Óbudai Egyetem

Progress presentation of the 1st semester (September 2023 – January 2024)

Toughening of high-entropy ceramics by low-dimensional nanomaterials

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Research Objectives of this PhD study

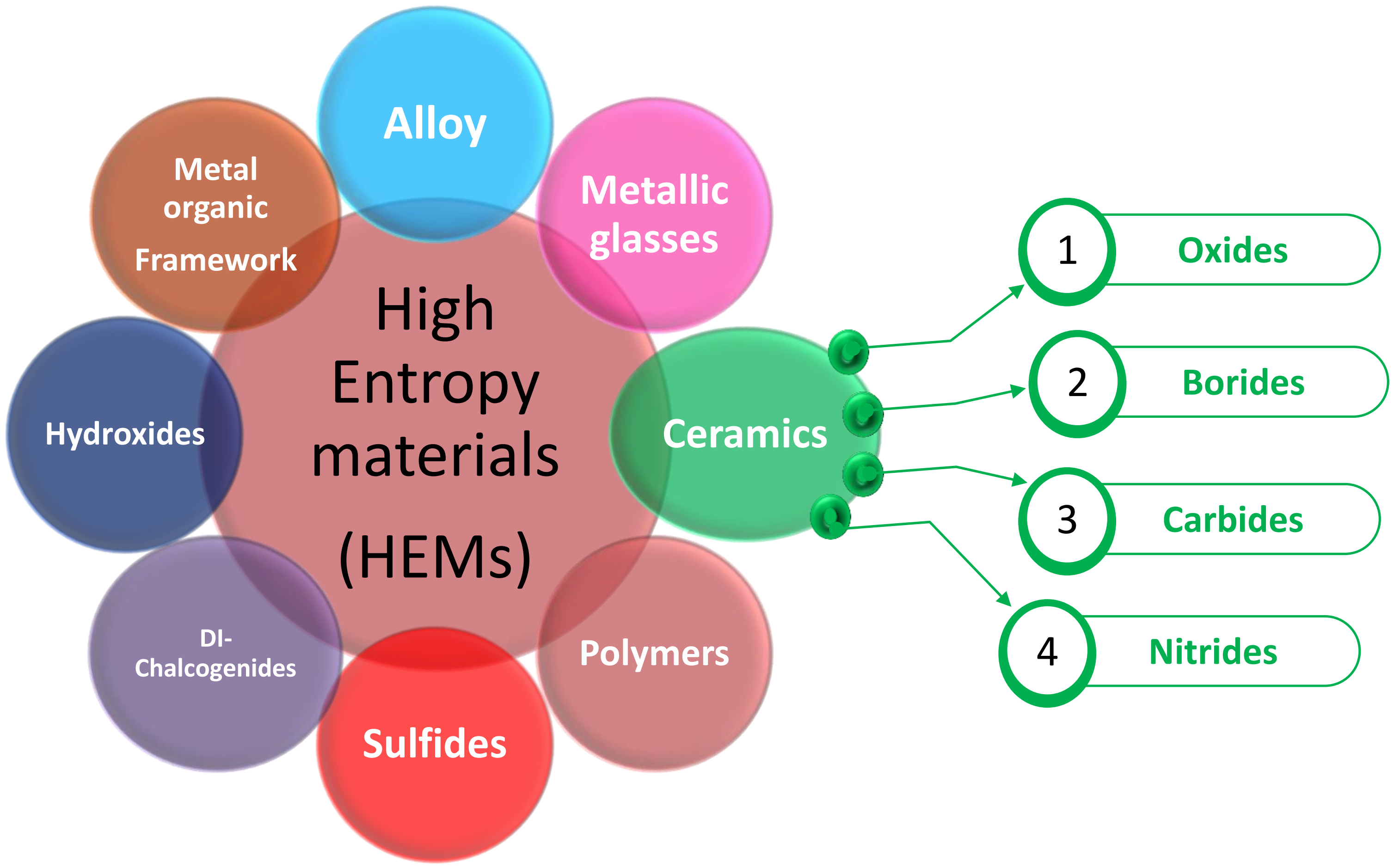
- Study the possible ways for toughening of high – entropy ceramics.
- Preparation of dual–phase high entropy ceramics with multilayer graphene and carbon fiber additives.
- Investigation the microstructure characteristics mechanical, tribological and high-temperature properties of developed systems.
- Optimization of the processing parameters to obtain suitable fracture toughness, mechanical and tribological properties of developed systems.

Introduction

- Driven by an age-old curiosity, our modern quest for material mastery explores High-Entropy Ceramics (HECs)

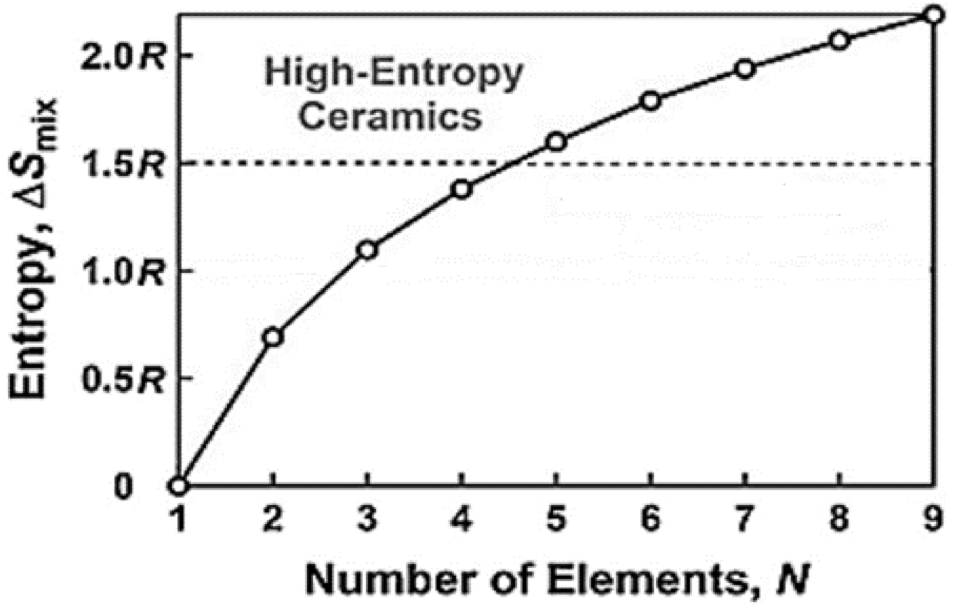
- A groundbreaking fusion of transition metal (Zr, Hf, Ti, Ta etc.) oxides, borides, carbides, and nitrides.

- In (nearly) equal proportions, offering unparalleled properties for extreme environments - temperature, chemical reactivity, mechanical stress, wear, radiation...



Transition to High-Entropy Ceramics (HECs):

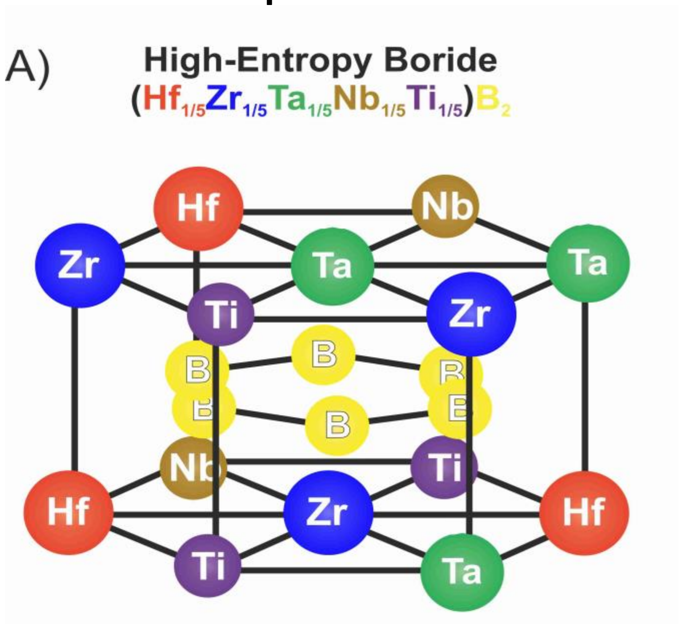
- High-entropy ceramics adhere to the dual definitions established for high-entropy alloys.
- Despite the inherent complexity of ceramic structures, high-entropy ceramics exhibit a uniform crystalline single-phase, showcasing exceptional homogeneity.
- The concept gained significant attention in 2015, marked by a clear demonstration of entropy stabilization.

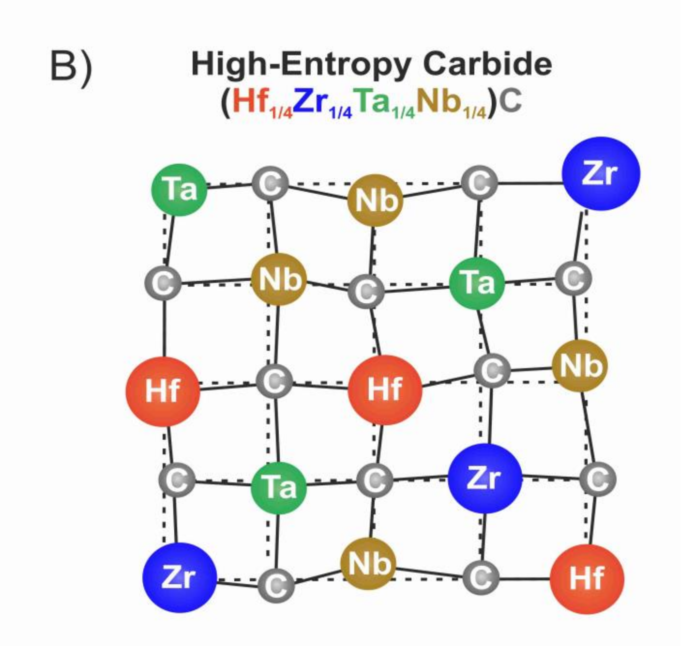


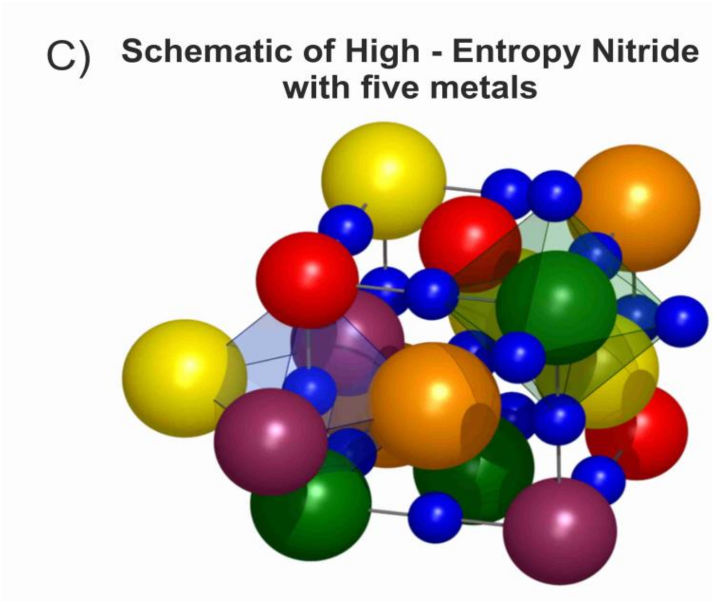
Relation between entropy mixing and number of elements and definition of high entropy ceramics

High – entropy bulk ceramics

- The development of HEC's in the last 8 years







- First published results for different HECs:

Oxide - C.M. Rost, E. Sachet, T. Borman, A. Moballegh, E.C. Dickey, D. Hou, et al., Nature Communications 6 (2015)

Boride - J. Gild, Y. Zhang, T. Harrington, S. Jiang, T. Hu, M.C. Quinn, et al., Scientific Reports 6, p.2–11, (2016)

Carbide - E. Castle, T. Csanádi, S. Grasso, J. Dusza, M. Reece, Scientific Reports 8, p.1–12, (2018)

Nitride - O. F. Dippo et all., Scientific Reports, 10:21288, (2020)

Dual Phase/Carbide + Boride - M. Qin et al., J Eurp Ceram Soc 40, p.5037–50, (2020)

Latest improvements in HECs

A notable subset, **dual-phase high-entropy ultra-high temperature ceramics (DPHE-UHTCs)**, has emerged as a potential candidate for ultra-high temperature applications.

01

The materials exhibited **superior hardness** compared to the weighted average of two single-phase high-entropy UHTCs, showcasing the tunability of microstructure and mechanical properties by adjusting phase fractions in DPHE-UHTCs.

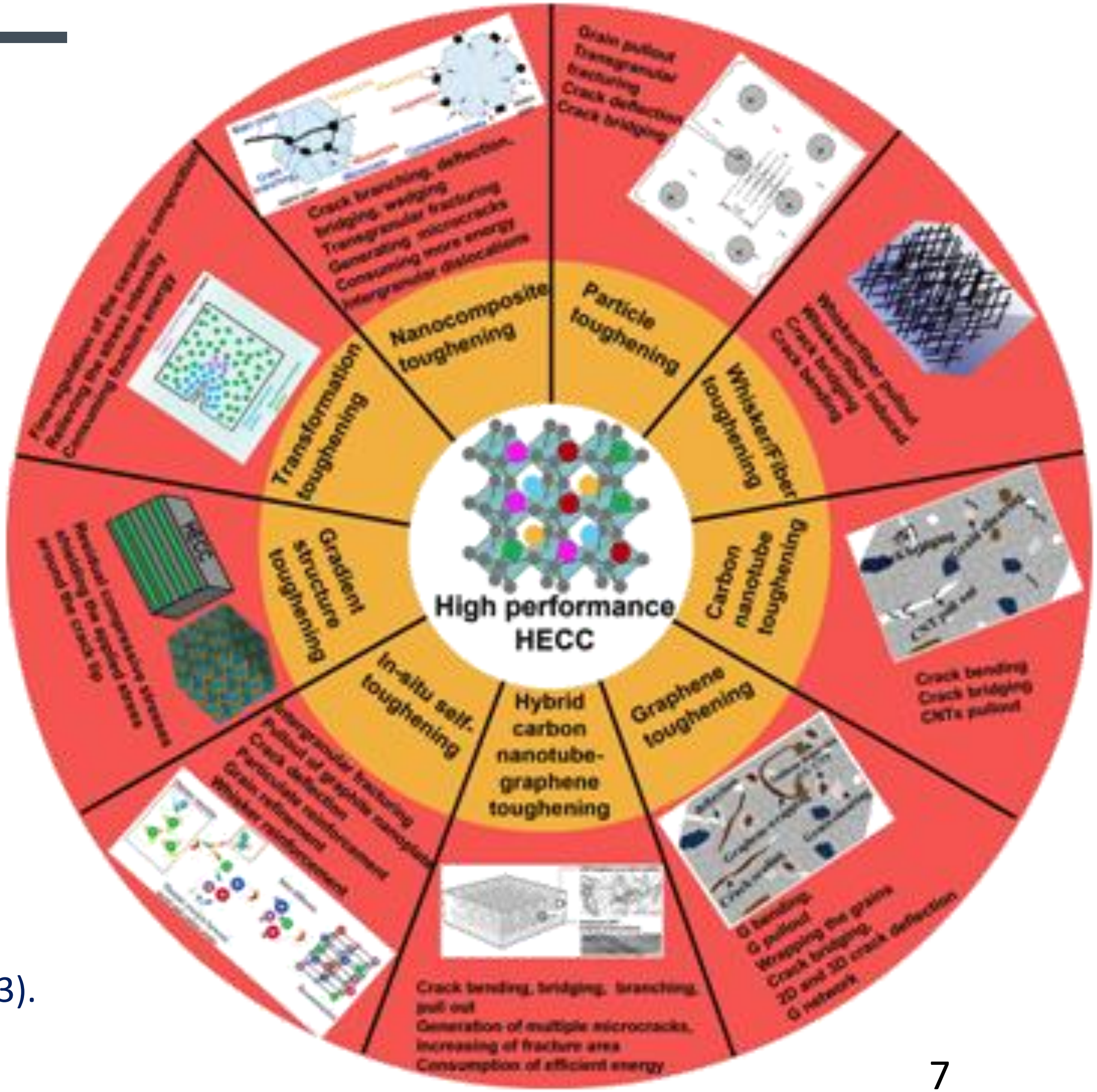
02

Nanotechnology holds potential for **optimizing the toughness of high-entropy ceramics**, facilitating the development of more resilient materials as nanoscale additives offer precise control over material characteristics

03

Toughening of high entropy ceramics

- Particle Toughening like SiC, Ni, Co, FeNi, and Mo enhance HECC toughness, requiring careful consideration to avoid compromising overall properties.
- Whisker/Fiber Toughening boosts ceramic toughness through mechanisms like pullout, bridging, and crack deflection.
- Adding raw materials enables controlled growth of uniformly distributed crystals in HECC, with advantages like no health hazards and lower sintering temperatures.
- CNTs and graphene enhance ceramic toughness, while their hybrid combination addresses toughness versus hardness/strength trade-offs in HECC, presenting ongoing research challenges and opportunities.



Cao, Z., Sun, J., Meng, L., Zhang, K., Zhao, J., Huang, Z., & Yun, X. (2023). Progress in densification and toughening of high entropy carbide ceramics. Journal of Materials Science & Technology.

Way of development

Computational approach

The Gibbs free energy calculations provide quantitative information regarding the phase stability and phase diagram. The Gibbs free energy is computed by:

- density functional theory
- molecular dynamics simulation
- machine learning
- CALPHAD and descriptors, etc.

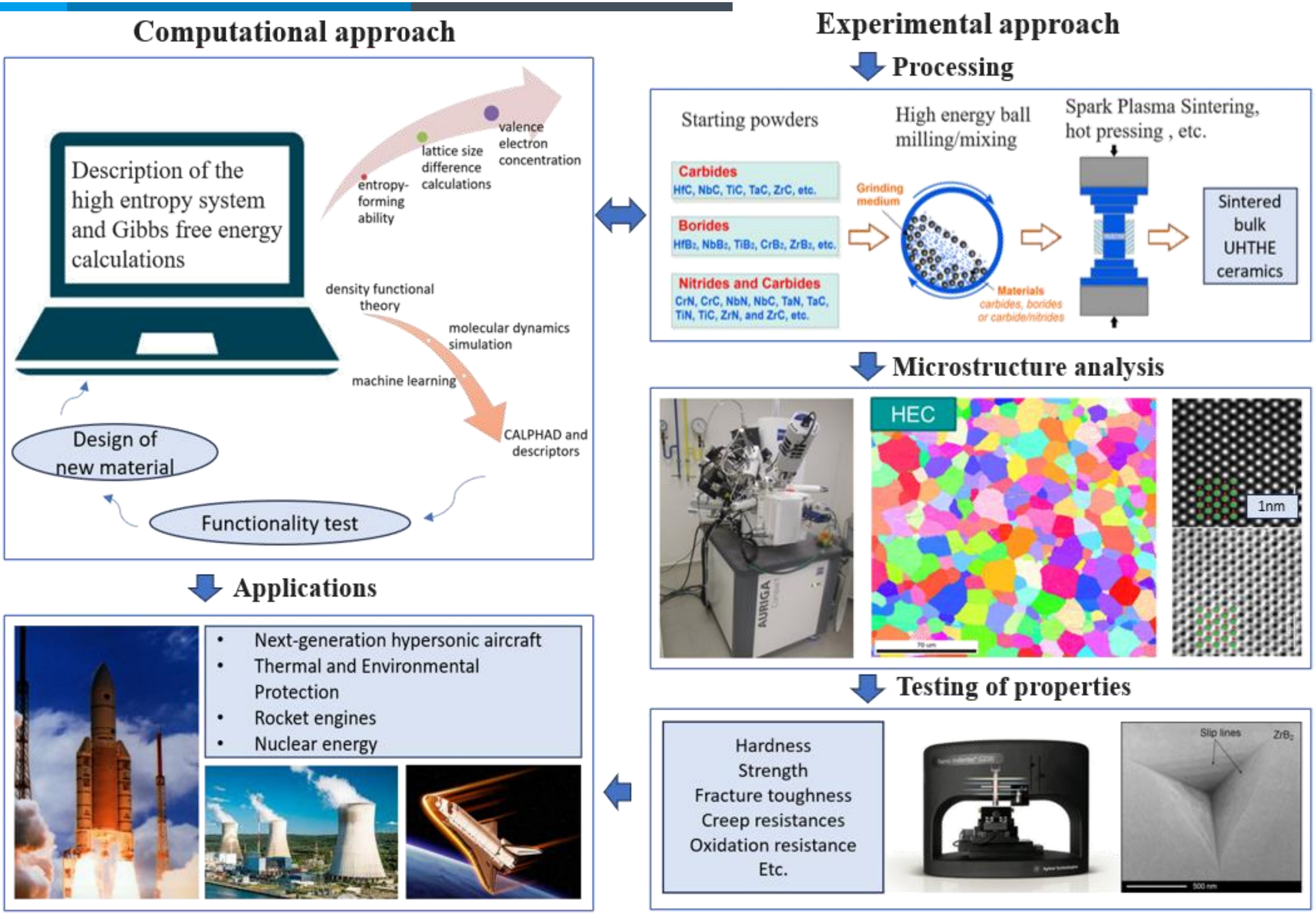
To describe the high entropy system:

- entropy-forming ability
- lattice size difference calculations
- valence electron concentrati on

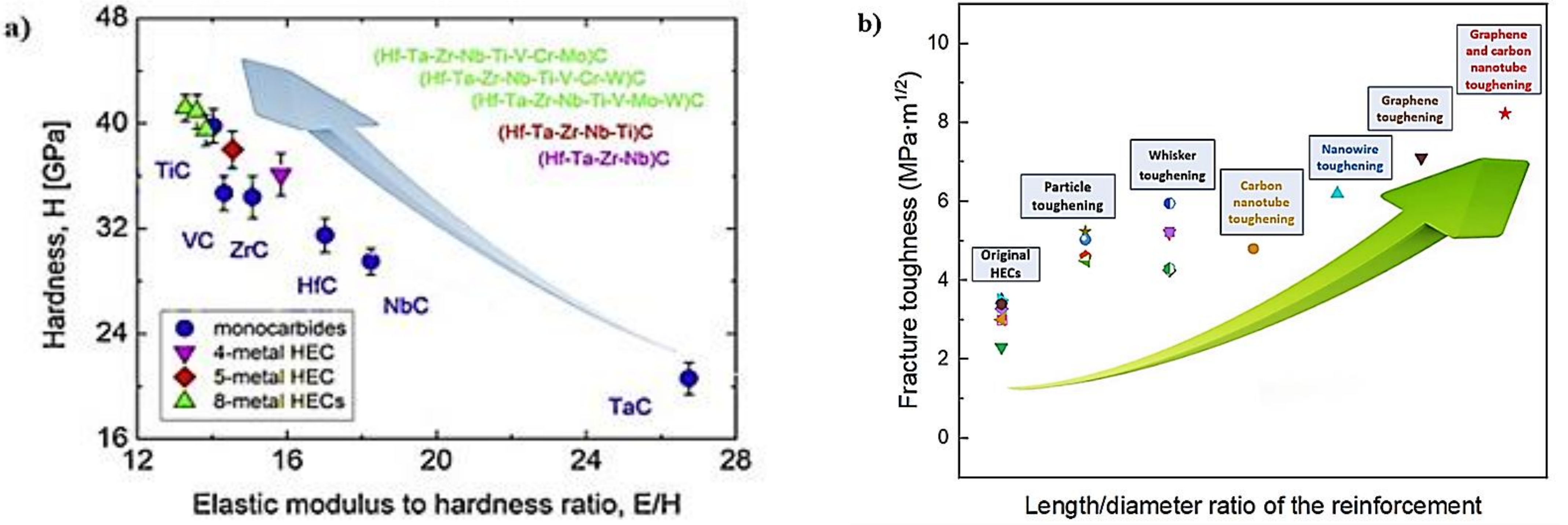
Experimental approach - processings

- Conventional solid-state reaction methods–ball milling
- High-pressure torsion
- Spark plasma sintering/hot pressing
- Reactive spark plasma sintering
- Self-propagating high-temperature synthesis

Way of development



PROPERTIES of HECs



Mechanical properties: nano/micro /macro hardness

Creep and fracture toughness

Thermal shock resistance

Corrosion and oxidation resistance

Tribological properties: friction, wear, and lubrication of interacting surfaces

Challenges

Complex compositions	Computational challenges	Property prediction discrepancies	Scale-up obstacles
• HE-UHTCs' intricate compositions challenge both computational and experimental studies	• Modeling faces hurdles due to numerous chemical species, demanding improved predictions for thermal transport and oxidation.	• Accurate computational predictions of thermal and electrical properties remain challenging, causing disparities with measured values.	• Understanding synthesis and densification mechanisms is crucial for successful scale-up, addressing issues like impurity levels and nonuniform microstructures.

- Addressing the hindered practical application of HECs, our future work focuses on improving fracture toughness, strength, and wear resistance.

- This involves the development of high-density dual-phase boride/carbide HEC composites reinforced with nanomaterials.

Publications and future work

1. Publication: Sara Ines Moussaoui, Péter Pinke, János Dusza: High Entropy Ceramics: A Brief Introduction, Engineering Symposium at Bánki (ESB 2023) http://bgk.uni-obuda.hu/esb/

2. Planned publication (in progress): A review article of Development of Dual-Phase Ultra-High Temperature High Entropy Ceramics

3. In collaboration with Instute of materials research SAS, Kosice, Slovakia - Processing of dual-phase high-entropy ceramic (Ti-Zr-Nb-Ta-Hf)C/(Ti-Zr-Nb-Ta-Hf)B2 boride/carbide system (HEC/HEB) based composites with graphene and carbon micro-fiber additives with different processing parameters as sintering temperature, time and pressure.

4. The microstructure characteristics will be studied by X – ray diffraction, scanning electron microscopy (SEM), aberration-corrected scanning transmission electron microscopy (STEM), energy dispersive X-ray spectroscopy (EDS) and electron energy loss spectroscopy (EELS).

5. Basic mechanical properties as hardness, indentation fracture resistance etc., will be measured.

6. Presentation of the results at conferences and meetings.

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Thank you for your attention