MCNP-4C2 and TRIM 98.01 based calculations of radiation damage in copper

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Aims

• Simulation of the defects movement based on the Embedded Atom Method (EAM) potentials and ab initio method in combination with Monte Carlo technique.

• Irradiation of specimens with neutrons and protons.

• Measurement of microstructure changes in materials using Positron annihilation techniques compared with TEM.

• Heat treatment, measurements and evaluations of results from microstructural point of view.
The interatomic potential used in the simulation is based on the Embedded Atom Method (EAM). For additional simulations we used Ab initio method in combination with the usual copper pseudopotential.
Distribution of neutron fluency, atomic displacement, nuclear heating and nuclear reactions in copper alloys foreseen for ITER first wall were calculated using Monte Carlo N-Particle Transport code MCNP-4C2.

Geometrical model consists of the slab with dimensions 12 x 12 x 0.5 mm, divided into 50 layers which create elementary cells with thickness 10 µm to provide detailed spatial distribution of the nuclear parameters. The shape of the modeled specimen correlates with the shape of samples used in the foreseen experiment - PAS measurements.
Specimens chemical composition and implantation dose.

<table>
<thead>
<tr>
<th>Specimen</th>
<th>Material</th>
<th>Implantation dose [C/cm²]</th>
</tr>
</thead>
<tbody>
<tr>
<td>SS</td>
<td>CuCrZr – 98.95w.%Cu; 0.6-0.9w.%Cr; 0.07-0.15w.%Zr</td>
<td>None</td>
</tr>
<tr>
<td>SA</td>
<td>CuCrZr</td>
<td>0.4</td>
</tr>
<tr>
<td>SM</td>
<td>CuCrZr</td>
<td>1.1</td>
</tr>
<tr>
<td>TT</td>
<td>CuAl25 – 99.5w.%Cu; 0.25w.%Al as Al₂O₃; 0.22w.%O as Al₂O₃; 0.025w.%B as B₂O₃</td>
<td>None</td>
</tr>
<tr>
<td>TA</td>
<td>CuAl25</td>
<td>0.4</td>
</tr>
<tr>
<td>TM</td>
<td>CuAl25</td>
<td>1.1</td>
</tr>
</tbody>
</table>

MATERIAL CHARACTERISTICS OF THE SPECIMEN.

<table>
<thead>
<tr>
<th>Material</th>
<th>Mass density (g/cm³)</th>
<th>Isotope fraction (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cu</td>
<td>8.93</td>
<td>⁶³Cu 67.17</td>
</tr>
<tr>
<td></td>
<td></td>
<td>⁶⁵Cu 30.83</td>
</tr>
</tbody>
</table>
Distribution of primary knock-out atoms in the specimen

<table>
<thead>
<tr>
<th>Particle</th>
<th>Total normalized production in the specimen [1/(source neutron-cm(^3))]</th>
<th>Total unnormalized production in the specimen (1/cm(^3))</th>
</tr>
</thead>
<tbody>
<tr>
<td>Photon</td>
<td>2.539E-01</td>
<td>7.842E11</td>
</tr>
<tr>
<td>Proton</td>
<td>1.324E-02</td>
<td>4.090E10</td>
</tr>
<tr>
<td>Deuterium</td>
<td>5.578E-04</td>
<td>1.723E09</td>
</tr>
<tr>
<td>(^3)He</td>
<td>2.002E-07</td>
<td>6.184E05</td>
</tr>
</tbody>
</table>
The decreasing of the specimen activity in the time dependence
NUCLEAR REACTION IN THE SPECIMEN.

<table>
<thead>
<tr>
<th>Target isotope</th>
<th>Reaction</th>
<th>Product</th>
<th>Number of reactions [1/(source neutron·cm³)]</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{63}$Cu</td>
<td>(n,2n)</td>
<td>$^{62}$Cu $\rightarrow$ $^{62}$Ni stable (st.)</td>
<td>1.82378E-02</td>
</tr>
<tr>
<td></td>
<td>(n,n'p)</td>
<td>$^{62}$Ni st.</td>
<td>1.06331E-02</td>
</tr>
<tr>
<td></td>
<td>(n,n'α)</td>
<td>$^{59}$Co st.</td>
<td>5.10850E-04</td>
</tr>
<tr>
<td></td>
<td>(n,p)</td>
<td>$^{63}$Ni $\rightarrow$ $^{63}$Cu st.</td>
<td>1.86386E-03</td>
</tr>
<tr>
<td></td>
<td>(n,d)</td>
<td>$^{62}$Ni st.</td>
<td>3.84324E-04</td>
</tr>
<tr>
<td></td>
<td>(n,3He)</td>
<td>$^{61}$Co $\rightarrow$ $^{61}$Ni st.</td>
<td>1.55531E-07</td>
</tr>
<tr>
<td></td>
<td>(n, α)</td>
<td>$^{60}$Co $\rightarrow$ $^{60}$Ni st.</td>
<td>1.70658E-03</td>
</tr>
<tr>
<td></td>
<td>(n,γ)</td>
<td>$^{64}$Cu $\rightarrow$ $^{64}$Ni st. (61%) $\rightarrow$ $^{64}$Zn st. (39%)</td>
<td>1.28033E-04</td>
</tr>
<tr>
<td>$^{65}$Cu</td>
<td>(n,2n)</td>
<td>$^{64}$Cu $\rightarrow$ $^{64}$Ni st. (61%) $\rightarrow$ $^{64}$Zn st. (39%)</td>
<td>1.62292E-02</td>
</tr>
<tr>
<td></td>
<td>(n,n'p)</td>
<td>$^{64}$Ni st.</td>
<td>3.02890E-04</td>
</tr>
<tr>
<td></td>
<td>(n,n'α)</td>
<td>$^{61}$Co $\rightarrow$ $^{61}$Ni st.</td>
<td>1.39880E-05</td>
</tr>
<tr>
<td></td>
<td>(n,p)</td>
<td>$^{65}$Ni $\rightarrow$ $^{65}$Cu st.</td>
<td>4.37881E-04</td>
</tr>
<tr>
<td></td>
<td>(n,d)</td>
<td>$^{64}$Ni st.</td>
<td>1.73503E-04</td>
</tr>
<tr>
<td></td>
<td>(n,3He)</td>
<td>$^{63}$Co $\rightarrow$ $^{63}$Ni $\rightarrow$ $^{63}$Cu st.</td>
<td>4.66784E-08</td>
</tr>
<tr>
<td></td>
<td>(n, α)</td>
<td>$^{62}$Co $\rightarrow$ $^{62}$Ni st.</td>
<td>1.82165E-04</td>
</tr>
<tr>
<td></td>
<td>(n,γ)</td>
<td>$^{66}$Cu $\rightarrow$ $^{66}$Zn st.</td>
<td>1.47605E-05</td>
</tr>
</tbody>
</table>
1 MeV cascade accelerator at STU Bratislava
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Total accelerating voltage: 0 - 1 MV
Ripple factor: < 1%
Energy range for singly charged particles: 5 keV to 1 MeV
Energy spread:
- 70 keV – 1 MeV: ≈ 2 keV
- < 70 keV: < 0.1%
Beam current: 1 - 100 µA
Ion implantation and measurements using PLEPS

- For the simulation of radiation stress of neutron flux, the ion implantation of protons has been applied.
- The proton mass is approximately identical as the neutron mass
- The energy of protons of about 95 keV was found out with the help of program TRIM 98.01
- Two implantation doses 1.1 and 0.4 C/cm² were chosen

- **Pulsed Low Energy Positron System** PLEPS seems to be optimal technique for the evaluation of defects structure and concentration in Cu-alloys
- This system enables to study the micro structural changes in the region from 20 to 500 nm
Reasons for application of PAS

• Mechanical tests give not enough information about changes in material microstructure. Therefore, additional methods should be applied.

• PAS technique is a well-established method for studying open-volume type atomic defects and defect’s interactions in metals

• Ability of PAS to detect very small defects as well as very low defects concentration

• PAS can give additional information about
  • radiation induced defects,
  • thermal annealing of these defects.

General overview of applicability range for some spectroscopy techniques [Howell, R., Physics Space Technology, 1987].

PAS Theory

Scheme of positron experiments (A),

Quantitative illustration of positron annihilation
γ-rays from different environments (B)
PAS Theory (PAS LT)

Lifetime spectra (PAS LT).
Scheme of Lifetime measurement system
Three detector set-up of PALS realization

PALS – electronic part

BaF₂ detectors and samples with $^{22}$Na source
Distribution of vacancies in Cu-alloys after irradiation (calculated by TRIM)
• Schematic view of the latest version of the pulsed low energy positron system.
PAS results

SS non-irradiated sample

- Non-irradiated CuCrZr sample (0 C/cm²)
- There have not been observed any defects

SM proton irradiated sample (1,1 C/cm²)

- Irradiated CuCrZr sample (1,1 C/cm²)
  - the shape of voids is spherical with diameter from 2 to 15nm
PLEPS Results for ITER

Decomposition of PASLT spectrum in two components
PAS and TEM results are usable for microstructural evaluation of new materials.

The calculated dependence of the positron lifetime on the size of the vacancy cluster in Cu.
PLEPS Results for ITER

Annealing behaviour of irradiated CuAl25-sample (TM)
Conclusions I

• The distinction between the two studied materials consists – in accordance with TEM observations – in the fact that the microstructure of CuAl25 material is almost intact to irradiation contrary to CuCrZr.

• In CuCrZr material positron annihilation characteristics are apparently changed due to irradiation.

• Annealing up to 600 °C seem to result to a state similar to annealed non-irradiated sample.

• It is important to be very precise by sample preparation and by excluding of contributions from oxides, source, back-diffusion, etc.
Neutron irradiation at BR2 reactor

Nominal power 60-120 MW
Maximum neutron flux (at 600 W/cm²)
thermal neutrons $1.2 \times 10^{15}$ n.cm⁻².s⁻¹
fast neutrons (E>0.1 MeV) $8.4 \times 10^{14}$ n.cm⁻².s⁻¹
Days of operation per year 168 to 240
Irradiation positions up to 100
Cannel diameter up to 200 mm
Dose dependence, measured on PA samples irradiated to different dose levels at $T_{irr}=50^\circ$C. Experimental values are compared with the trapping model value $\tau_{calc}$, using trapping rate $\kappa$ values.
The intensity $I_2$ of the long-lived component in dose dependence for neutron irradiated prime aged CuCrZr (Outokumpu) alloy.
Trapping rate $\kappa$ into the defect (trapping site) in neutron irradiated PA sample at $T_{irr}=50^\circ$C for three different irradiation doses.
Results from PAS experiment at neutron irradiated samples (collaboration with Risoe N.L. and SCK.CEN Mol)

• In all neutron irradiated samples from dose $1 \times 10^{-1}$ dpa up to $3 \times 10^{-1}$ dpa the values for the positron mean lifetime increased and the defect lifetime of $175 \pm 5$ ps was observed. Thus, compared to the non-irradiated state an additional trapping site was observed, probably in the form of Stacking Fault Tetrahedra.

• In all the positron lifetime measurements on neutron irradiated samples no lifetime component longer than 206 ps was observed, implying that no voids are created in the neutron irradiated CuCrZr (Outokumpu) alloy.