High-Entropy Carbide Ceramics for Extreme Environments

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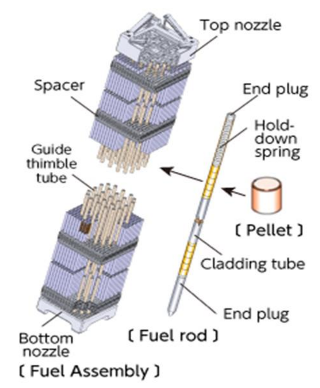
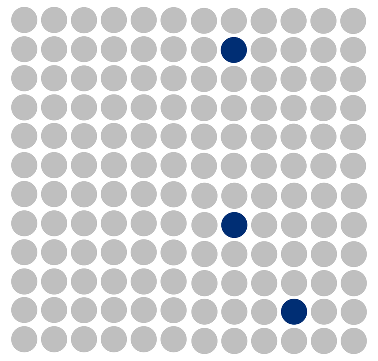
Motivation

- **Use the 'high-entropy' concept to develop novel ceramic materials for the extreme environments in next-generation nuclear energy systems**
- **Potential applications: fuel cladding, structural component**
- **High melting temperature, high irradiation damage resistance, high-temperature strength, thermal shock resistance and corrosion resistance.**

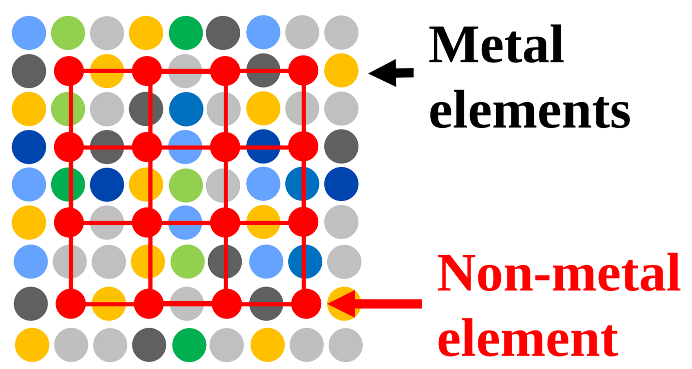


Background



Traditional alloys
Example: Steel

High-entropy alloys
Example: CoCrFeMnNi

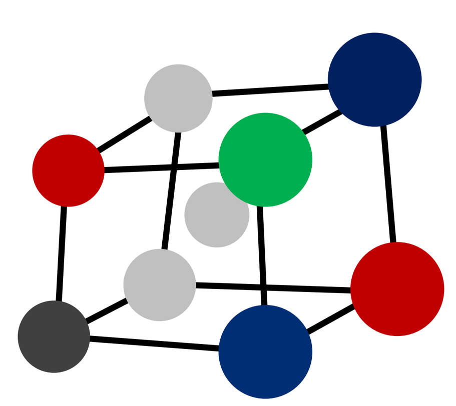


High-entropy ceramics
Example: $(Hf_{0.2}Zr_{0.2}Ta_{0.2}Nb_{0.2}Ti_{0.2})C$

- **Traditional alloys**: base element and trace amount of other elements.
- **High-entropy alloys**: multiple principal elements mixed in an equimolar or near equimolar composition but form a single BCC, FCC or HCP structure.
- **High-entropy ceramics**: single-phase structure with disorder of multiple metal elements at cation positions, ordered C, B or O elements at the anion positions.

High-Entropy Effects

1. **Thermodynamically more stable**
2. **Lattice distortion**
3. Sluggish diffusion
4. Cocktail effect



Lattice distortion

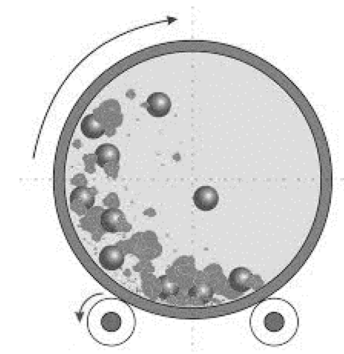
Configuration entropy of mixing per mole:

$$\Delta S_{mix} = -R\sum_{i-1}^{n} c_i \ln c_i \quad G = H - TS$$

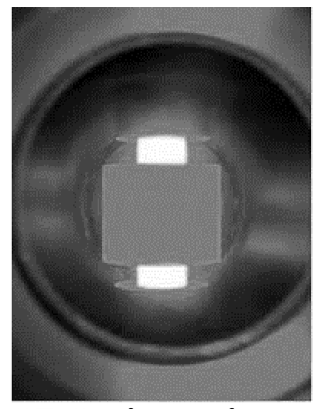
Results

1. Synthesis of $(Hf_{0.2}Zr_{0.2}Ta_{0.2}Nb_{0.2}Ti_{0.2})C$

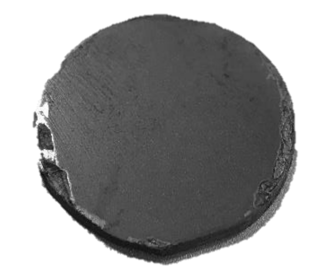
ZrC, TiC, NbC, TaC and HfC powders mixed with an equimolar composition



Ball milling
250 rpm/6 hours/Ar

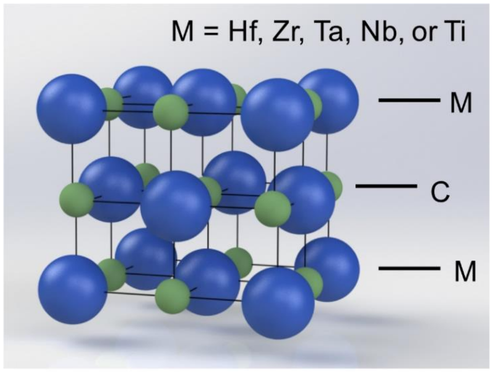


SPS sintering
2000°C/30 MPa
5 min/vacuum

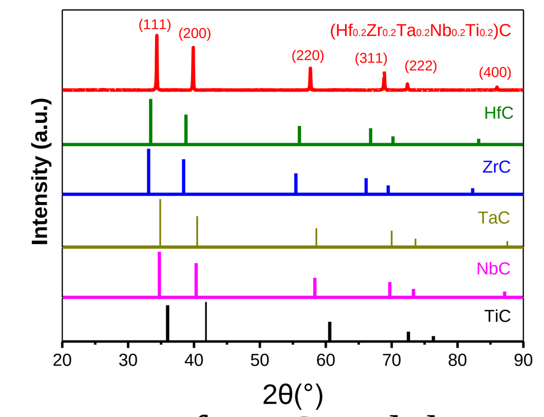


Sintered sample:
Size:⌀ 20*3 mm

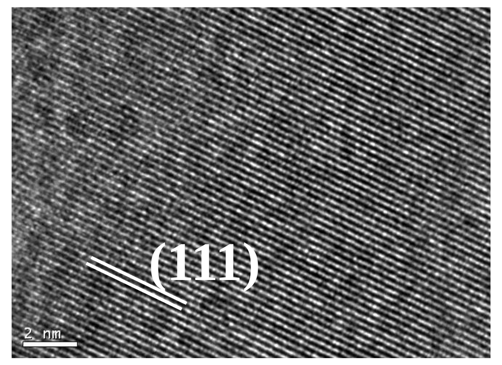
2. Single phase rock-salt crystal structure of HEC



Schematic of the rock salt structure of HEC



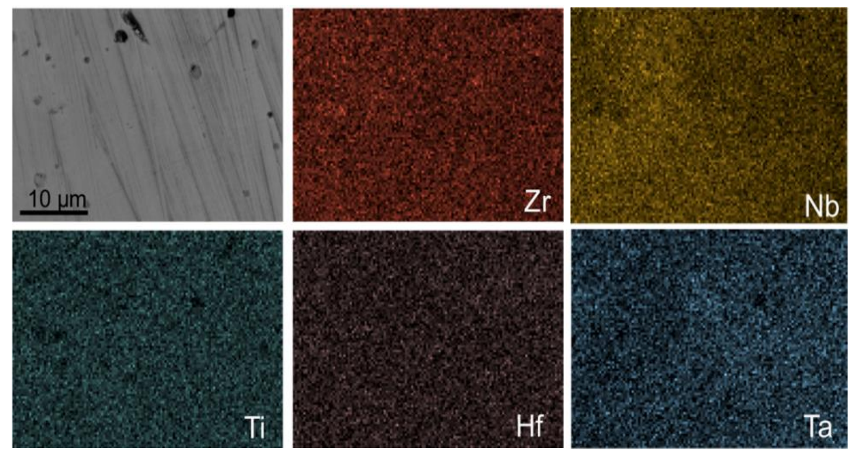
XRD of HEC and the 5 binary carbides



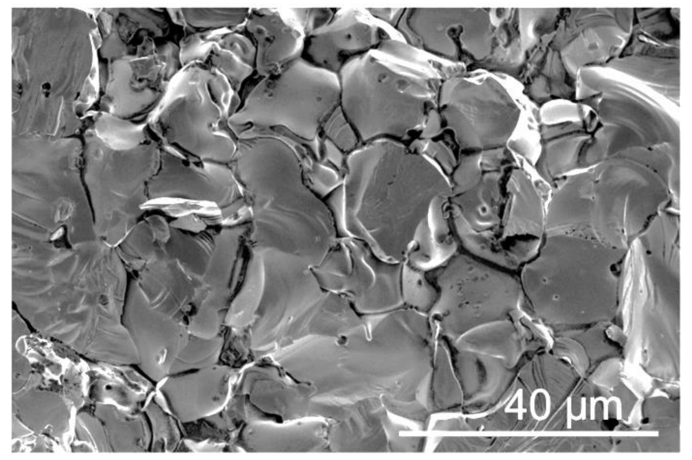
High-resolution TEM

Lattice parameter: 4.518 Å, which is close to the average lattice parameter (4.513 Å) of 5 binary carbides.

3. Homogenous elements distribution in HEC



Microscale EDS of HEC

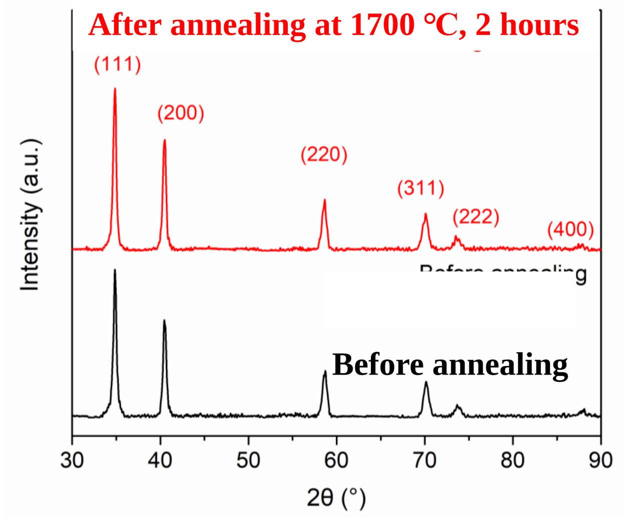


SEM image of the grains of HEC

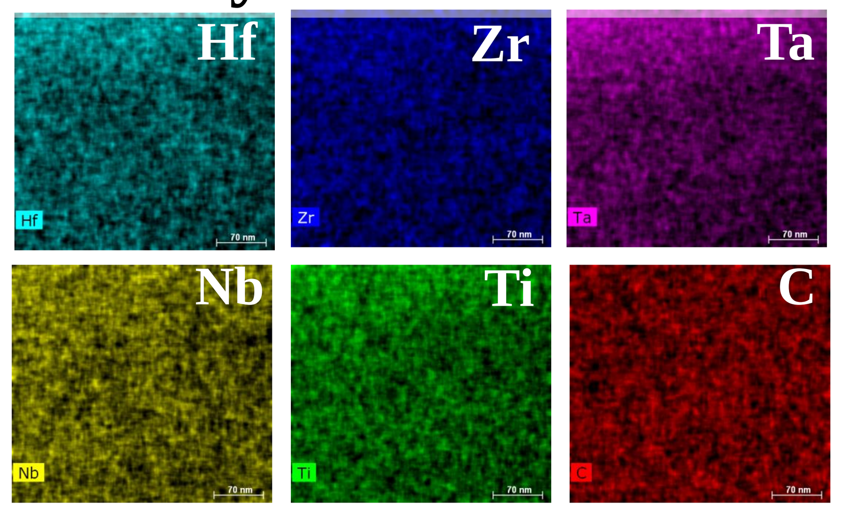
4. Ultra-high nanoindentation hardness of HEC

Materials	Elastic modulus, *E*(GPa)	Vickers hardness, H_V(GPa)	Nanoindentation hardness (GPa)
HEC	**479**	**15**	**48.9**
HfC	**450-500**	**18.3**	**29.0**
ZrC	**464**	**17.6**	**32.5**
TaC	**458**	**13.9**	**18.6**
NbC	**392**	**22.1**	**24.5**
TiC	**448**	**21.87**	**25.6**

5. High temperature thermal stability of HEC



XRD before and after annealing

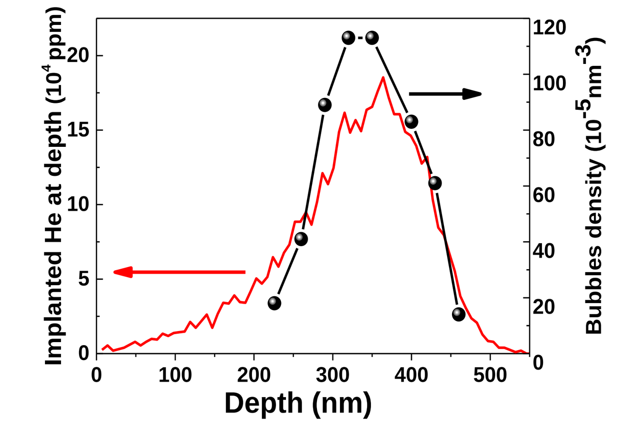


EDS of sample after annealing at 1700 °C

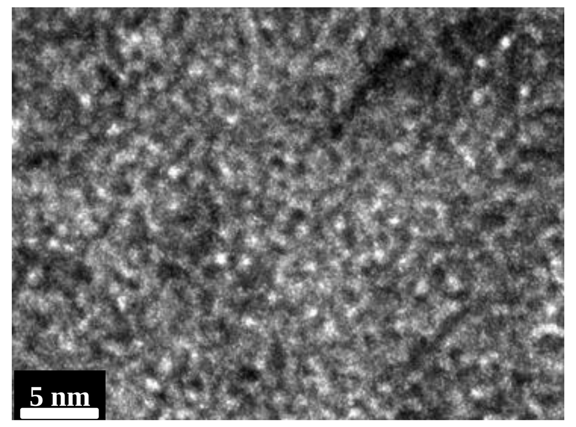
6. Low thermal diffusivity and conductivity of HEC

Materials	Thermal diffusivity, α (mm^2/s)	Thermal conductivity, *k* (W/m·K)	Thermal expansion coefficient, $\alpha_L(10^{-6}\,K^{-1})$
HEC	**3.6**	**6.45**	**6.44**
NbC	**6.29**	**6.3**	**6.65**
TiC	**8.32**	**22.2**	**6.99**
HfC	**12.3**	**29.3**	**6.6**
TaC	**12.4**	**33.5**	**6.29**
ZrC	**15.2**	**33.5**	**6.74**

7. Good irradiation resistance of HEC



SRIM-calculated helium concentration and the measured number density of helium bubbles



Bright-field TEM image of helium bubbles

Conclusions

1. **Single-phase $(Hf_{0.2}Zr_{0.2}Ta_{0.2}Nb_{0.2}Ti_{0.2})C$ was synthesized by SPS.**
2. **Thermal stability at least up to 1700°C due to the low Gibbs free energy.**
3. **Ultra-high nanoindentation hardness is due to distorted lattice.**
4. **Low thermal conductivity and diffusivity due to scattering of phonons by lattice distortions.**
5. **Helium bubble coalesce and growth is suppressed by high-entropy effect.**

Acknowledgements

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