Journal of the Serbian Chemical Society
JSCS-info@shd.org.rs • www.shd.org.rs/JSCS
J. Serb. Chem. Soc. **00(0)** S1-S3 (2024) *Supplementary material*

SUPPLEMENTARY MATERIAL TO

Molecular dynamics simulation of uranium nitride oxidation

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PREPARATION OF THE SIMULATED SYSTEMS

An amorphous particle was created in a preliminary molecular dynamics calculation by melting a crystal at a temperature of 3000 K and then holding it at this temperature for 200 ps (2 million time steps). The process of the amorphous UN structure preparation is shown in Fig. S1. After the particle acquired an irregular structure, it was uniformly and rapidly cooled over a time interval of 100 ps until a temperature of 300 K was reached.

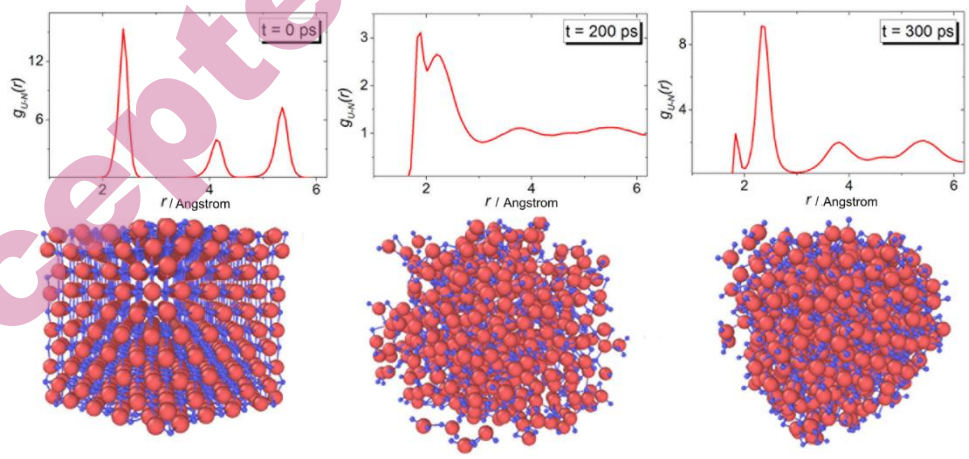

Fig. S1. The process of the amorphous UN structure preparation, the insets above show the partial radial distribution functions $g_{\mathrm{U-N}}(r)$ corresponding to each of the above states; uranium atoms are marked in red, nitrogen atoms are in blue.

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The preparation of the simulated systems “UN crystalline/amorphous particle in the Ar-O gaseous medium” was started from the creation of the argon-gaseous medium. The process of its preparation was carried out in the NPT ensemble as follows: 1) initially, an argon crystal was melted at a fixed barostat pressure of 1 bar, converting it into a gaseous state at T = 1273 K for 400 ps; 2) at the second stage, oxygen was gradually introduced into the gaseous argon mixture, by adding one atom every 0.2 ps (2000 time steps) until the desired oxygen concentration was reached; 3) finally, the mixed argon-oxygen gas system was equilibrated for 400 ps. The barostat relaxation time was $\tau_T = 10$ fs.

The previously obtained and equilibrated argon-oxygen medium was further used to introduce an amorphous or crystalline UN particle into it. For that, the volume of the simulation cell was increased by moving its upper face upward. This face was raised until an amorphous particle (its volume was greater than the volume of the crystalline particle) could be placed between it and the previously obtained surface of the gas system. The new position of the upper face of the MD cell was fixed, and the UN particle (crystalline or amorphous) was placed in the upper part of the MD cell. The lower part of the cell was occupied by the gaseous Ar-O mixture.

A microheterophase system “UN crystalline/amorphous particle in a gaseous Ar-O mixture” was created in the MD calculation using Maxwellian boundary conditions. Already at the beginning of this calculation, Ar and O atoms occupied the free volume formed at the top of the MD cell. Due to the use of Maxwellian boundary conditions, the UN particle immersed in the gaseous medium performed random movements over the cell.

MORSE POTENTIAL FUNCTION

In the demonstration calculation, the EAM potential describing U-N interactions in the system was replaced by the pair potential. Pair potential built on the basis of the Morse potential function[14] has a form:

$$\Phi = \frac{z_i z_j e^2}{r_{ij}} + (b_i + b_j)\exp\left[\frac{a_i + a_j - r_{ij}}{b_i + b_j}\right] + \\ + D_{ij}\{\exp[-2\beta_{ij}(r_{ij} - r_0)] - 2\exp[-\beta_{ij}(r_{ij} - r_0)]\} \quad \text{(S1)}$$

here e (= 1) is the elementary electrical charge, r_{ij} is the distance between i and j ions, other potential parameters are presented in Table SI.

TABLE SI. Parameters of the potential represented by equation (S1).

Ion	z	a, Å	b, Å	Bond	D_{ij}, eV	β_{ij}, Å^{-1}	r_0, Å
N^{3-}	-1.450	1.797	0.080	N^{3-}– U^{3+}	7.00	1.25	2.364
U^{3+}	+1.450	1.228	0.080				

The form of potential (S1) is presented in Fig. S2.

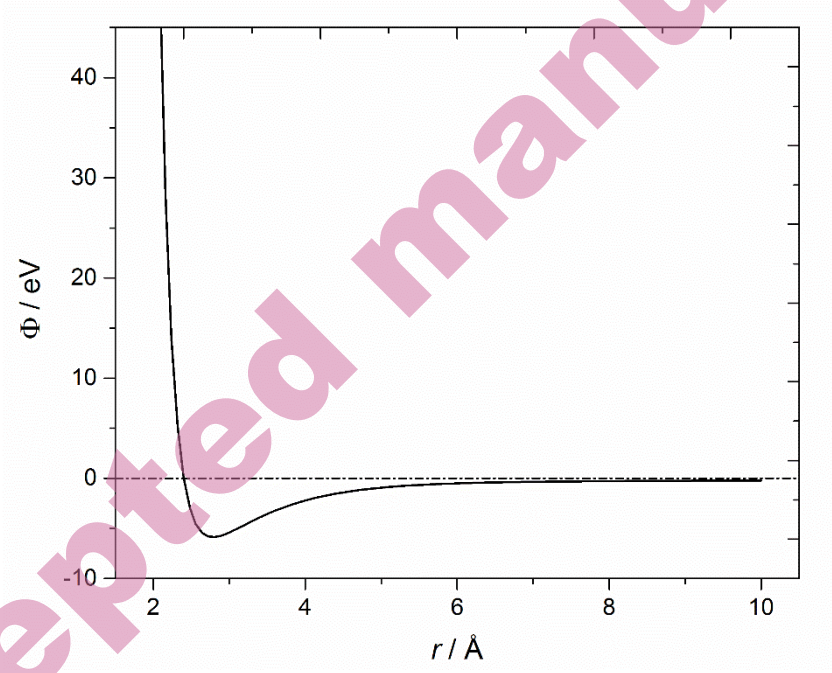

Fig. S2. The U-N interaction potential represented by formula (S1).

The replacement of the potential describing the U-N interaction was performed to simulate a system with an amorphous particle that had already split into fragments, the largest of which had an irregular shape and tended to undergo further segmentation. The newly used (pair) potential had been previously employed to simulate the crystalline UN phase in the temperature range of 300-2000 K at a pressure of 1 bar. Our calculation with this pair potential at a temperature of 923 K is presented in the demonstration movie (external link):

(https://watercluster.ucoz.ru/publ/2024/supplementary/supplementary_for_journal_of_the_serbian_chemical_society/190-1-0-222).