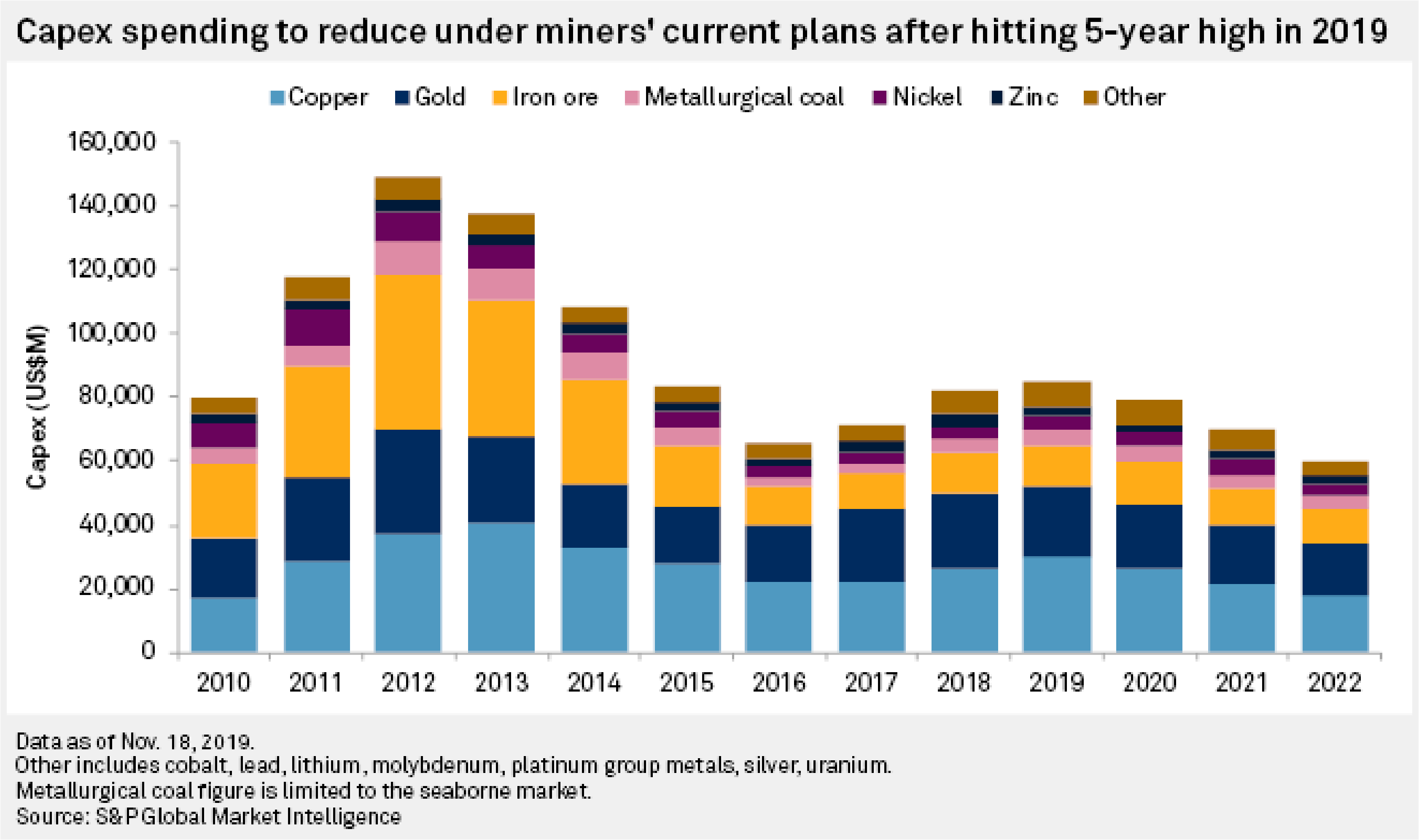
Byproducts

Strongly non-linear problem !



Adapted from Vincent Donnen - CDMR

Primary mining



Investment in Mining is at its lowest for 10+ yrs!

Shortage

Shortage:

- Became a reality after Covid 19 due to a mismatch between the restart of the different sector of economy (automotive vs. Semiconductors)

- Such shortage may happen more often due to “geologic reasons” which is more concerning!

- Shortage are much more dangerous than commodity price variation (hard to hedge)

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Autos & Transportation

2 minute read · October 20, 2021 7:08 PM GMT+2 · Last Updated 2 years ago

EXCLUSIVE Renault eyes bigger production cut as chip shortage rumbles on -sources

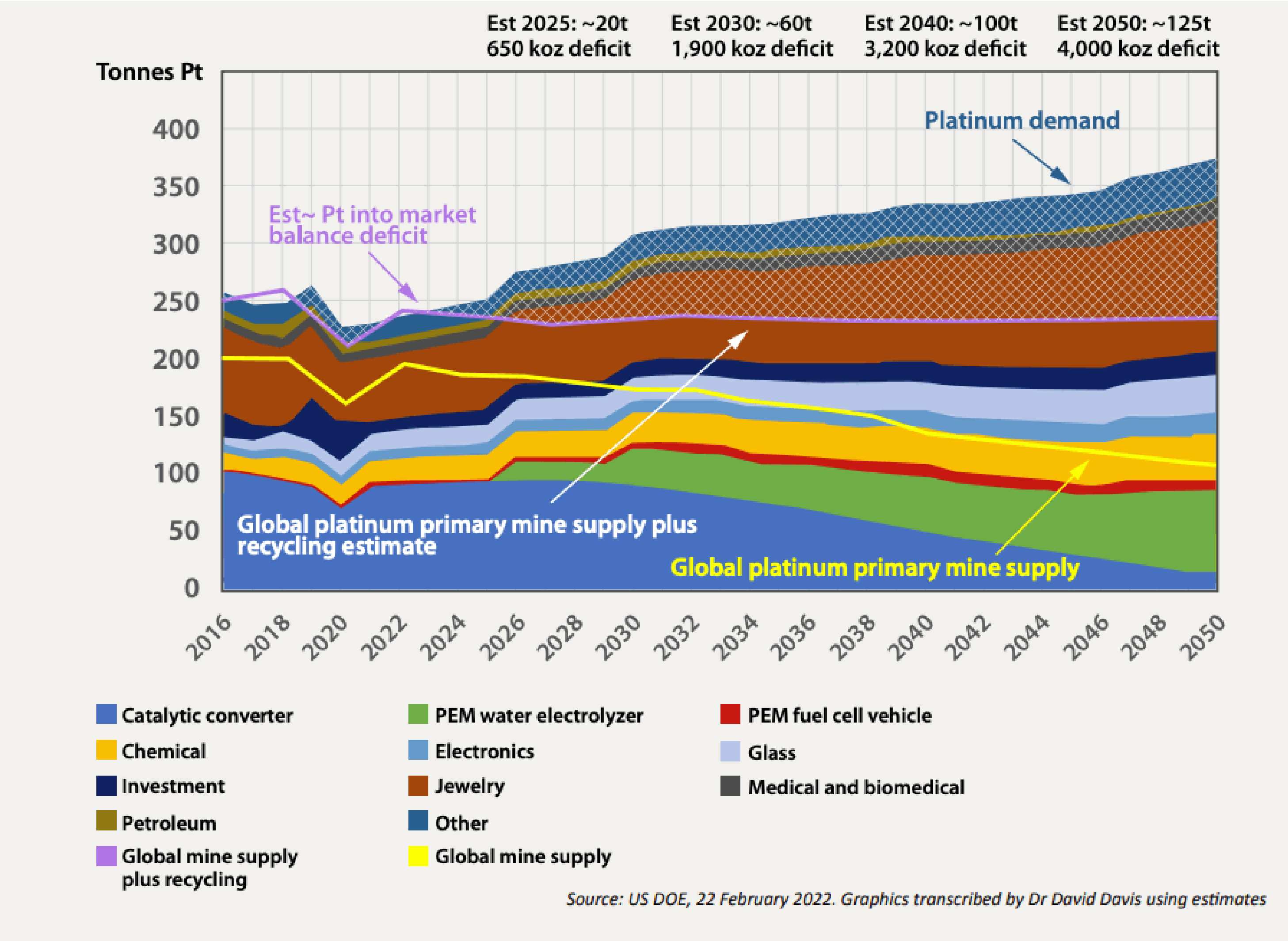
By Gilles Guillaume

Summary Companies

- Renault sees production shortfall of at least 300,000 vehicles this year - sources
- Co had previously estimated a shortfall of 220,000 vehicles
- Chip shortage could cost sector 11 mln lost vehicles this year - IHS

Shortage for Hydrogen economy?

Shortage for Hydrogen economy?

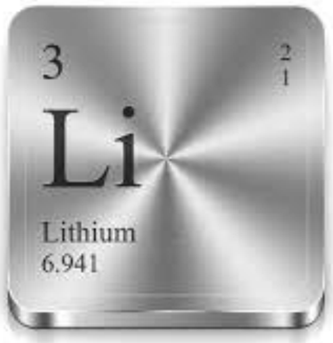


100s of billions €

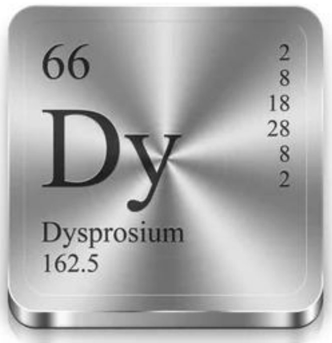
invested in Hydrogen.

These investment might be **very soon** in jeopardy due to Platinum Group metals shortage

Context



2109 %



433%

403%

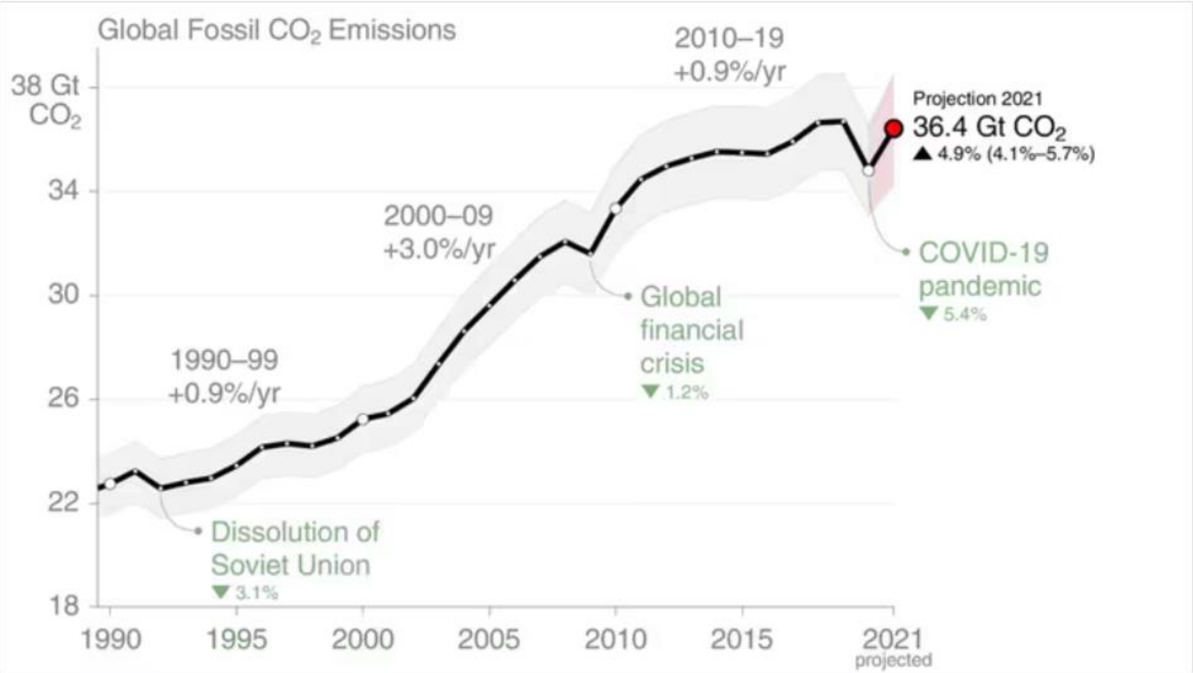
% metal required in 2050 for clean energy technologies vs. 2020 overall use (Source: KU Leuven – Eurometaux)

What can we do?

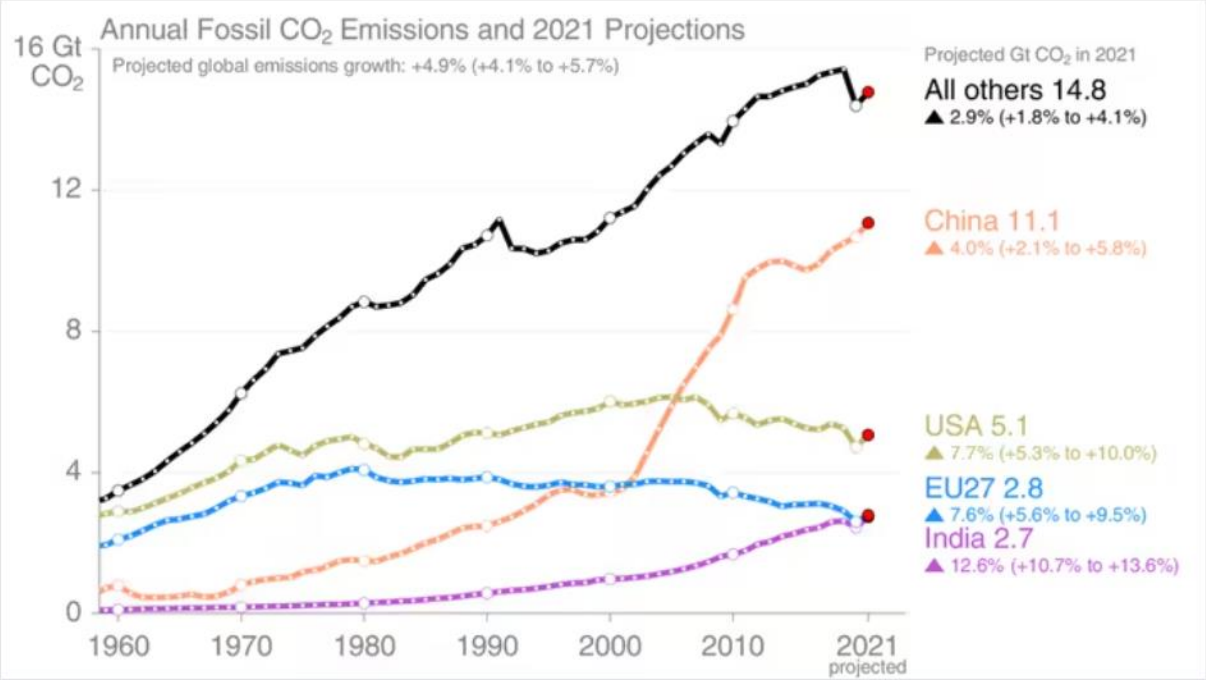
Reduce overconsumption?

Reduce overconsumption

1. A lever with **immediate & substantial effect** but hard to maintain **over long time period** (*e.g.* COVID 19)
2. High risk of **social unrest** if too brutal (*e.g.* 2008 financial crisis, COVID 19)
3. More a **‘Western’** view rather than a **global** view



Émissions mondiales annuelles de CO2 d'origine fossile. Les projections du Global Carbon Project montrent une hausse de 4,9% sur les projections en 2021. (GLOBAL CARBON PROJECT 2021)



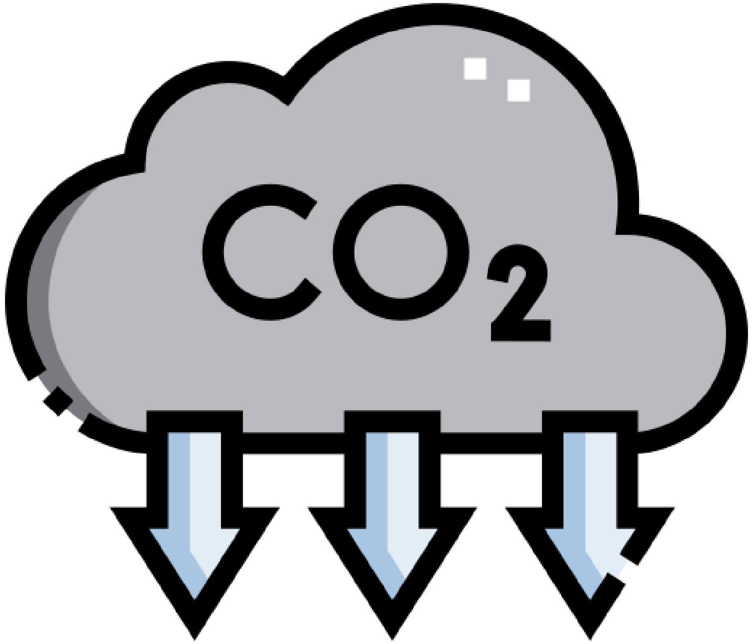
Émissions mondiales annuelles de CO2 d'origine fossile par pays. (GLOBAL CARBON PROJECT 2021)

Recycle ?

Recycling

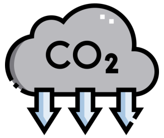

PRIMARY RESOURCES
EXPLORATION
WASTE
MINING
WASTE
PROCESSING
WASTE
RAW MATERIALS
WASTE
DESIGN
Substitution of Raw Materials
PRODUCTION
WASTE
USE, REUSE
WASTE
COLLECTION
WASTE
RECYCLING
WASTE
eit RawMaterials
Connecting matters

Recycling



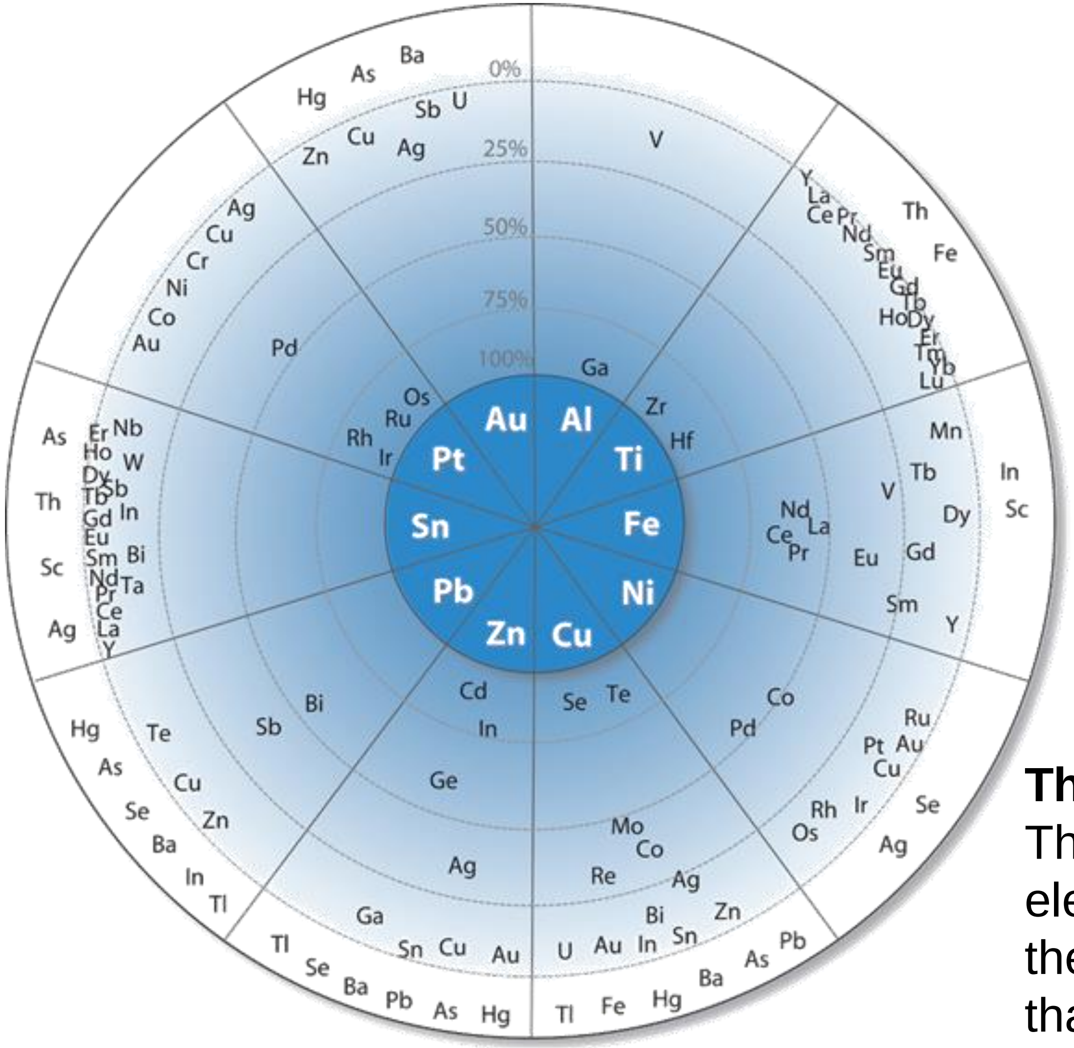
Primary extraction
2 kg CO_2/kg of Iron
12 kg CO_2/kg of Aluminum
380 kg CO_2/kg of Silver
50t CO_2/kg of Gold

Carbon footprint divided by **3-6** for abundant materials **10-50** for precious metals



Recycling
0.7 kg CO_2/kg of Iron
2.5 kg CO_2/kg of Aluminum
38 kg CO_2/kg of Silver
1t CO_2/kg of Gold

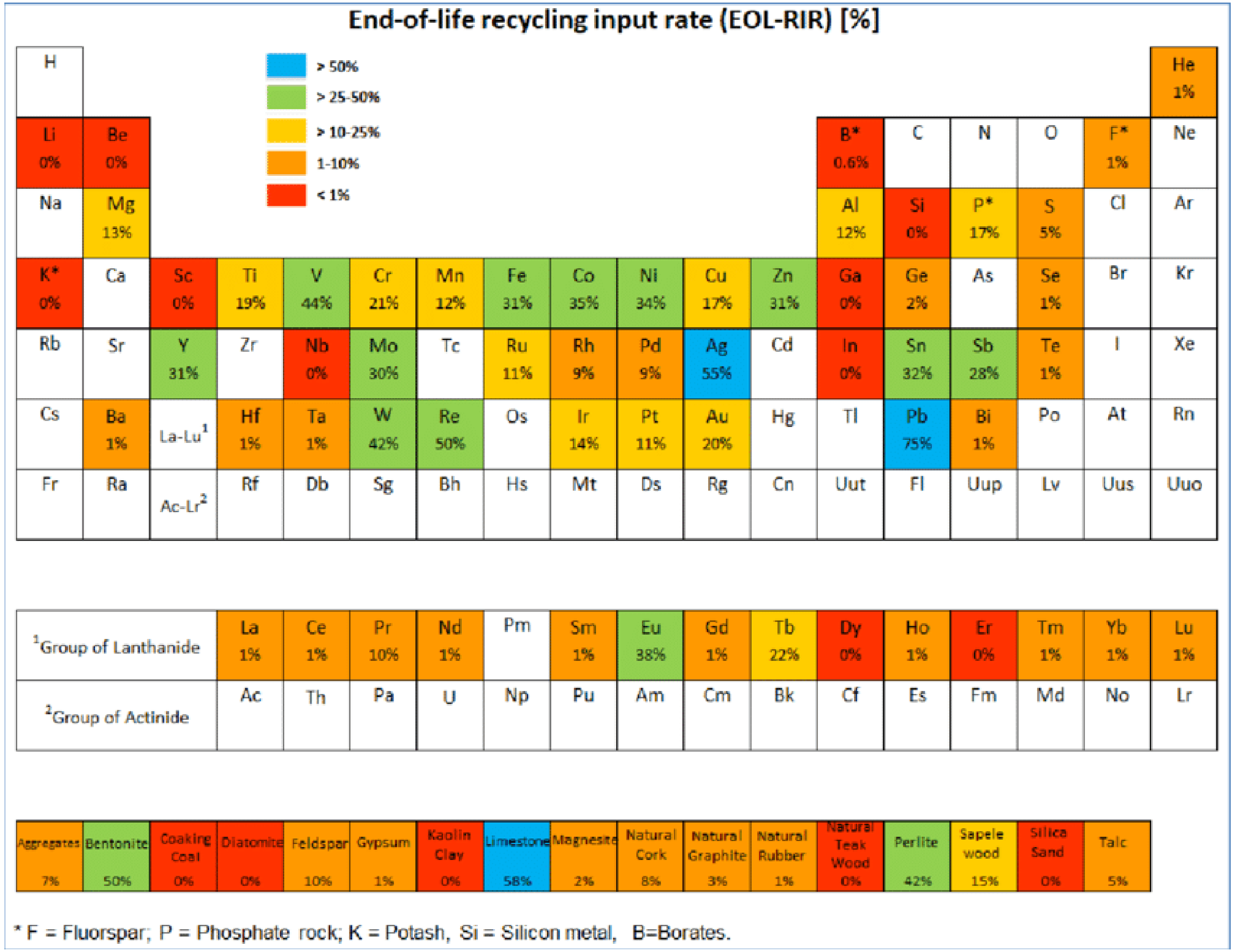
Recycling



Recycling may be a risk for the by products!

The wheel of metal companionality (Nassar *et al.)*
The principal host metals form the inner circle. Companion elements appear in the outer circle at distances proportional to the percentage of their primary production (from 100 to 0%) that originates with the host metal indicated.

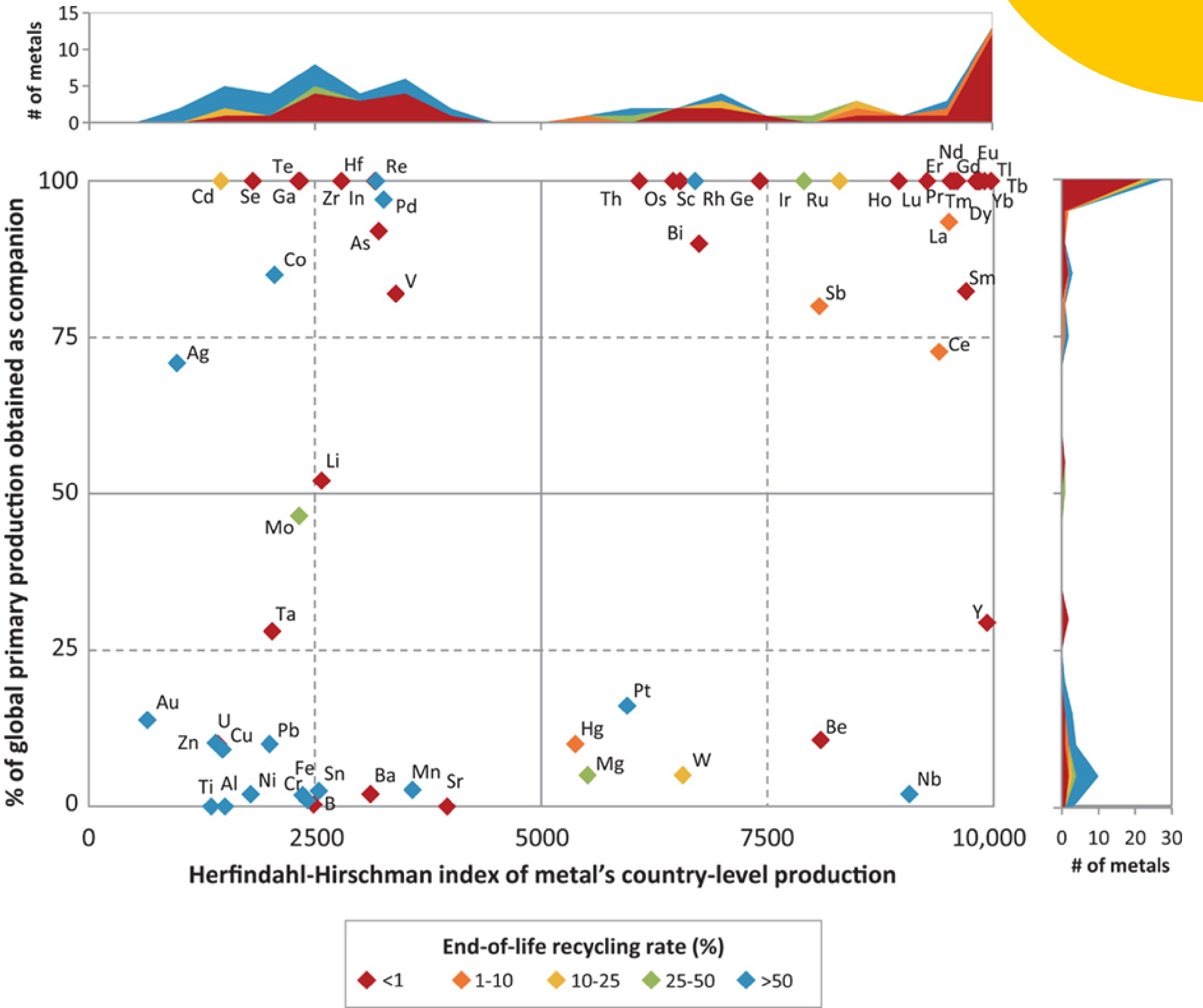
Recycling



By products are (ironically) poorly recycled…

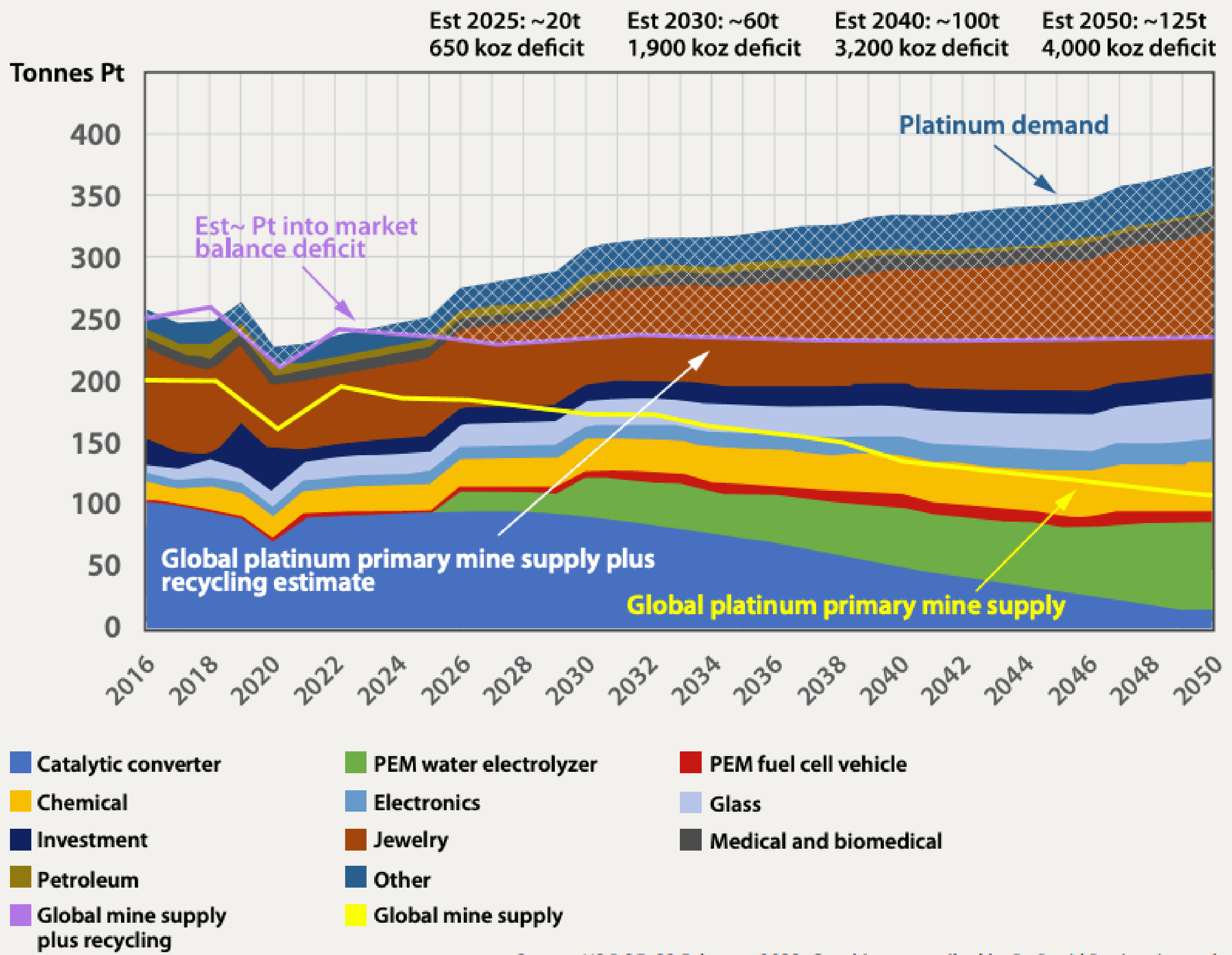
End-of-life recycling input rates (EOL-RIR) for the EU-28

Recycling



# of metals
% of global primary production obtained as companion
Herfindahl-Hirschman index of metal's country-level production
# of metals
End-of-life recycling rate (%)
<1
1-10
10-25
25-50
>50
0
2500
5000
7500
10,000
0
25
50
75
100
5
10
15
20
30
Cd Se Te Ga Zr Hf In Re Pd As Co V Ag Li Mo Ta Au U Zn Cu Pb Ti Al Ni Fe Cr Sn B Ba Mn Sr
Th Os Sc Rh Ge Ir Ru Bi Ho Lu Er Nd Pr Gd Tm Eu Tl Dy Tb Yb La Sb Sm Ce Y Pt Hg Mg W Be Nb

The case of Platinum



Source: US DOE, 22 February 2022. Graphics transcribed by Dr David Davis using estimates

Recycling will **compensate** the decrease in mine supply but will not avoid Platinum **shortage**

Substitute ?

The substitution equation

- **Constraint 1:** Green products must save more CO_2 usage than the extraction of its raw materials generated
- **Constraint 2:** Green products must be affordable
- **Constraint 3:** Green products must equal or outperform the existing ones
- **Constraint 4:** Materials used must be abundant
- **Constraint 5:** Materials must not be produced in non-democratic countries
- **Constraint 6:** Coal&Gas must be replaced by electricity or H_2 in the industrial processes but not from nuclear power

Solution of the equation:

$$\{\emptyset\}$$

A possible solution: Expand the library of Functional Nanomaterials

What do we do?

Do it fast!

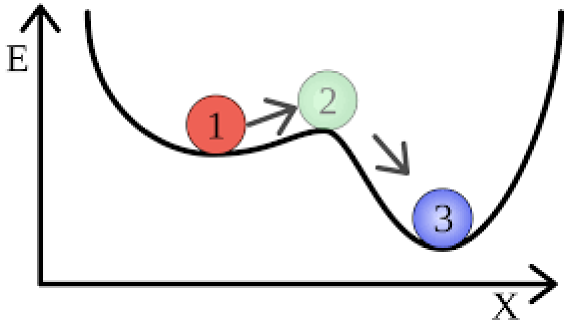
Explore &Discover

New materials with high performance

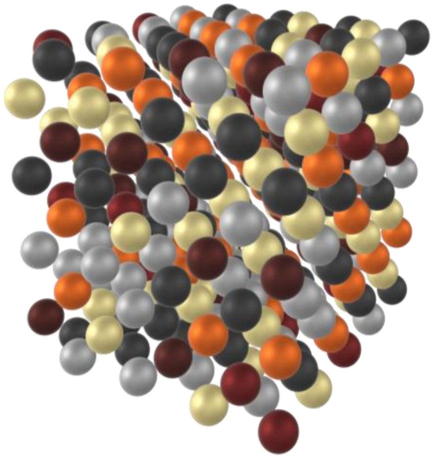
Assess

Materials sustainability, even at low TRL

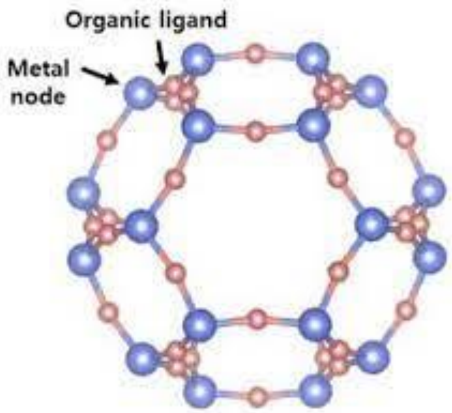
New constallations of Materials



Metastable Materials



High Entropy Alloys

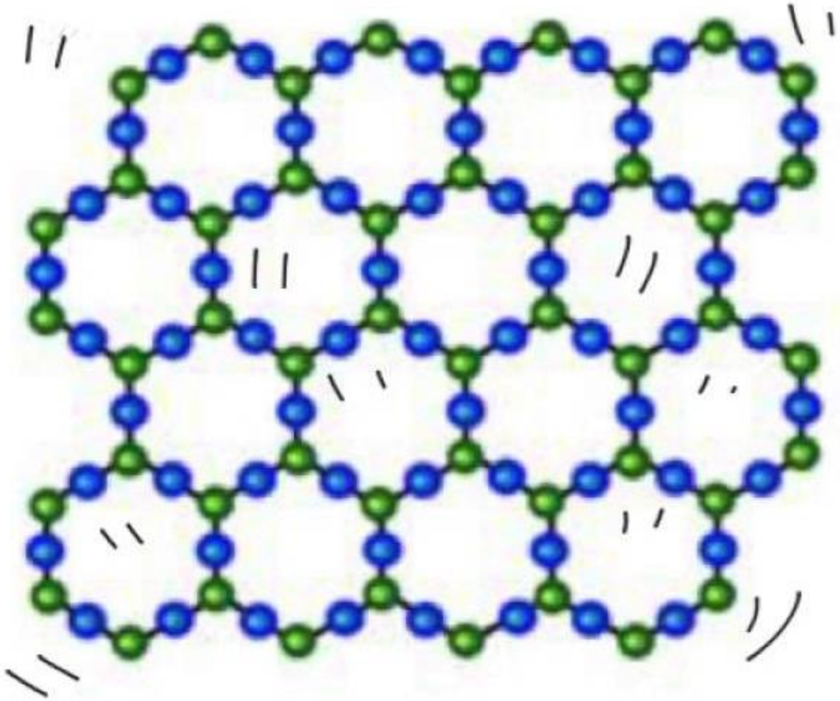


Metal-Organic Frameworks

What are High Entropy Alloys?

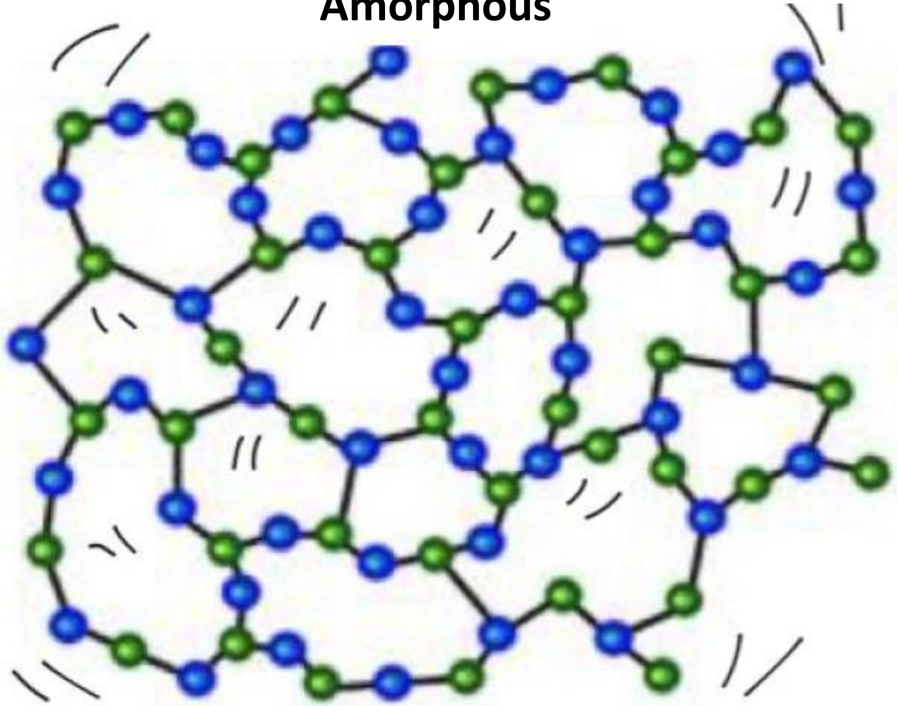
Classical model: crystalline vs. amorphous

Crystalline



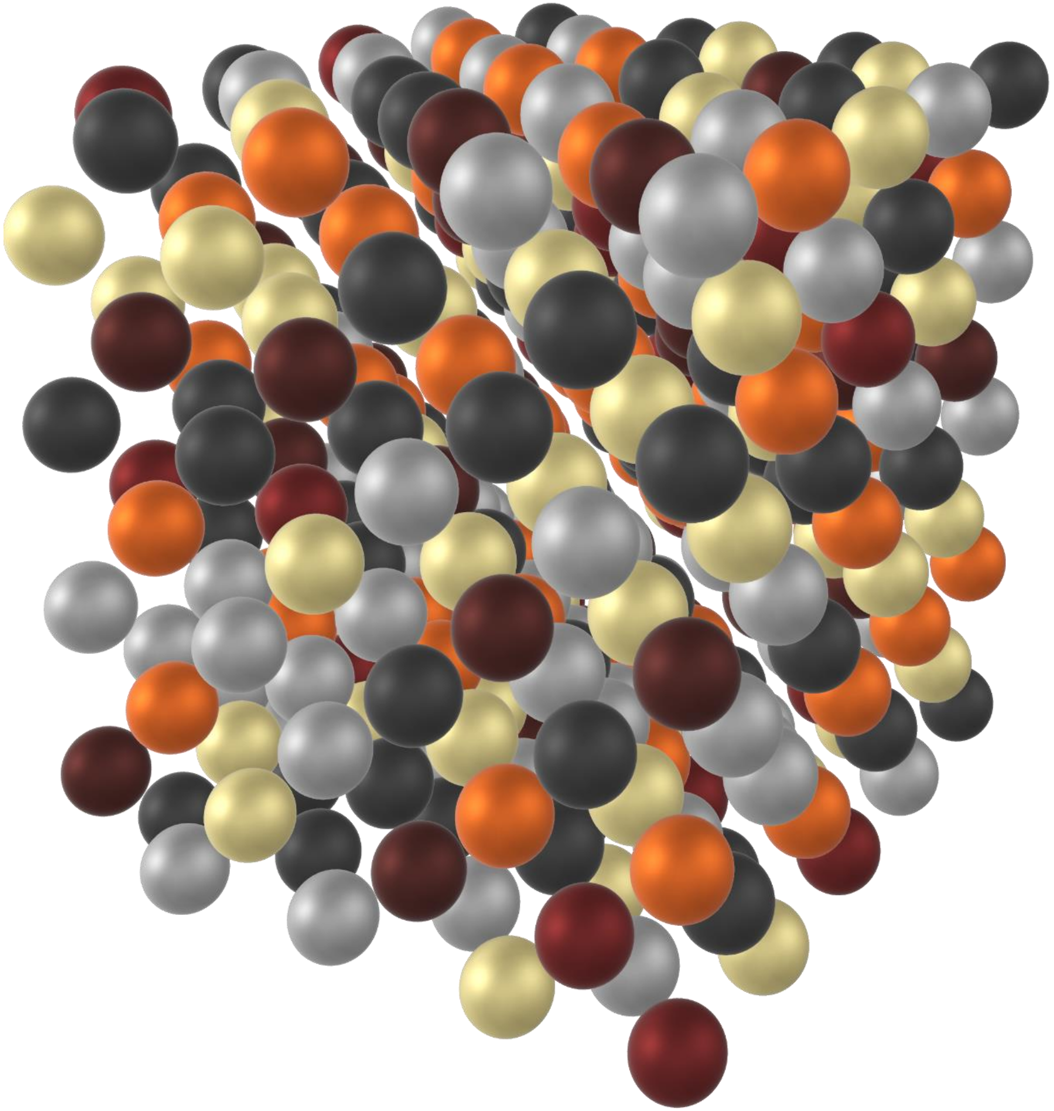
Atoms vibrate in place in a fixed pattern

Amorphous



Atoms vibrate in place in more random arrangements

High Entropy alloys: ordered and disordered



- A three-dimensional periodic atomic network with random chemical decoration.
- A single-phase solid solution containing at least 5 elements with a molar composition of at least 5%

Why HEA are interesting?

Enhanced properties

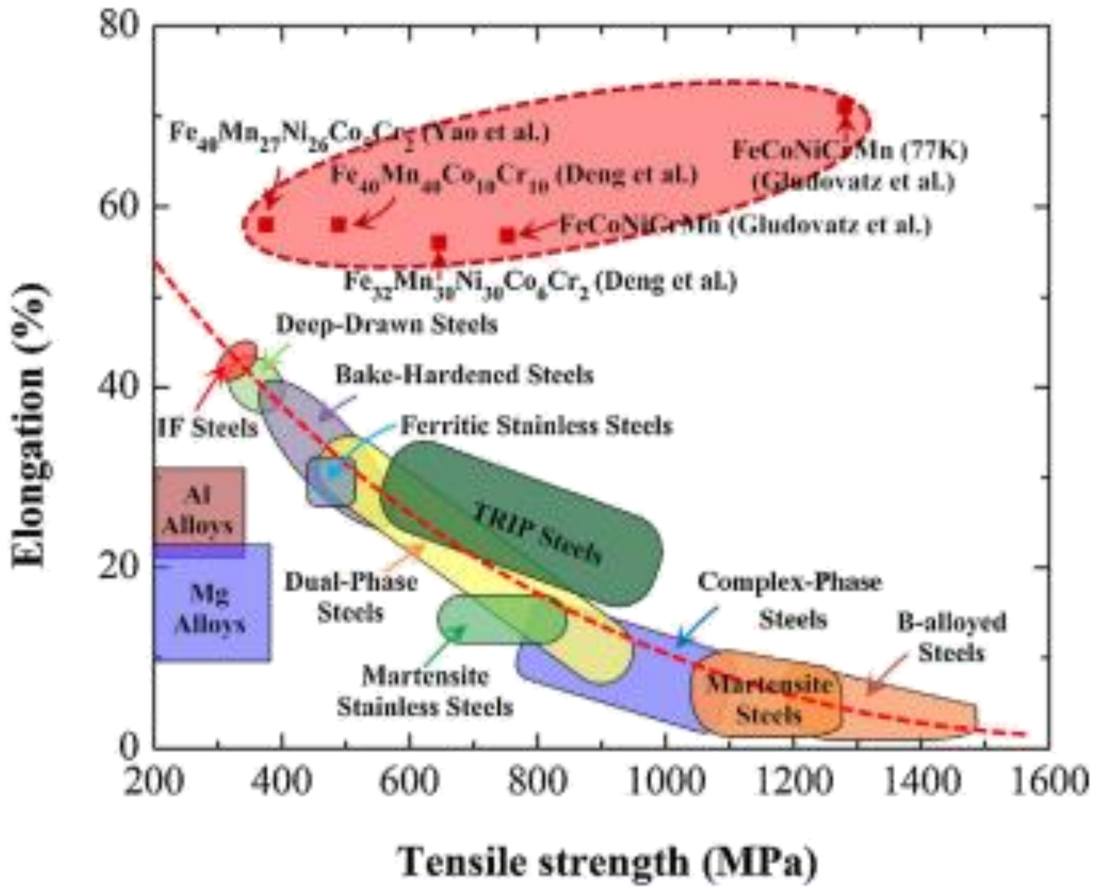


FIGURE 9

Strength versus ductility properties for low-SFE HEAs such as $Fe_{40}Mn_{27}Ni_{26}Co_5Cr_2$ [74], $Fe_{40}Mn_{40}Co_{10}Cr_{10}$ [65], $Fe_{32}Mn_{30}Ni_{30}Co_6Cr_2$ [65], and FeCoNiCrMn [6] at room temperature and FeCoNiCrMn [6] at 77 K, compared with other conventional alloys [75].

High-entropy alloy: challenges and Prospects
Ye *et al.* Materials Today Volume 19, 2016

Non-linear thinking

nature COMMUNICATIONS

COMMENT

https://doi.org/10.1038/s41467-019-09700-1 OPEN

High entropy alloys as a bold step forward in alloy development

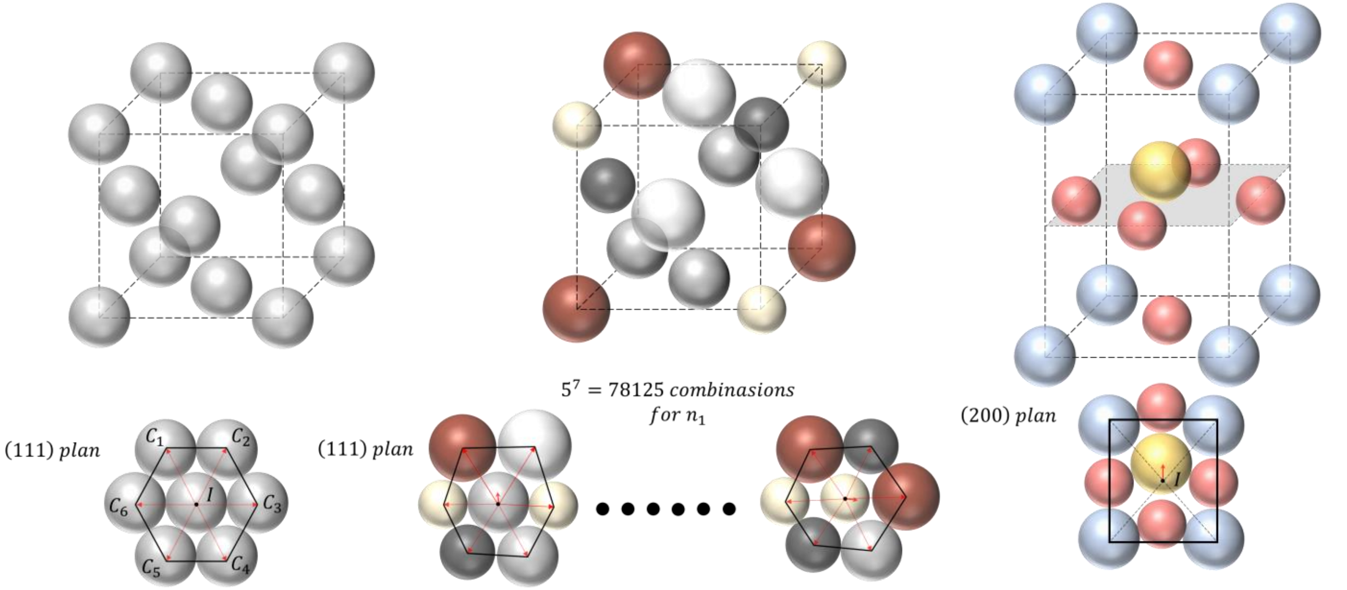
D.B. Miracle[1]

Diluting a base element with small amounts of another has served as the basis for developing alloys for thousands of years since the advent of bronze. Today, a fundamentally new idea where alloys have no single dominant element is giving new traction to materials discovery.

$$\boldsymbol{P_{HEA} \neq \sum x_i P_i}$$

- P_i is a property of the element *i*
- x_i is the concentration of the element i in the HEA

Outliers



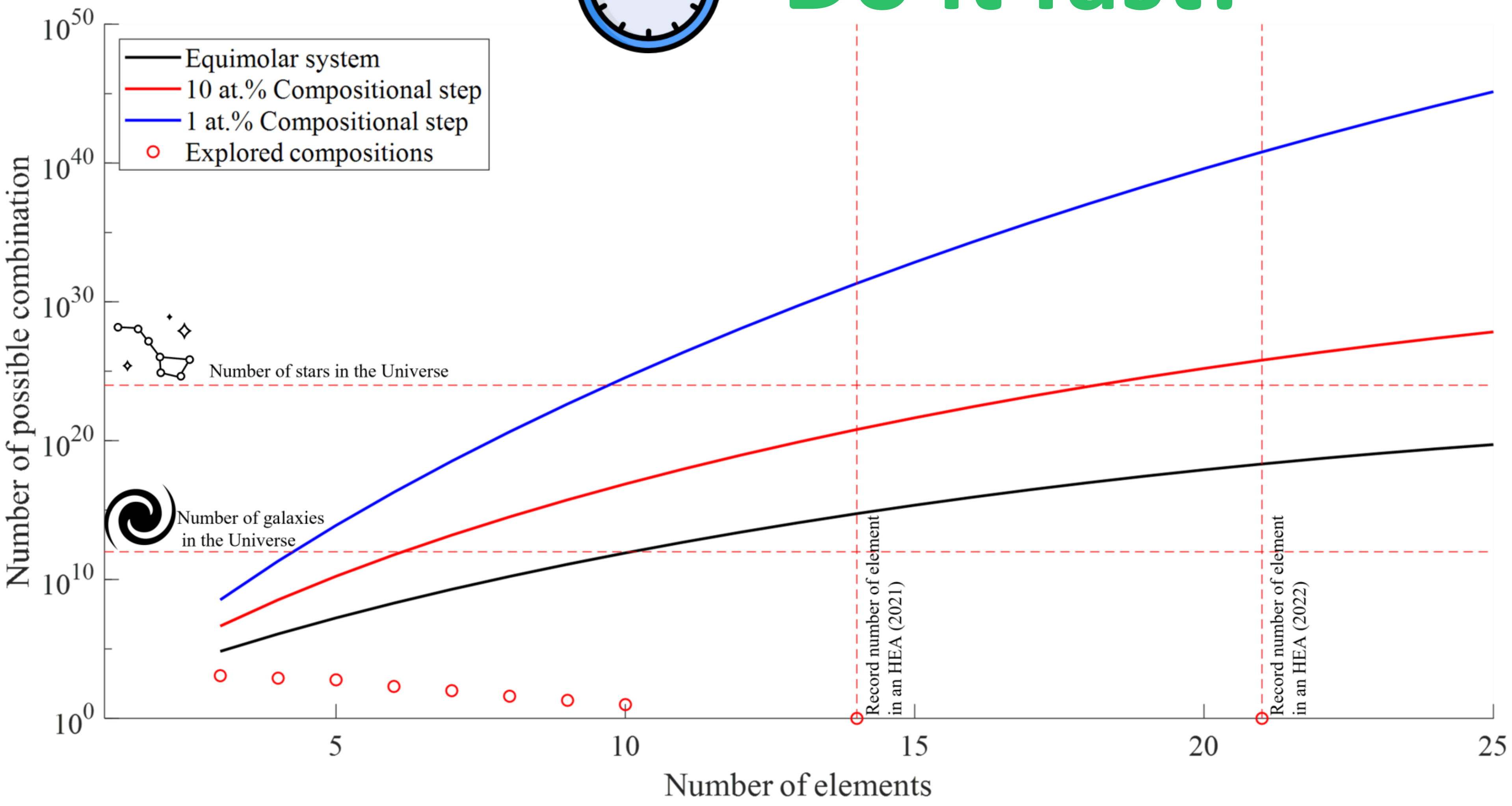
	Pure Metal	High Entropy Alloys	Non − linear crystal
Unit displacement vector: $\vec{d} = \sum_{i=1}^{6} \overrightarrow{IC_i}$	$\vec{d} = \vec{0}$	$\vec{d} \neq \vec{0}$	$\vec{d} \neq \vec{0}$
Displacement vector over n lattice: $\vec{D} = \sum_{j=1}^{n} \vec{d}_j$	$\vec{D} = \vec{0}$	$\vec{D} = \vec{0}$	$\vec{D} = n\vec{d}$
Average unit displacement: $\bar{d} = \sum_{j=1}^{n} \frac{\lVert \vec{d}_j \rVert}{n}$	$\bar{d} = 0$	$\bar{d} \neq 0$	$\bar{d} = \lVert \vec{d} \rVert$
Standard deviation σ_d	$\sigma_d = 0$	$\sigma_d \neq 0$	$\sigma_d = 0$

- Many theories predicting physical properties cannot accommodate High Entropy Alloys

Why AI is needed?

Answers

Do it fast!



The classical (simplified) theory

- A stable material is a material with the lowest G (free enthalpy)

$$G = H - TS$$

***H* , Enthalpy**
related to the energy needed to bind the atoms
(the lowest the better)

***S*, Entropy**
Related to the number of possible combination
(the disorder)

The classical theory

G = H - TS

The conditions under which an element could dissolve (Hume-Rothery rules) in are empirically defined as :

- The atomic radii of the solute and solvent atoms must differ by no more than 15%:
- The crystal structures of solute and solvent must match.
- Maximum solubility occurs when the solvent and solute have the same valency. ...
- The solute and solvent should have similar electronegativity

William Hume-Rothery OBE FRS (15 May 1899 – 27 September 1968) was an English metallurgist and materials scientist who studied the constitution of alloys.

Birds of a feather flock together

Qui se ressemble s'assemble

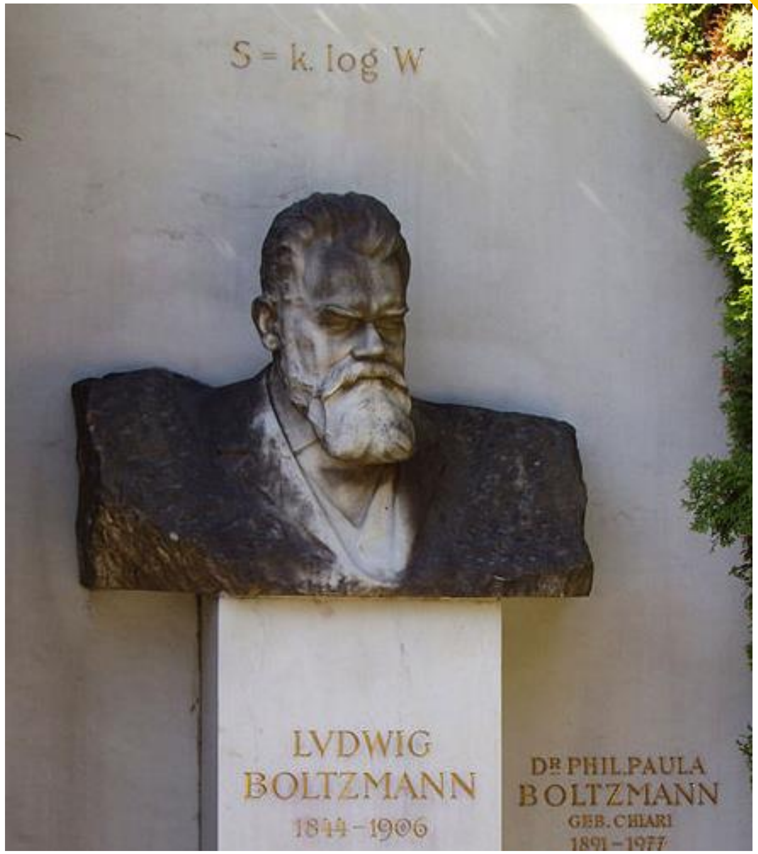
The classical theory

$$G = H - TS$$

The entropy is proportional to the logarithm of the number of combination Ω: $S = k_B \ln \Omega$

The number of combination increases with the number of alloying element *n*: $\Omega = N! \prod_{i=1}^{n} \frac{1}{N_i!}$

$$S = k_B N_{Av} \sum_{i=1}^{n} x_i \ln x_i$$



The classical theory

G = H - T**S**

The entropy is proportional to the logarithm of the number of combination Ω: $S = k_B \ln \Omega$

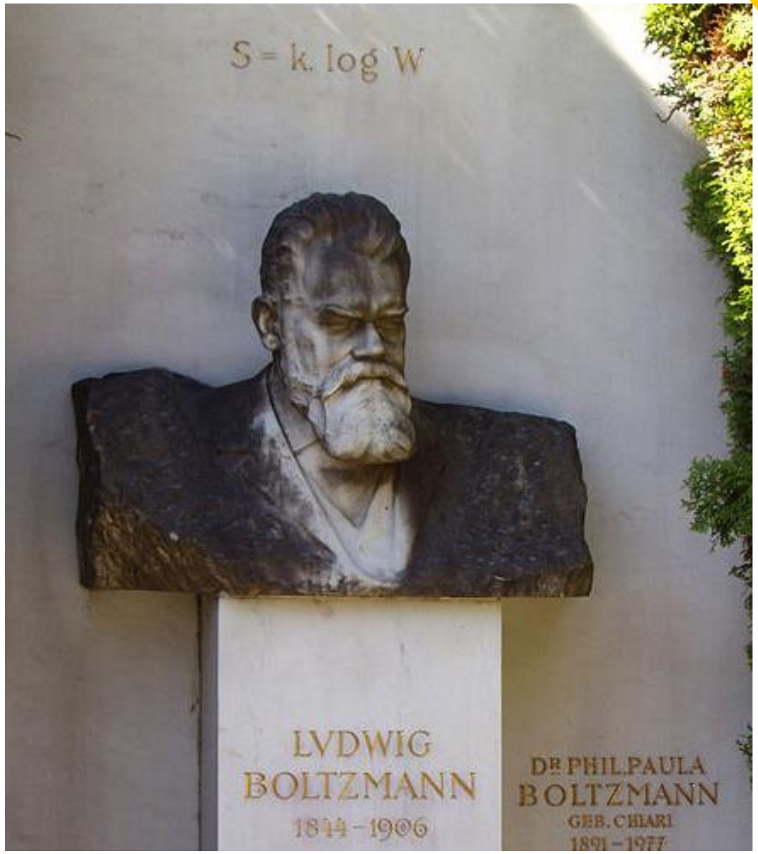
The number of combination increases with the number of alloying element *n*: $\Omega = N! \prod_{i=1}^{n} \frac{1}{N_i!}$

The entropy is maximal if the **number of element increases**, and if they are in **equimolar concentration**:

$$S = k_B N_{Av} \sum_{i=1}^{n} x_i \ln x_i$$

Boredom was born from uniformity

L'ennui naquit un jour de l'uniformité – La Motte



Basic theory

1. Configurational entropy is:
    a. Maximal for equimolar alloys
    b. Increases with the number of alloying element

2. According to this theory High entropy alloy should be more likely:
    a. At equimolar composition
    b. For higher number of alloying element

First massive modeling

Contents lists available at ScienceDirect

Scripta Materialia

journal homepage: www.elsevier.com/locate/scriptamat

ELSEVIER

Regular Article

Computational design of light and strong high entropy alloys (HEA): Obtainment of an extremely high specific solid solution hardening

Edern Menou [a,b], Franck Tancret [a,c,*], Isaac Toda-Caraballo [c,d], Gérard Ramstein [b], Philippe Castany [e], Emmanuel Bertrand [a], Nicolas Gautier [a], Pedro Eduardo Jose Rivera Díaz-Del-Castillo [c,f]

An interesting fact is that the Pareto set contains no equiatomic alloy. Although equimolarity was initially considered to favour the stability of HEAs and to confer them a high SSH [27], it was already recognised that configurational entropy was not a major feature triggering the formation of a single solid solution [26]; this is confirmed here. Actually, it seems that the algorithm pushed compositions as far as possible away from equimolarity; this is evidenced by calculating the average deviation (*AD*) from equimolar compositions as:

$$AD = \frac{1}{n}\sum_{i=1}^{n}\left|x_i - \frac{1}{n}\right|$$

According to this theory High entropy alloy should be more likely:

~~a. At equimolar composition~~

b. For higher number of alloying element

First massive modeling

ARTICLE

Received 6 Nov 2014 | Accepted 5 Feb 2015 | Published 5 Mar 2015

DOI: 10.1038/ncomms7529

OPEN

Accelerated exploration of multi-principal element alloys with solid solution phases

O.N. Senkov[1], J.D. Miller[1], D.B. Miracle[1] & C. Woodward[1]

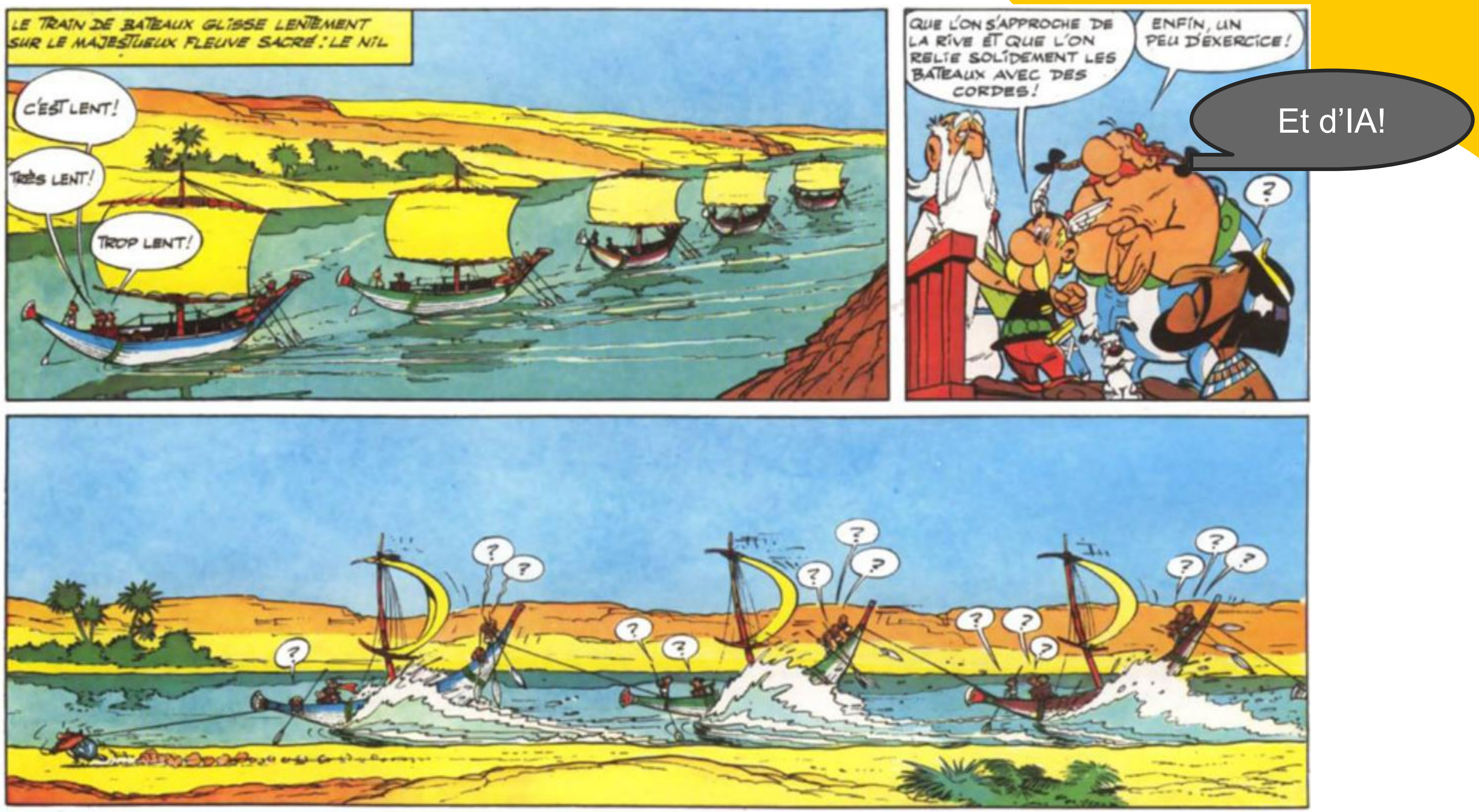
Recent multi-principal element, high entropy alloy (HEA) development strategies vastly expand the number of candidate alloy systems, but also pose a new challenge—how to rapidly screen thousands of candidate alloy systems for targeted properties. Here we develop a new approach to rapidly assess structural metals by combining calculated phase diagrams with simple rules based on the phases present, their transformation temperatures and useful microstructures. We evaluate over 130,000 alloy systems, identifying promising compositions for more time-intensive experimental studies. We find the surprising result that solid solution alloys become less likely as the number of alloy elements increases. This contradicts the major premise of HEAs—that increased configurational entropy increases the stability of disordered solid solution phases. As the number of elements increases, the configurational entropy rises slowly while the probability of at least one pair of elements favouring formation of intermetallic compounds increases more rapidly, explaining this apparent contradiction.

According to this theory High entropy alloy should be more likely:

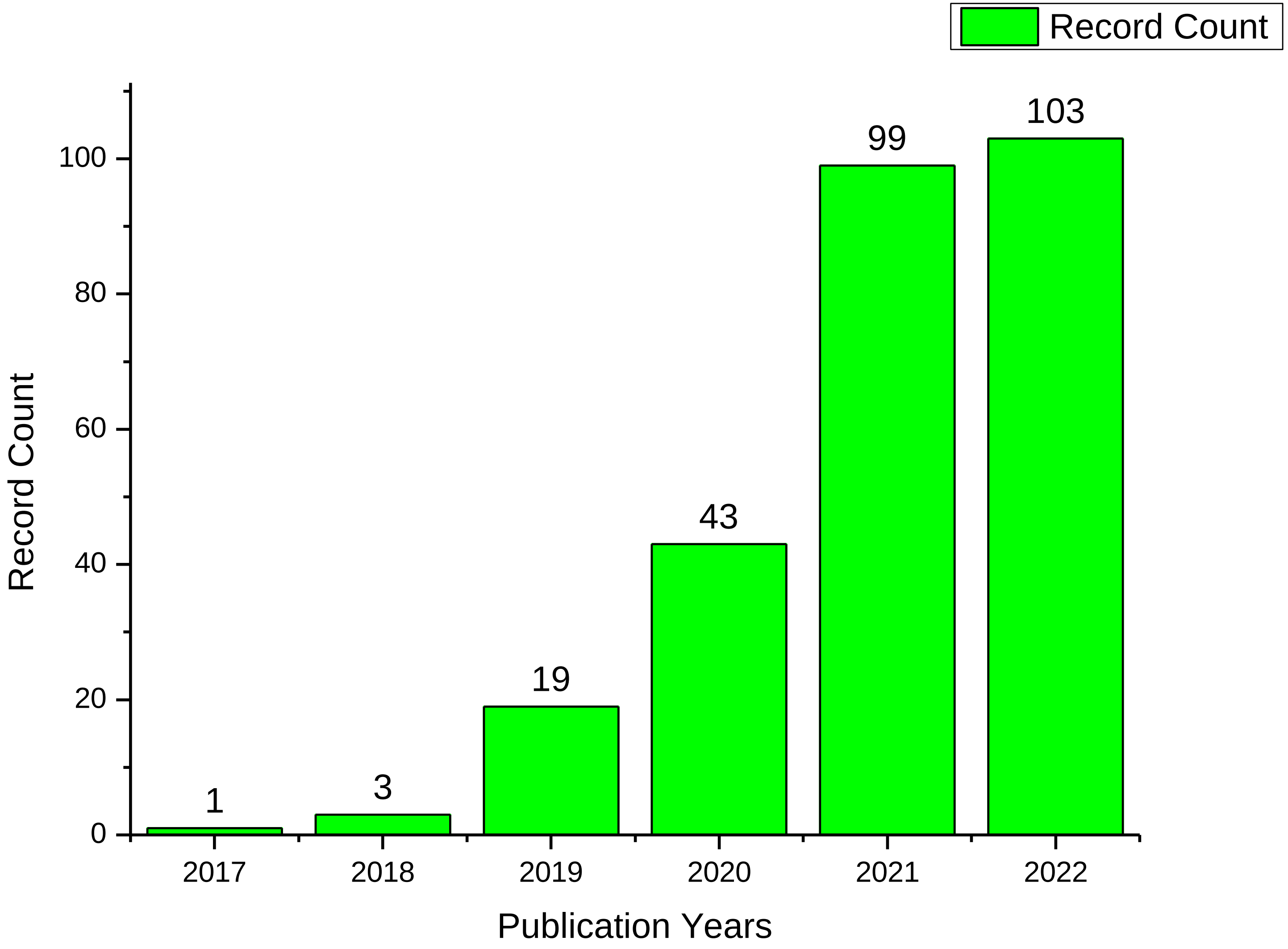
~~a. At equimolar composition~~

~~b. For higher number of alloying element~~

Materials discover without AI



The start of Machine Learning



- Source: Web of Science
- Keywords 'Machine Learning' & 'High Entropy Alloys'

ARTICLE OPEN

Check for updates

Machine-learning informed prediction of high-entropy solid solution formation: Beyond the Hume-Rothery rules

Zongrui Pei[1,2], Junqi Yin[3], Jeffrey A. Hawk[1], David E. Alman[1] and Michael C. Gao[1,4]

The empirical rules for the prediction of solid solution formation proposed so far in the literature usually have very compromised predictability. Some rules with seemingly good predictability were, however, tested using small data sets. Based on an unprecedented large dataset containing 1252 multicomponent alloys, machine-learning methods showed that the formation of solid solutions can be very accurately predicted (93%). The machine-learning results help identify the most important features, such as molar volume, bulk modulus, and melting temperature. As such a new thermodynamics-based rule was developed to predict solid–solution alloys. The new rule is nonetheless slightly less accurate (73%) but has roots in the physical nature of the problem. The new rule is employed to predict solid solutions existing in the three blocks, each of which consists of 9 elements. The predictions encompass face-centered cubic (FCC), body-centered cubic (BCC), and hexagonal closest packed (HCP) structures in a high throughput manner. The validity of the prediction is further confirmed by CALculations of PHAse Diagram (CALPHAD) calculations with high consistency (94%). Since the new thermodynamics-based rule employs only elemental properties, applicability in screening for solid solution high-entropy alloys is straightforward and efficient.

npj Computational Materials (2020)6:50 ; https://doi.org/10.1038/s41524-020-0308-7

AI Challenges

AI for HEA

Small Data

The largest data set contains less than 2000 entries

Expensive Data

Create new entry experimentally takes about 2 days, requires 3 instruments (synthesis apparatus, X-Ray diffractometer and EDS analysis). The cost can be roughly estimated around **100-1000€** per entry

Explainable AI

AI results must contribute to the elaboration of a new HEA theory

Moderate performances

70% accuracy would lead to spectacular experimentalist efficiency improvement

False positive are OK but not the false negative

05

First results

Accuracy

npj | Computational Materials

www.nature.com/npjcompumats

ARTICLE OPEN

Machine learning guided appraisal and exploration of phase design for high entropy alloys

Ziqing Zhou[1], Yeju Zhou[2], Quanfeng He[1], Zhaoyi Ding[1], Fucheng Li[1] and Yong Yang[1,3*]

```
# Model Accuracy, how often is the classifier correct?
print("Accuracy:",metrics.accuracy_score(y_test, y_pred))
```

```
Accuracy: 0.9662921348314607
```

96%

Accuracy


"That's all Folks!"

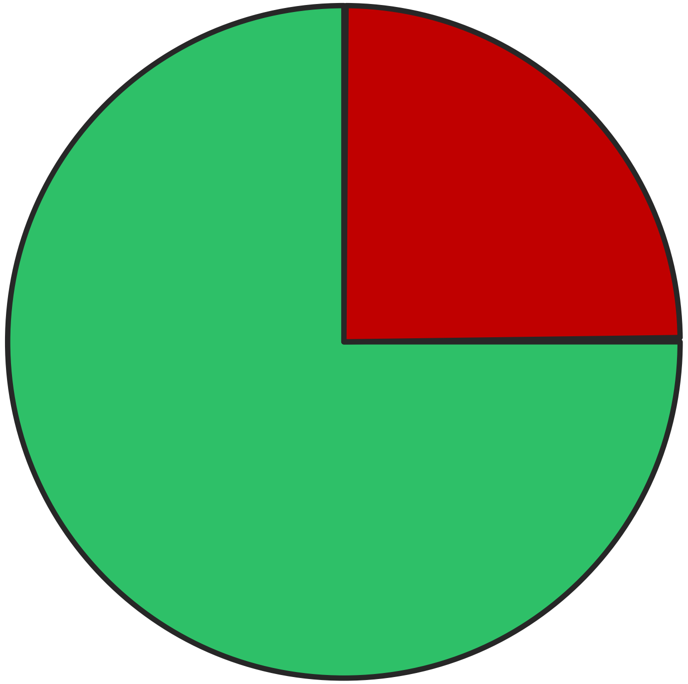
Why then, no significantly new HEA has been predicted by AI so far?

Reason 1: ratio HEA vs. Non HEA

Dataset 1

No of entries: 650 000
Proportion of HEA: 4.6%
Method: DFT
Representation of elements: All combination of 40 éléments
Source: Chen et al. *Nat. Comm (2023)*

Dataset 2



No of entries: 1600
Proportion of HEA: 75%
Method: Experiment
Representation of elements: Overepresentation of Fe, Co, Mn, V, Ti and refractory metals (Ta, W, Zr, Hf)
Source: Machaka *et al. Data in Brief (*2021)

Reason 1: ratio HEA vs. Non HEA

Dataset 1

No of entries: 650 000
Proportion of HEA: 4.6%
Method: DFT
Representation of elements: All combination of 40 éléments
Source: Chen et al. *Nat. Comm (2023)*

Learning: 80% of Dataset 1

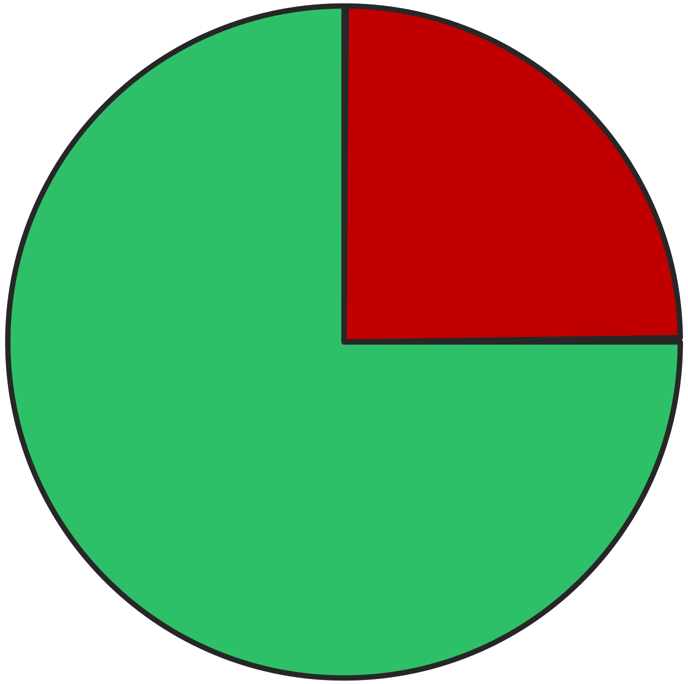
Predicting: 20% of Dataset 1

Accuracy: ~ 95%

Explanation: The model predicts that no HEA exists!

Reason 1: ratio HEA vs. Non HEA

Dataset 2



No of entries: 1600
Proportion of HEA: 75%
Method: Experiment
Representation of elements: Overepresentation of Fe, Co, Mn, V, Ti and refractory metals (Ta, W, Zr, Hf)
Source: Machaka *et al. Data in Brief (*2021)

Learning: 80% of Dataset 2

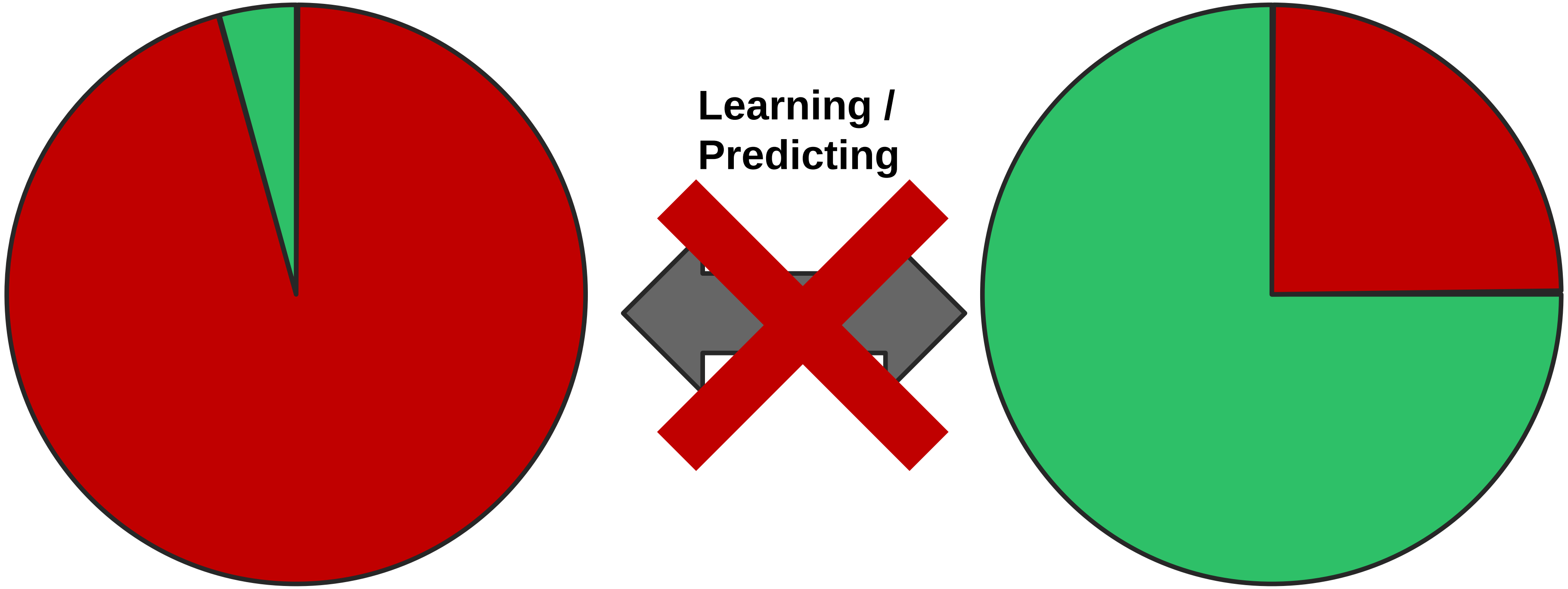
Predicting: 20% of Dataset 2

Accuracy: ~ 90%

Explanation: The model predicts a few non-HEA, otherwise it assumes these are HEAs (high false positive rate)

Reason 1: ratio HEA vs. Non HEA

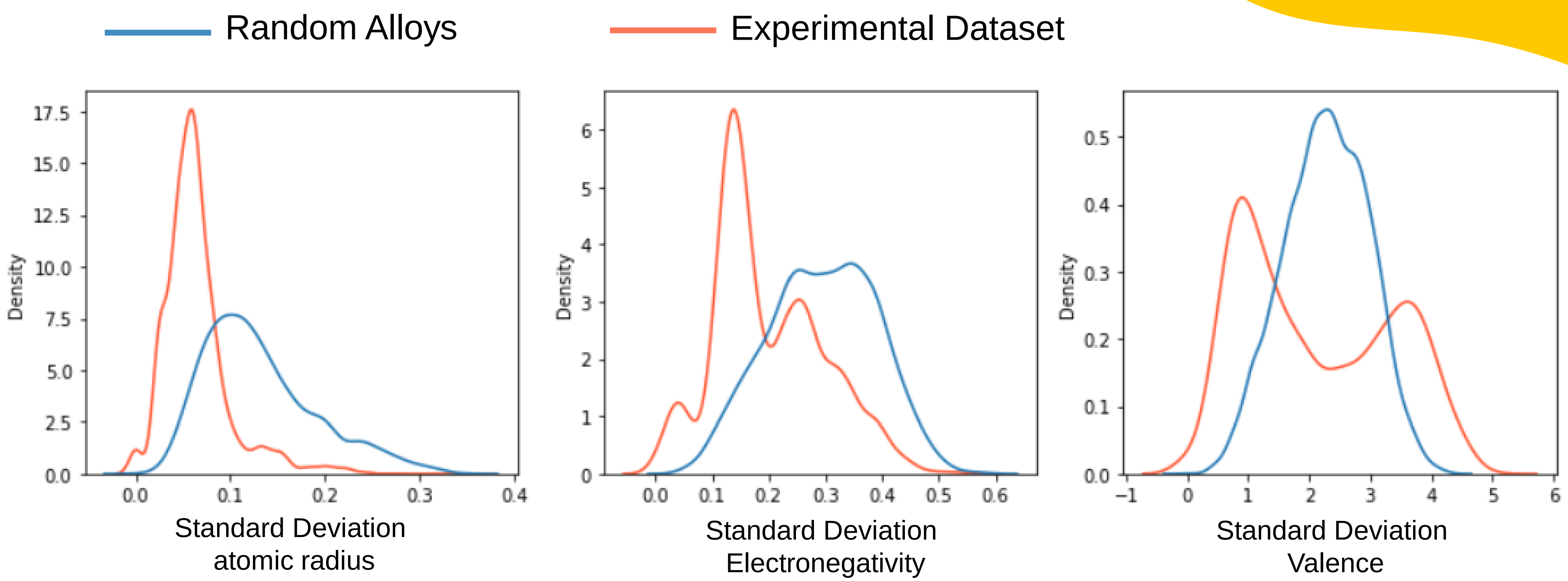
Dataset 1 **Dataset 2**



No of entries: 650 000
Proportion of HEA: 4.6%
Method: DFT
Representation of elements: All combination of 40 éléments
Source: Chen et al. *Nat. Comm (2023)*

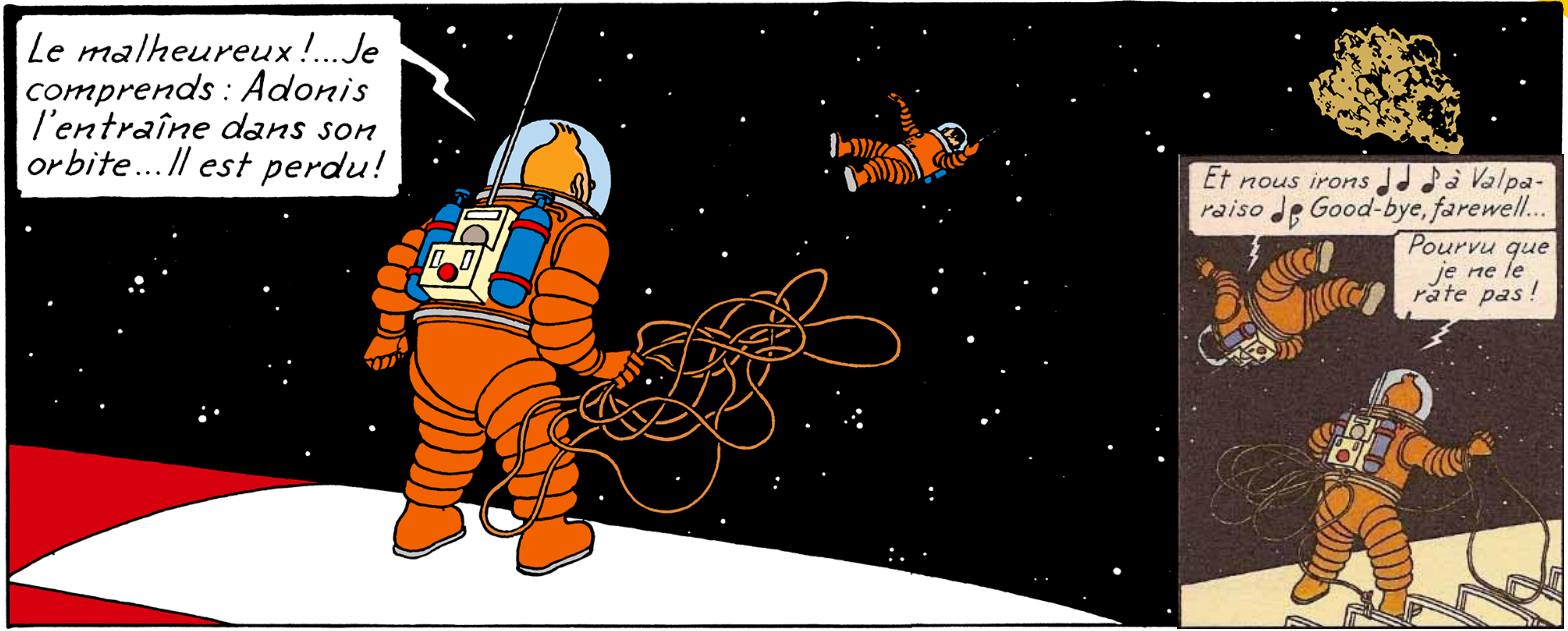
No of entries: 1600
Proportion of HEA: 75%
Method: Experiment
Representation of elements: Overepresentation of Fe, Co, Mn, V, Ti and refractory metals (Ta, W, Zr, Hf)
Source: Machaka *et al. Data in Brief (*2021)

Reason 2: poor coverage of the parameter space



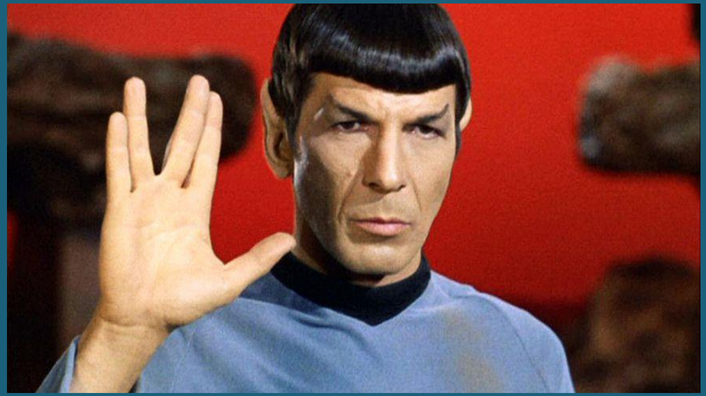
Experimental Dataset is NOT representative

AI with wrong dataset….




- HERGÉ -
LES AVENTURES DE
TINTIN
ON A MARCHÉ
SUR LA LUNE
casterman

How to continue the HEA Exploration?



Do it fast!

No Data – No AI!

1. AI needs experiment!
2. Experiment are long and expensive → Selection of the region of interest is of the utmost importance!
    a. Cover the parameter space: accept risky experiment, or to fail voluntarily!
    b. Focus experiment on materials with a substitution potential (more sustainable)

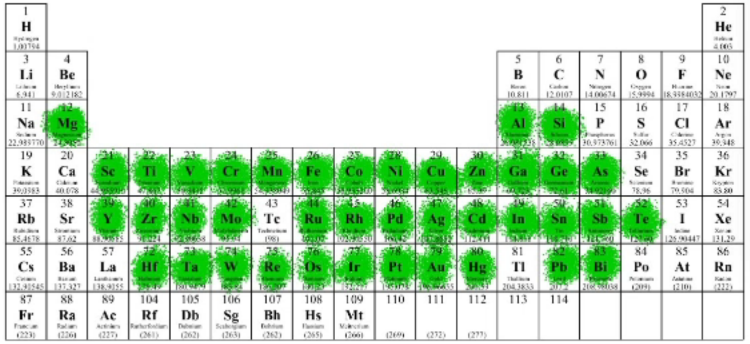
Sustainability Assessment

Calculate sustainability 👇

Please enter the alloy as the following form : `An1Bn2Cn3`

TiPbFeCuAl

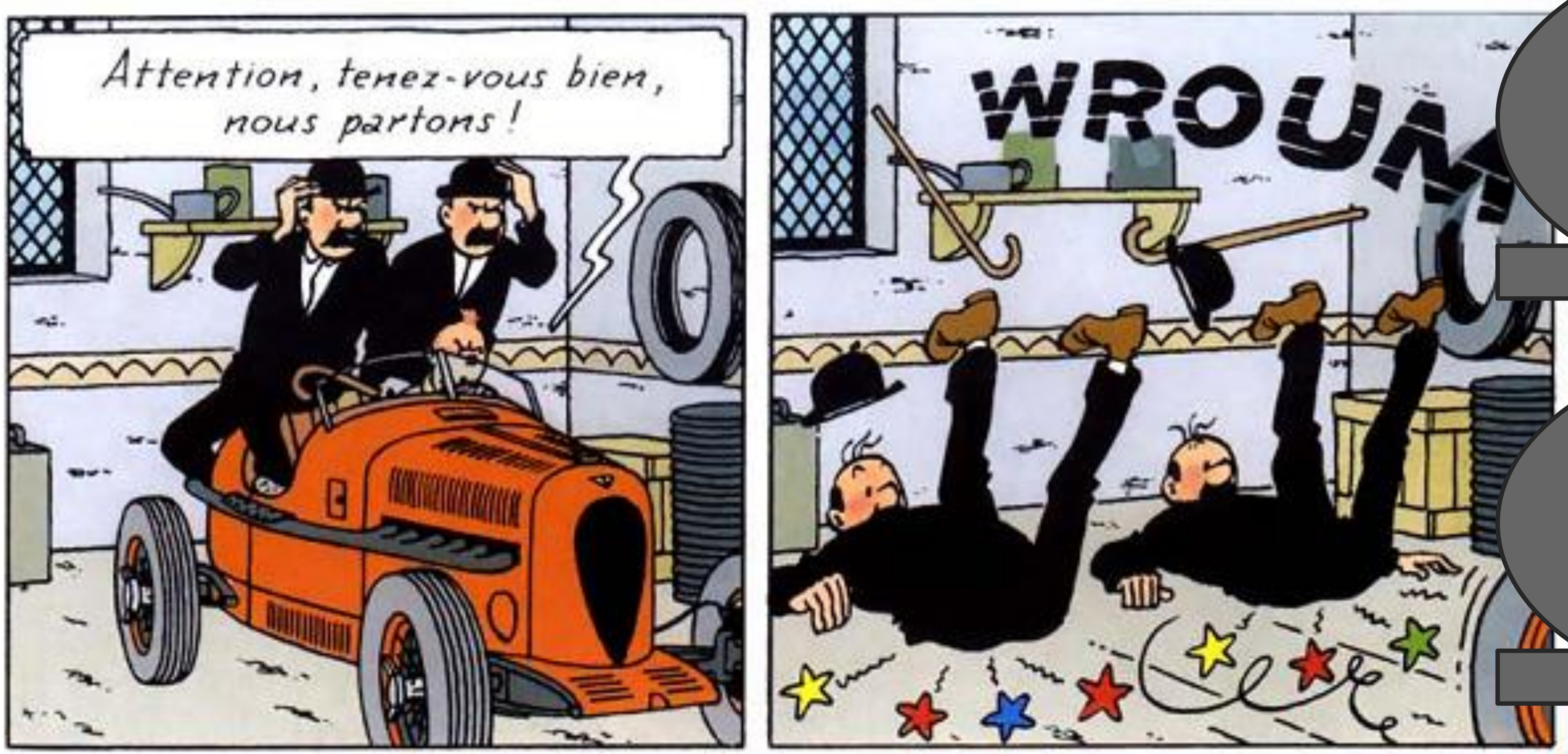
Calculate



Only colored elements are accepted

Carbon Footprint (g CO2/… 0.65 <10%	ESG risk 2.98 <10%	Maximal Companionality 10 <10%	Most critical element Ti in term of production
Energy Footprint (MJ/mol) 10.51 <10%	Supply risk 0.87 <10%	Maximal Production sollicitat… 3.95e-05 <10%	Most critical element Pb in term of reserve
Herfindahl-Hirschman In… 0.26 <10%	Average Companionality 4.0 <10%	Maximal Reserve sollicitation… 1.2e-06 <10%	Maximal Market size 2530000.0 <90%

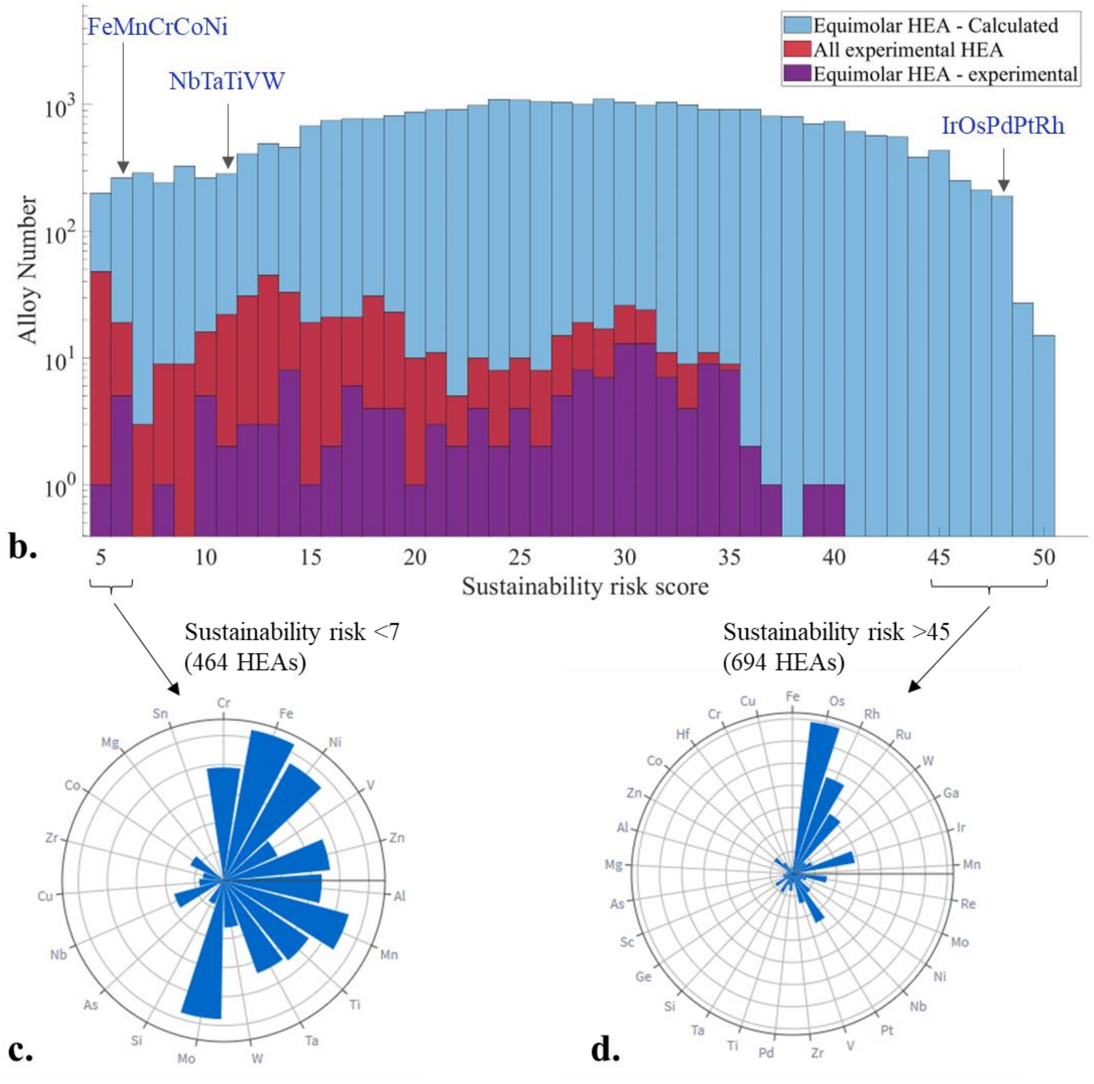
Materials development without sustainability assessement



Je t’avais pourtant bien dit Dupond que mettre de l’Osmium dans un matériaux de structure n’était pas une bonne idée

Saperlipopette Dupont, On avait pourtant mis septante millions de francs (belges) dans cette manip!

Sustainable HEA by Design

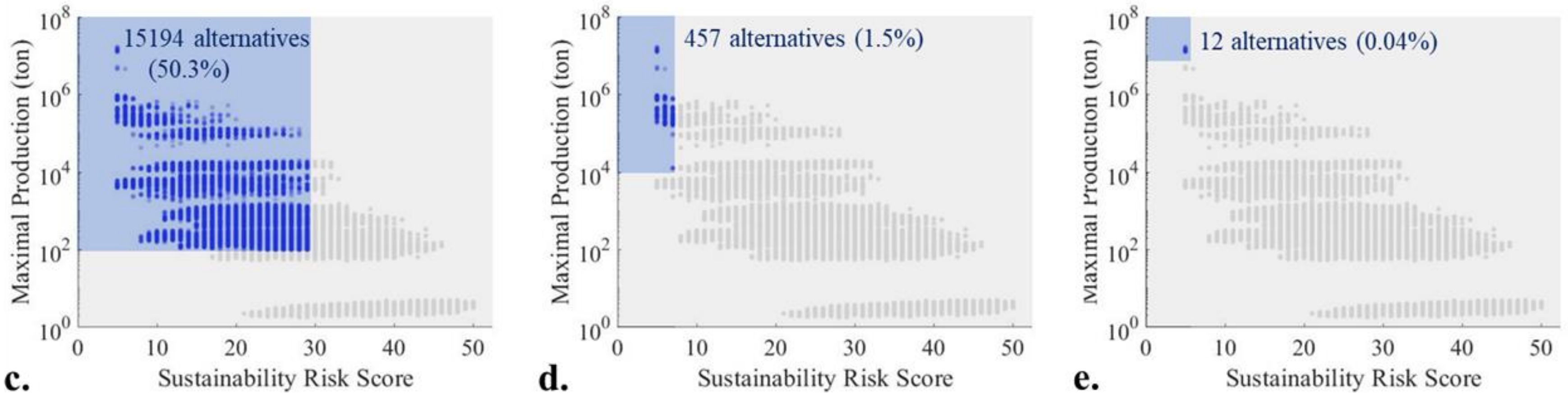


Only 6 of the 464 most sustainable predicted HEA systems have been synthesised!

Sustainable HEA by Design

Material to be substituted	Platinum	Grade 5 Titanium Alloy	304 Steel Alloy
Targeted Production (tons)	100	10000	10000000
Sustainability Risk Score	29	7	5
Possible application	H_2 Production	Aircraft Engine	Stainless Steel

b.



15000
HEA systems are candidates to substitute Platinum.

12
HEA systems to substitute 15% of Stainless Steel (3% of Steels)

Conclusions

Conclusions

Take home messages

1. The Green and Digital transition is **highly dependant on Metals**

2. Recycling & Reduction of overconsumption are of the utmost importance but **are not « magic sticks »**

3. **New materials** have to be discovered to substitute **the most critical and polluting ones**

4. AI is a fantastic **accelerator** for discovery…providing the **use of proper dataset!**

5. Materials must be assessed beyond a simplistic « cost-performance » paradigm, but with the sustainability taken into account

Acknowledgement

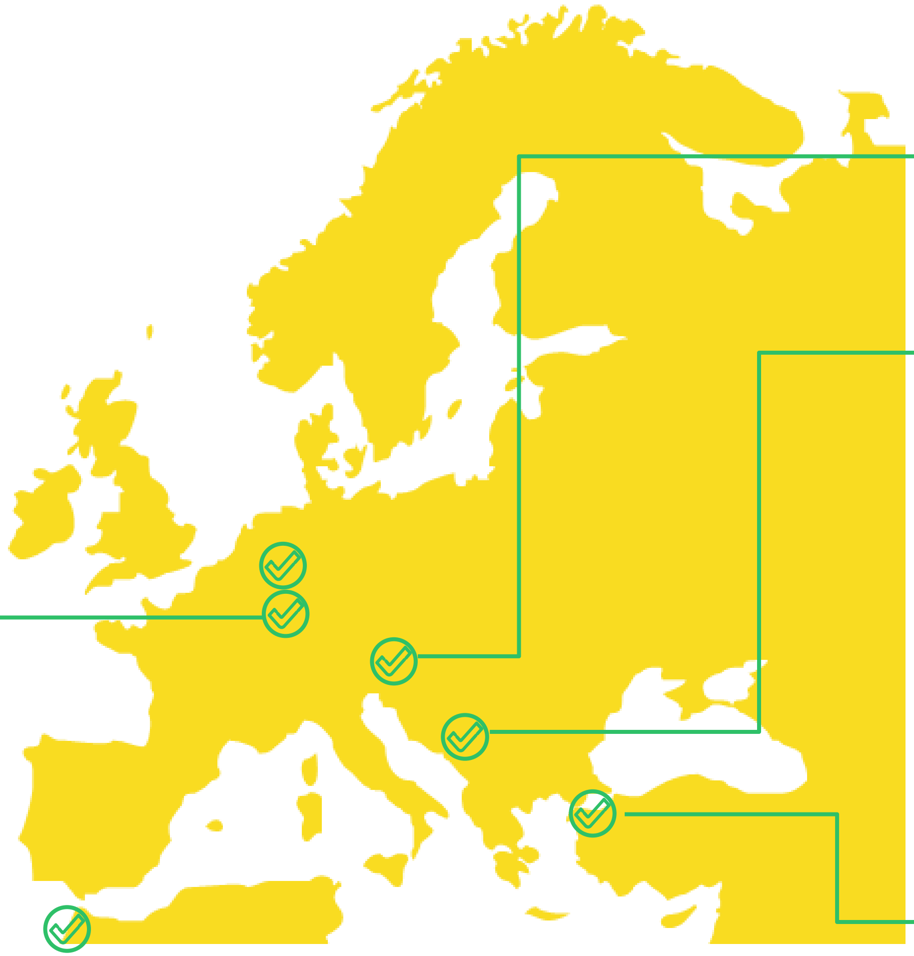
Co-funded by the
Erasmus+ Programme
of the European Union

Ecole des Mines / Institut Jean Lamour
François Rousseau
Stephane Mangin
JF Pierson
Tatiana Banderova

Loria
Anne Boyer
Chahrazed Labba

Labex R21
Alexandre Chagnes
Michel Cathelineau
Olga Chernoburova

CRAN
Eric Rondeau
Sevra Cicekli



Institute Jozef Stefan
Janez Zavasnik
Uros Cvelbar

University of Belgrade
Tijana Vulevic

University of Queensland
Agnes Samper

University Mohamed VI Ayyoub El Kasmi
Outhmane Ouahri

Galatasaray University
Tuncay Gürbüz

Follow us!

1. **Sustainability Assessment Software** (upon acceptance of the paper): January 2024
2. **We can be HERawS:** podcast on raw materials (November 2023)
3. **Life long learning courses:** from Spring 2024 in Ljubljana, Nancy, Belgrade and Luxemburg
4. **Our LinkedIn page:** https://www.linkedin.com/company/heraws/

Thanks for your attention

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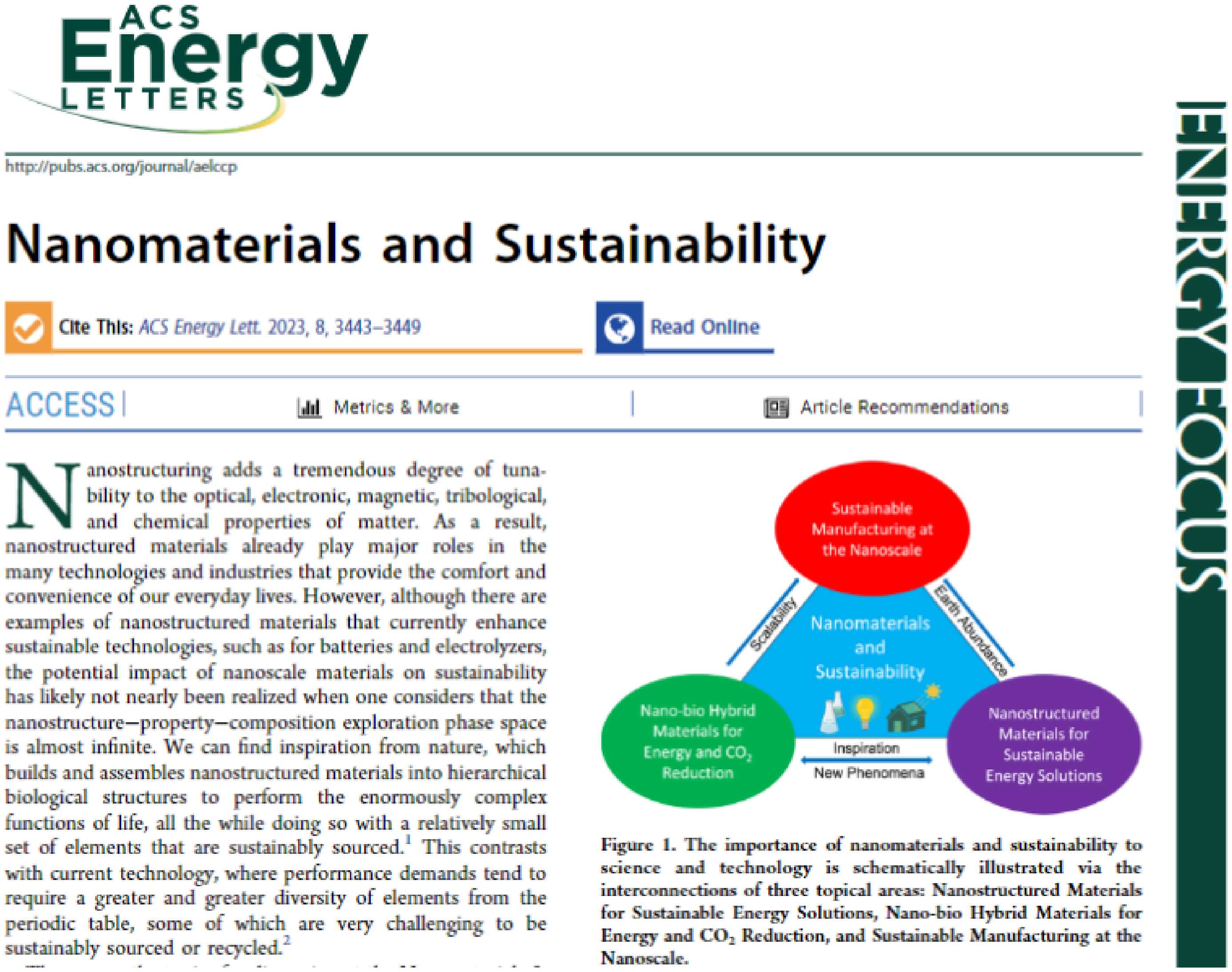

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ENERGY FOCUS

Nanomaterials and Sustainability

Cite This: *ACS Energy Lett.* 2023, 8, 3443–3449 | Read Online

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Nanostructuring adds a tremendous degree of tunability to the optical, electronic, magnetic, tribological, and chemical properties of matter. As a result, nanostructured materials already play major roles in the many technologies and industries that provide the comfort and convenience of our everyday lives. However, although there are examples of nanostructured materials that currently enhance sustainable technologies, such as for batteries and electrolyzers, the potential impact of nanoscale materials on sustainability has likely not nearly been realized when one considers that the nanostructure—property—composition exploration phase space is almost infinite. We can find inspiration from nature, which builds and assembles nanostructured materials into hierarchical biological structures to perform the enormously complex functions of life, all the while doing so with a relatively small set of elements that are sustainably sourced.[1] This contrasts with current technology, where performance demands tend to require a greater and greater diversity of elements from the periodic table, some of which are very challenging to be sustainably sourced or recycled.[2]

Figure 1. The importance of nanomaterials and sustainability to science and technology is schematically illustrated via the interconnections of three topical areas: Nanostructured Materials for Sustainable Energy Solutions, Nano-bio Hybrid Materials for Energy and CO_2 Reduction, and Sustainable Manufacturing at the Nanoscale.

Conference given for the US Department of Energy and published at the American Chemical Society (link here)