Coupled first-principles and experimental positron annihilation study of defects in actinide mixed oxides

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Coupled first-principles and experimental positron annihilation study of defects in actinide mixed oxides

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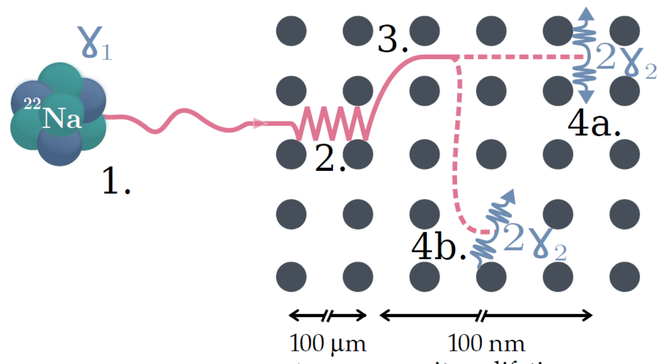
Context

- Positron Annihilation Spectroscopy (PAS) is a powerful technique to detect vacancy-type defects in materials
- We demonstrated its capability to identify irradiation-induced defects in UO_2 by coupling experiments and ab initio calculations [1]
- MOX fuels ($UPuO_2$) will be the fuel of Gen IV reactors
- MOX fuels fabricated from spent fuels → contains few % of minor actinide elements like Am
- Under irradiation, defects are created in the fuel and have strong impact on behaviour of fission products and evolution of microstructure



1. Positron generation in β^+ decay
2. Positron thermalization
3. Diffusion
4a. Annihilation in the bulk
4b. Trapping in a defect and annihilation

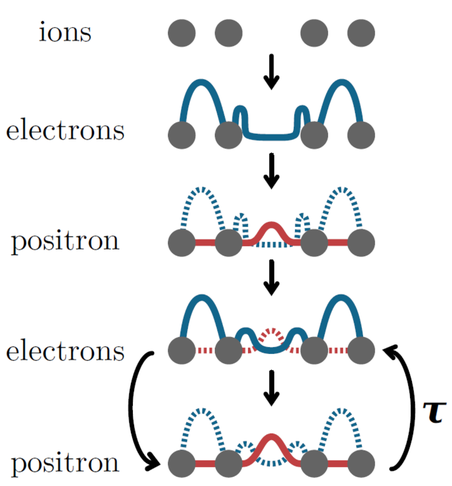
γ_1 -1.27 MeV
γ_2 -≈0.51 MeV
$\Delta\gamma_2$ -Doppler broadening

Scheme of the main positron-solid interactions

➡ Strong interest in characterizing vacancy defects created in actinides mixed oxides

Methods

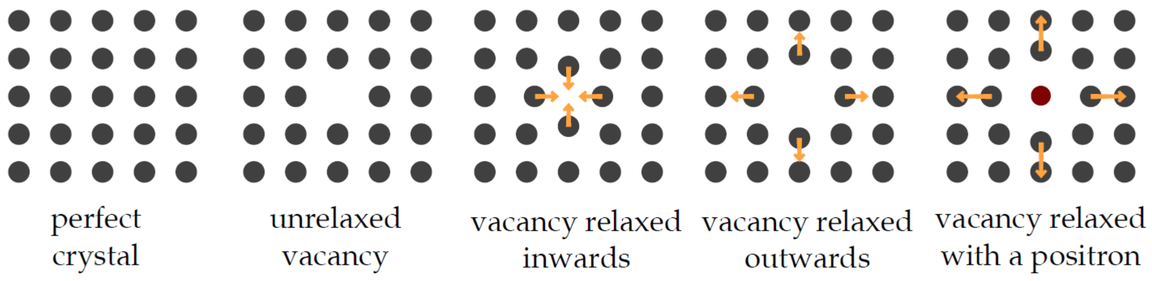
Positron lifetimes are computed using electronic structure calculations in the two component density functional theory (TCDFT) [2].
Self-consistent calculations are performed to obtain the electron and positron densities.



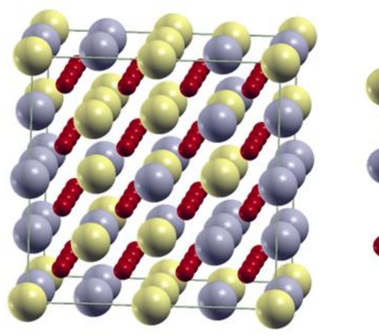
1. Positions of ions in the system are defined.
2. Electronic density is calculated for given positions of ions.
3. Density of the positron interacting with the electrons is calculated.
4. Electronic density is recalculated in the presence of the positron.
5. Positron density is recalculated and this cycle is continued until convergence.

Positron lifetime calculation scheme

In our study the relaxation effect due to the vacancy creation and the positron localized in the defect is taken into account.



Calculations are performed with the ABINIT code [3] using 96 atom supercells. The DFT+*U* [4] method is used to take into account the strong correlations between the 5*f* electrons.



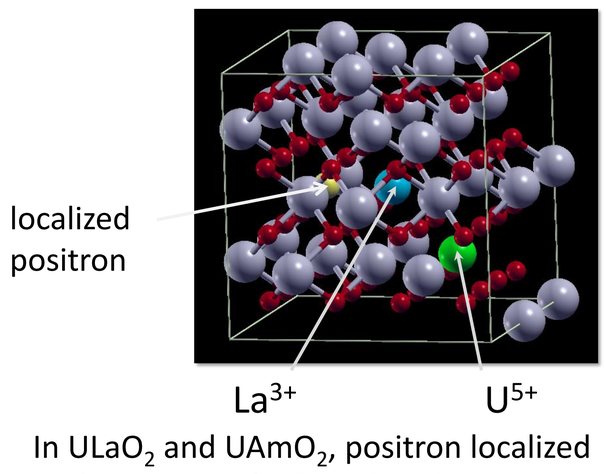
Mixed oxides supercell are simulated using the SQS method.

Preliminary results : unexpected localization of positron in bulk actinide oxides

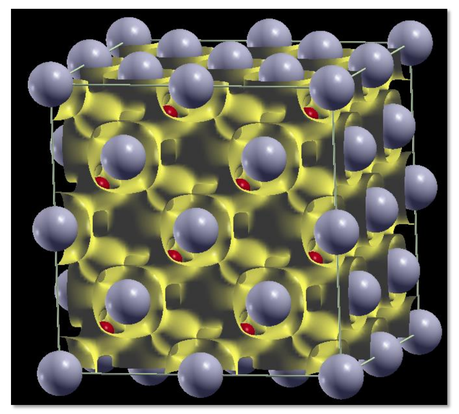
4 compounds investigated : U**Ce**O_2, U**Pu**O_2, U**Am**O_2 and U**La**O_2

For pure tri-valent elements, bulk lifetime within the GGGC-GGA scheme is much higher than expected.

compounds	$\tau_{GGGC-GGA}$ (ps)	$\tau_{CONV-GGA}$ (ps)
U**La**O_2 3,125 at%	199	168
U**La**O_2 6,25 at%	201	168
U**La**O_2 12,5 at%	202	168
U**La**O_2 25 at%	206	168
U**La**O_2 50 at%	211	168
U**Am**O_2 6,25 at%	200	168
U**Ce**O_2 25 at%	168	168
U**Pu**O_2 6,25 at%	168	168
UO_2 τ_{exp}=169±1ps	168	168



In $ULaO_2$ and $UAmO_2$, positron localized in the vicinity of La^{3+} and Am^{3+} respectively.

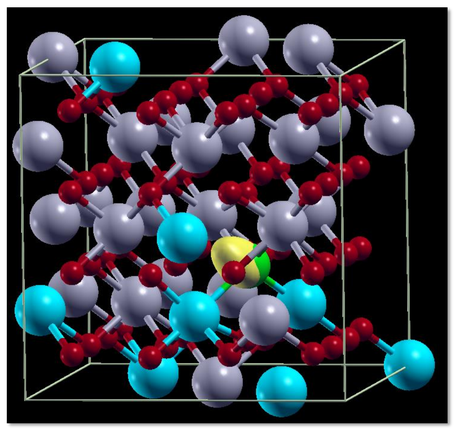


Delocalized positron in bulk UO_2, $UCeO_2$ and $UPuO_2$.

For oxygen mono-vacancy :

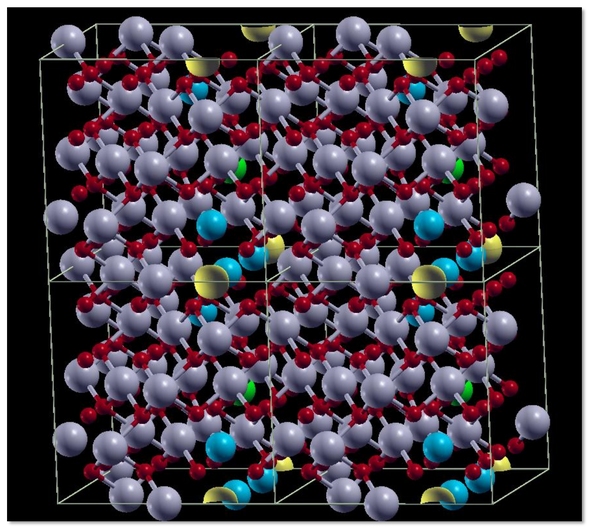
Positron localization in the vacancy for $UCeO_2$ and $UPuO_2$

$\tau_{V_O\text{-}UCeO_2}$ =227ps



Same positron localization as in the bulk for $ULaO_2$ and $UAmO_2$

$\tau_{V_O\text{-}ULaO_2}$ =202ps



Experimental work in progress on $UCeO_2$ and $ULaO_2$ at CNRS/CEMHTI : DFT predictions soon confirmed?

References: [1] J. Wiktor, G. Jomard, M. Torrent et al., Phys. Rev. B 90, 184101 (2014)
[2] R. M. Nieminen, E. Boronski and L. J. Lantto, Phys. Rev. B 32, 1377 (1985)
[3] X. Gonze et al., CPC 205 (2016) 106
[4] B. Dorado, B. Amadon, M. Freyss and M. Bertolus, Phys. Rev. B 79, 235125 (2009)