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RADIATION DAMAGE EFFECT ON STRUCTURAL AND MECHANICAL PROPERTIES OF INERT ZrN LAYER: CORROSION MITIGATION IN LBE COOLING ENVIRONMENT

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Plan

- Status of the art,
  1. the LBE advantages compared to other liquid alloys ;
  2. Po-210 production issue;
  3. corrosion issue;
  4. compatibility with materials;
  5. proposed solution such as coatings and inhibitors in which the formed thin film of ZrN can play a key role in corrosion mitigation.
- ZrN thin film applications and performance,
- Experiments
  1. ZrN irradiation and characterization techniques
- Results and discussion
- Summary
- Perspective

Introduction

Lead-Bismuth Eutectic (LBE) present multiple advantages:

- low thermal neutron absorption cross section,
- radiation damage tolerance,
- low melting temperature (123.5°C),
- high boiling temperature (1670°C) and
- non-reactive with air and water,

Introduction

- Due to these different advantages, Lead-Bismuth Eutectic (LBE) is a primary candidate for use as a target for the production of a spallation neutron source and as the coolant for advanced nuclear reactors systems (Gen. IV).
- Thus, it has been the subject of much research since its development in the 1950s, where extensive studies were carried out across the world as part of R&D activities.

Po-210 production

- ^{210}Po production from neutron irradiation of Bismuth (^{209}Bi).
- ^{210}Po is alpha emitter with a half-life about 138 days and energy of 5.3 MeV.

^{210}Po poses no safety hazard during normal operation.

ZHANG, J., Lead–Bismuth Eutectic (LBE): A Coolant Candidate for Gen. IV Advanced Nuclear Reactor Concepts, Adv. Eng. Mater. 16, N° 4 (2014) 349.

Corrosion issue

- Both experimental and theoretical results have demonstrated that the current structural materials will be destroyed by corrosion in the LBE environment with high-temperature and high neutron flux;

Hao Wang , Jun Xiao, Hui Wang, Yong Chen , Xing Yin and Ning Guo, Corrosion Behavior and Surface Treatment of Cladding Materials Used in High-Temperature Lead-Bismuth Eutectic Alloy: A Review , Coatings 2021, 11, 364.

Compatibility with materials

- Thus, the compatibility between the cladding materials and LBE is considered as the main problem for the lead-cooled fast reactor (LFR) systems.

Hao Wang , Jun Xiao, Hui Wang, Yong Chen , Xing Yin and Ning Guo, Corrosion Behavior and Surface Treatment of Cladding Materials Used in High-Temperature Lead-Bismuth Eutectic Alloy: A Review , Coatings 2021, 11, 364.

Proposed solution

It has been found that corrosion by liquid lead alloys **can be successfully mitigated by control of the oxygen concentration in the coolant**.

ZHANG, J., Lead–Bismuth Eutectic (LBE): A Coolant Candidate for Gen. IV Advanced Nuclear Reactor Concepts, Adv. Eng. Mater. 16, N° 4 (2014) 349.

Proposed solution

The review discussed:

- the corrosion behavior of different candidate cladding materials for LBE-LFR and their coatings in the different LBE environment (temperature, oxygen concentration, flowing rate);
- the latest coating technologies that are expected to be applied to the cladding materials in LBE.

Hao Wang , Jun Xiao, Hui Wang, Yong Chen , Xing Yin and Ning Guo, Corrosion Behavior and Surface Treatment of Cladding Materials Used in High-Temperature Lead-Bismuth Eutectic Alloy: A Review , Coatings 2021, 11, 364.

Proposed solution

Several experiments were conducted on differents clading materials:

T91, HT-9, 1.4718, EP823, HCM12A, …

It found by carefully controlling the oxygen concentration the formation of duplex oxide layer:

- Outer layer: Fe_3O_4;
- Inner layer ($[Fe, Cr]_3O_4$)

Hao Wang , Jun Xiao, Hui Wang, Yong Chen , Xing Yin and Ning Guo, Corrosion Behavior and Surface Treatment of Cladding Materials Used in High-Temperature Lead-Bismuth Eutectic Alloy: A Review , Coatings 2021, 11, 364.

Proposed solution

This type of oxide layer is believed to protect SS from the LBE corrosion

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Hao Wang , Jun Xiao, Hui Wang, Yong Chen , Xing Yin and Ning Guo, Corrosion Behavior and Surface Treatment of Cladding Materials Used in High-Temperature Lead-Bismuth Eutectic Alloy: A Review , Coatings 2021, 11, 364.

Proposed solution

Surface Coatings of Cladding Materials

- ➢ Al-containing coatings;
- The introduction of Al_2O_3 layer on the SS
- ➢ SS surface aluminizing
- Intermetallic compound formation « FeAL »

Hao Wang , Jun Xiao, Hui Wang, Yong Chen , Xing Yin and Ning Guo, Corrosion Behavior and Surface Treatment of Cladding Materials Used in High-Temperature Lead-Bismuth Eutectic Alloy: A Review , Coatings 2021, 11, 364.

Proposed solution

- Al_2O_3 layer on the SS surface significantly improve the corrosion resistance in LBE;
- The alloying layer containing FeAl had exellent corrosion resistance

Hao Wang , Jun Xiao, Hui Wang, Yong Chen , Xing Yin and Ning Guo, Corrosion Behavior and Surface Treatment of Cladding Materials Used in High-Temperature Lead-Bismuth Eutectic Alloy: A Review , Coatings 2021, 11, 364.

Proposed solution

We note:

- ✓ Other elements, such as Si, Ti, W, and Mo, have been introduced in Addition to Al coating;
- ✓ Ceramc coatings: SiC, Ti_3SiC_2, TiAlN, TiSiN,…
- ✓ Amorphous and High-Entropy Alloy Coatings:

Amorphous SiO_2, Si_3N_4, Al_2O_3, Al0.7CoCrFeNi HEA,…

Hao Wang , Jun Xiao, Hui Wang, Yong Chen , Xing Yin and Ning Guo, Corrosion Behavior and Surface Treatment of Cladding Materials Used in High-Temperature Lead-Bismuth Eutectic Alloy: A Review , Coatings 2021, 11, 364.

Proposed solution

Amorphous metallic glass coatings

We recentely investigated radiation damage in amorphous metallic glass based on zirconium material: **$Zr_{70}Ni_{30}$**.

The results revealed: Smoothening of $Zr_{70}Ni_{30}$: this makes it a suitable protective coating in nuclear applications since the decrease in roughness increases the corrosion resistance

Wafa Boukhemkhem, Mahmoud Izerrouken, Matteo Ghidelli, Thomas Pardoen, Ali Sari, Abdel Yazid Khereddine and Ali Meftah, Swift heavy ion irradiation effect on structural, morphological and mechanical properties of Zr70Ni30 metallic glass, Phys. Scr. 98 (2023) 085311

Proposed solution

Corrosion inhibitors

Corrosion inhibitors such as Ti and Zr have been proposed. The reaction with the nitrogen and carbon of the steel container leads to the formation of protective layers ZrN, TiN, or TiN+TiC at the steel/LBE interface which inhibit corrosion.

ZHANG, J., Lead–Bismuth Eutectic (LBE): A Coolant Candidate for Gen. IV Advanced Nuclear Reactor Concepts, Adv. Eng. Mater. 16, N° 4 (2014) 349.

ZrN thin film applications and performance

Zirconium nitride is a promising material that exhibits a good combination of ceramics and metallic properties,

- very high melting point;
- good wear;
- corrosion resistance;
- low resistivity;
- high mechanical properties;
- excellent thermal and electrical conductivity and
- low thermal neutron absorption cross section

ZrN thin film applications and performance

It is used as:

- an inert matrix in some types of nuclear fuels;
- inert matrix material to burn plutonium or to transmute long-lived actinides;
- a diffusion barrier between the U-Mo fuel particles and the aluminum to avoid the formation of the interaction layer (U-Mo)Al_x during reactor neutron operation.

Radiation damage tolerance

As well known, fast neutrons and charged particles from fission or fusion reactions affect the properties of the protective layer and therefore accelerate the corrosion process.

Radiation damage tolerance

Thus, theoretical and experimental studies should be carried out to identify the irradiation tolerance of such protective layers.

So most of the coatings discussed above should be investigated

- ✓ **ZrN**, SiC, Ti_3SiC_2, TiAlN, TiSiN, Amorphous SiO_2, Si_3N_4, Al_2O_3, Al0.7CoCrFeNi HEA, …

Experiments

- Elaboration

ZrN thin films used in this study was elaborated using high power impulse magnetron sputtering technique and deposited on Zircaloy-4 substrate.

- Irradiation

The samples were irradiated at room temperature with 2 MeV proton in a fluence range of 1×10^{13}–1×10^{15} p.cm^{-2}. at 5UDH-2 Pelletron Tandem accelerator Islamabad, Pakistan.

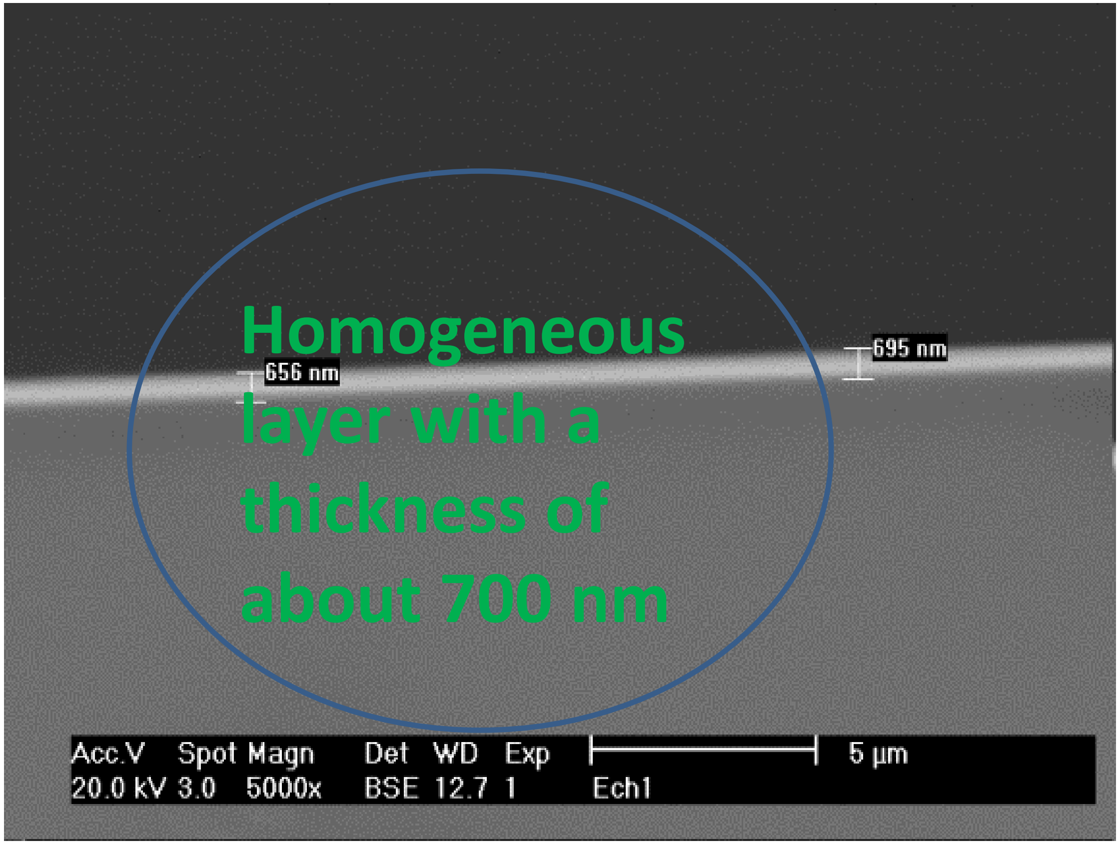
Experiments

- Characterization

After irradiation the defect formation was investigated using grazing incidence X-ray diffraction (GIXRD), atomic force microscopy (AFM) and nanoindentation tools.

Results and discussion

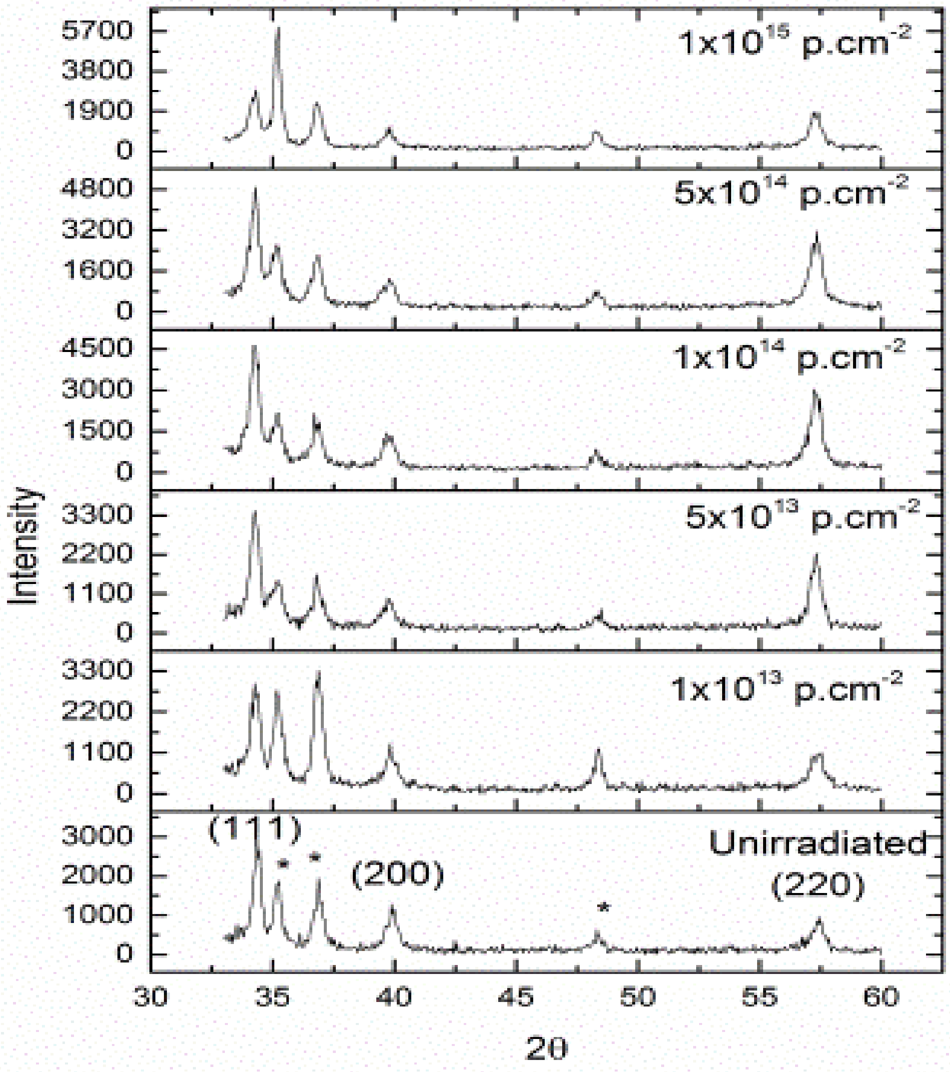
1. SEM cross section image



Homogeneous layer with a thickness of about 700 nm

Results and discussion

1. Structure analysis



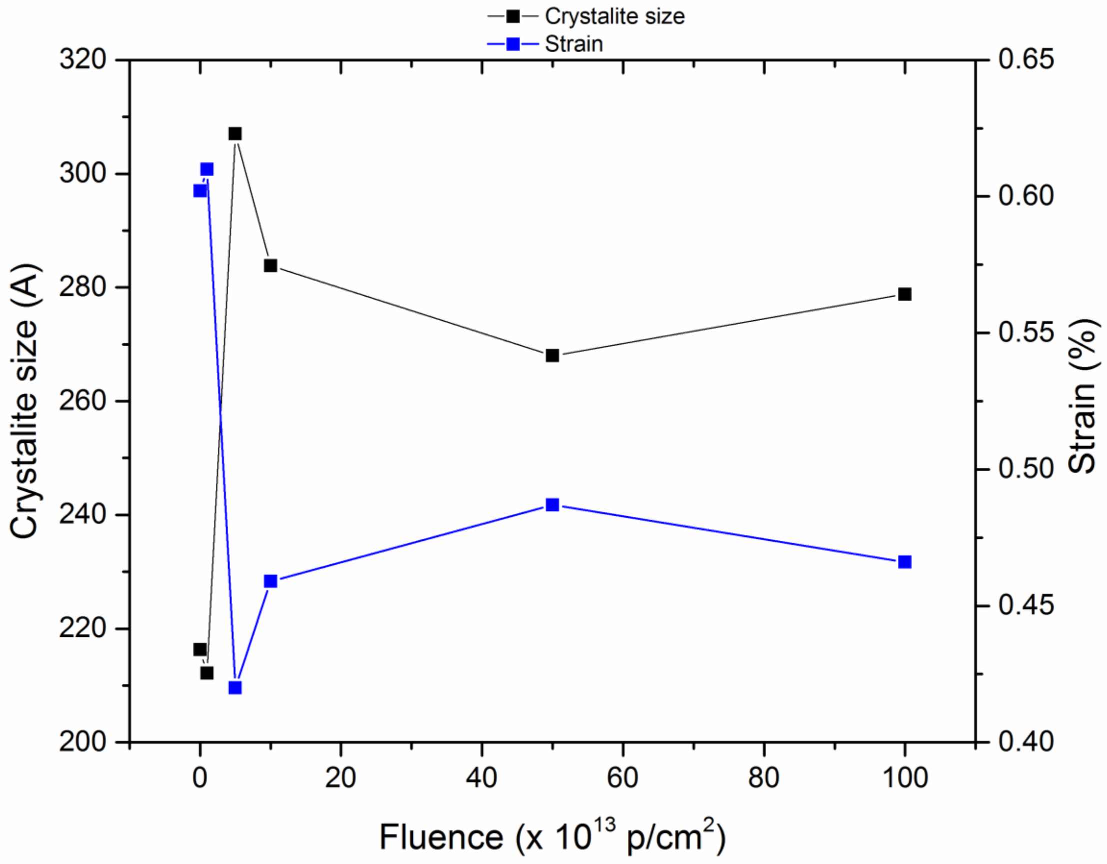
These suggest stable crystallinity and no swelling or contraction under irradiation

After irradiation, the intensity and position of the peaks remain unchanged

The DRX spectra of as elaborated sample show the diffraction from 111, 200 and 220 planes of cubic lattice of ZrN film

Results and discussion

1. Structure analysis

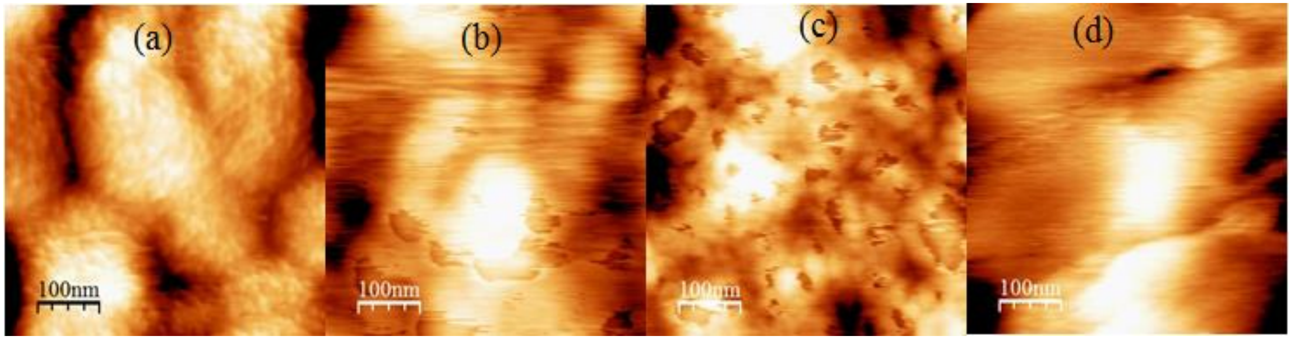


it is worthy to note the crystallite smallness improves the corrosion resistance

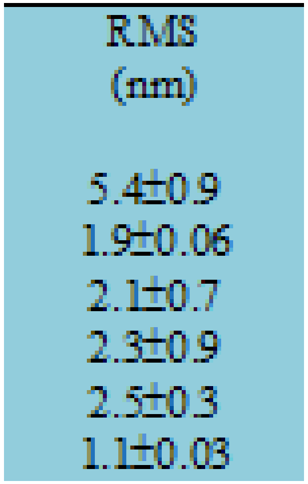
ALIYU A., SRIVASTAVA C., Correlation between growth texture, crystallite size, lattice strain and corrosion behavior of copper-carbon nanotube composite coatings, Surface & Coatings Technology 405 (2021) 126596.

Results and discussion

2. Surface morphology



Tapping-mode AFM images of ZrN film (a) before irradiation and after irradiation by 2 MeV proton at different fluence: (b) 1×10^{13} p.cm^{-2}, (c) 1×10^{14} p.cm^{-2}, (d) 1×10^{15} p.cm^{-2}.



RMS (nm)
5.4±0.9
1.9±0.06
2.1±0.7
2.3±0.9
2.5±0.3
1.1±0.03

The RMS evolution with fluence indicates the irradiation-induced smoothening which improve the corrosion resistance.

ALSHAMSI A. S. AND ALBLOOSHI A., Effect of surface roughness on the corrosion behavior of pure iron in acidic solutions Int. J. Electrochem. Sci 14 (2019) 5794–5812.

Results and discussion

2. Surface morphology

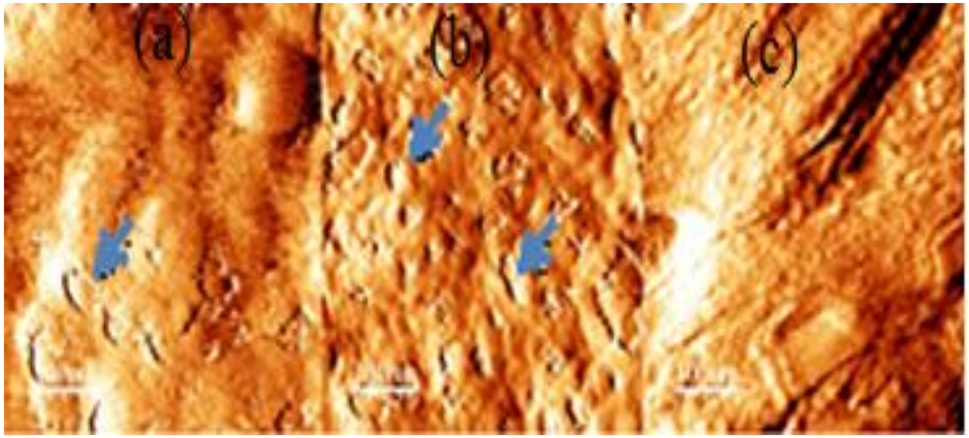
The surface smoothening may be interpreted by several mechanisms :

- plastic flow;
- evaporation-condensation;
- bulk diffusion and
- surface diffusion

Results and discussion

2. Surface morphology

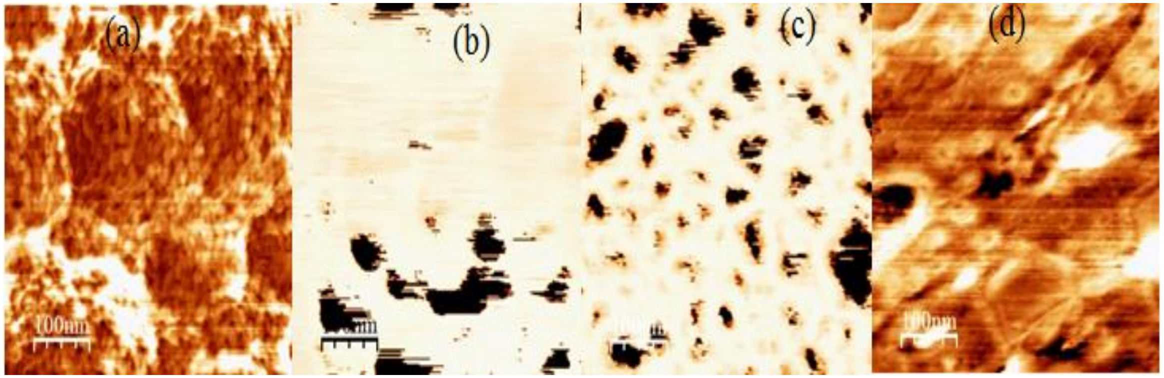
- In our case, the smoothening may be interpreted as evaporation-condensation mechanism caused by the sputtering.



Amplitude AFM images showing the formation of craters on the grains surface after irradiation by 2 MeV proton at different fluence:(a) 1×10^{13} p.cm^{-2}, (b) 1×10^{14} p.cm^{-2}, (c) 1×10^{15} p.cm^{-2}.

Results and discussion

2. Surface morphology

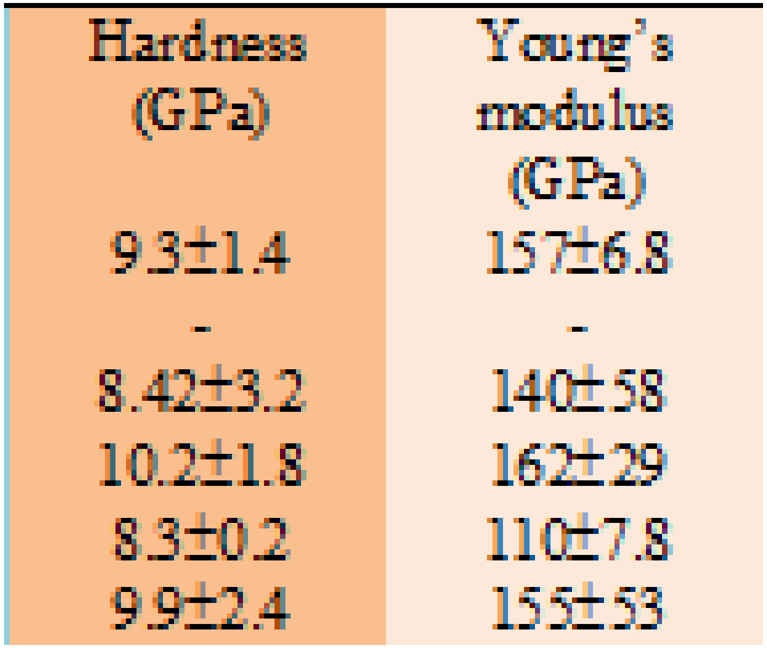


Phase AFM images of (a) as elaborated ZrN film compared to the phase AFM images of the 2 MeV proton-irradiated ZrN film:(b) 1×10^{13} p.cm^{-2}, (c) 1×10^{14} p.cm^{-2}, (d) 1×10^{5} p.cm^{-2}

The interpretation is confirmed by the Phase AFM images indicating a difference in contrast between the crater zones and the rest of the sample. This difference in contrast is probably due to the difference in hardness in the two regions

Results and discussion

3. Mechanical property



Hardness (GPa)	Young's modulus (GPa)
9.3±1.4	157±6.8
-	-
8.42±3.2	140±58
10.2±1.8	162±29
8.3±0.2	110±7.8
9.9±2.4	155±53

The data revealed no significant changes in both hardness and Young's moduls values after irradiation

Summary

The results revealed:

- stable crystallinity;
- no amorphization, swelling and contraction;
- both crystallite size and lattice strain, respectively, decrease and increase linearly with the fluence above 1×10^{14} p.cm^{-2};
- surface smoothening;
- no significant change in hardness and Young's modulus.

Summary

It is concluded from the results that:

- high radiation tolerance of the ZrN coating with strong structural stability under irradiation;
- surface smoothening and smallness of the crystallite under irradiation improve the corrosion resistance of ZrN film;
- **ZrN protective layer formed will not be affected by irradiation, which will mitigate LBE corrosion.**

Perspective

- Further investigation should be made to understand the craters formation and its effect on ZrN mechanical properties and corrosion evolution;
- Although a large number of corrosion experiments have been conducted, radiation damage tolerance of the developped coatings must be well investigated under different irradiation conditions.

So, additional experiments are needed to avoid any surprises during reactor operation