CERIUM DIFFUSION IN IRRADIATED AND NON-IRRADIATED SINGLE AND POLYCRYSTALLINE YTTRIA STABILIZED ZIRCONIA

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MOTIVATION OF THE STUDY

Cubic yttria-stabilized zirconia (YSZ) is considered as one of the most promising candidates as an inert matrix for the transmutation of actinides produced in nuclear power plants. Safety considerations require the evaluation of the retention properties of volatile fission products in the selected matrices. Such a problem can be studied by following the cationic diffusion upon subsequent thermal treatments. For example, cerium as tetravalent actinide and gadolinium as trivalent actinide can be used.

The present work is focused on cerium (Ce) diffusion in non-irradiated and irradiated YSZ. Samples were irradiated at room temperature with 4 MeV or 20 MeV Au ions at fluences ranging from $5\,10^{14}$ to $5\,10^{15}$ cm^{-2}. The Ce diffusion profiles were determined by secondary ion mass spectrometry (SIMS) and allowed the determination of bulk diffusion (D_B) in single crystals and effective (D_{eff}), bulk and grain boundary diffusion parameters (αD_{GB}) in polycrystals. The dependence of the diffusion coefficients on temperature is described by means of Arrhenius plots and the influence of radiation defects on cationic diffusion is discussed.

EXPERIMENTAL

Materials

8YSZ and 10YSZ polycrystals
9.5YSZ single crystals

Irradiation

4 MeV Au ions (CSNSM, F-Orsay)
20 MeV Au ions (Porto Alegre, Brasil)

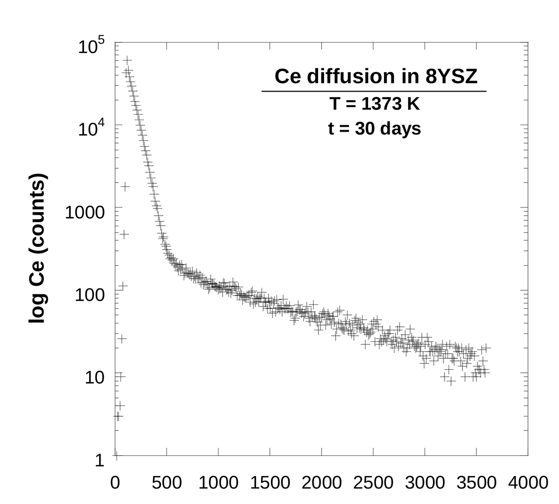
Tracer deposition

CeO_2 thin film deposited either by spin coating or physical vapor deposition (PVD) on the surface of polished samples

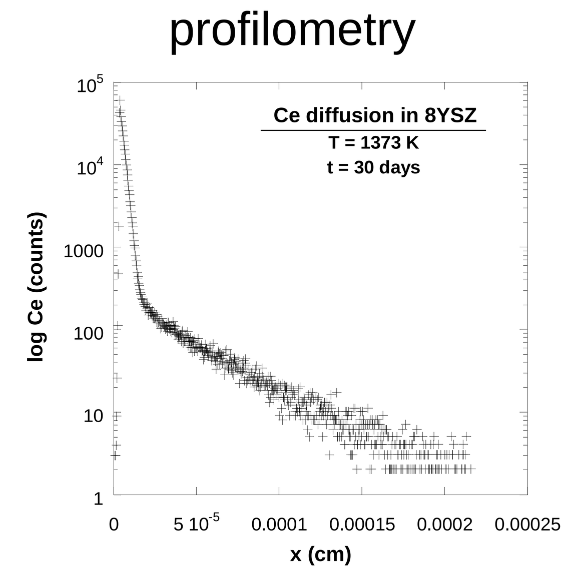
Diffusion annealing conditions

Sample	Irradiation conditions		T (°C)	t (days)
Polycrystal 8YSZ	Au 4 MeV	10^{15} cm^{-2}	1673 1373 1273	2 30 45
	Au 20 MeV	$5\,10^{14}$, $5\,10^{15}$ cm^{-2}	1673 1373 1273	2 30 45
Polycrystal 10YSZ	Au 4 MeV	10^{15} cm^{-2}	1673 1373 1273	2 19 45
Single crystal 9.5YSZ	Au 4 MeV	10^{15} cm^{-2}	1673 1373 1273	2 30 45

Depth profiling by SIMS

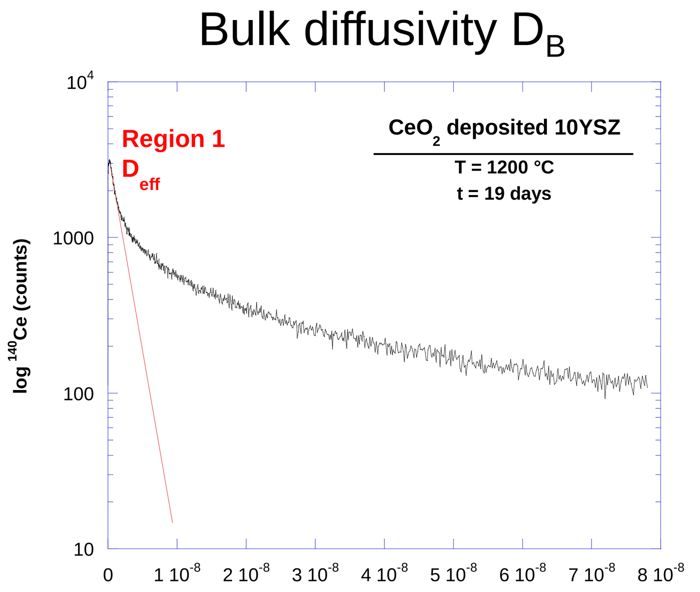


After conversion of the sputtering time into crater depth determined by profilometry



D_B and αD_{GB} CALCULATIONS

Bulk diffusivity D_B



Type-B kinetics regime

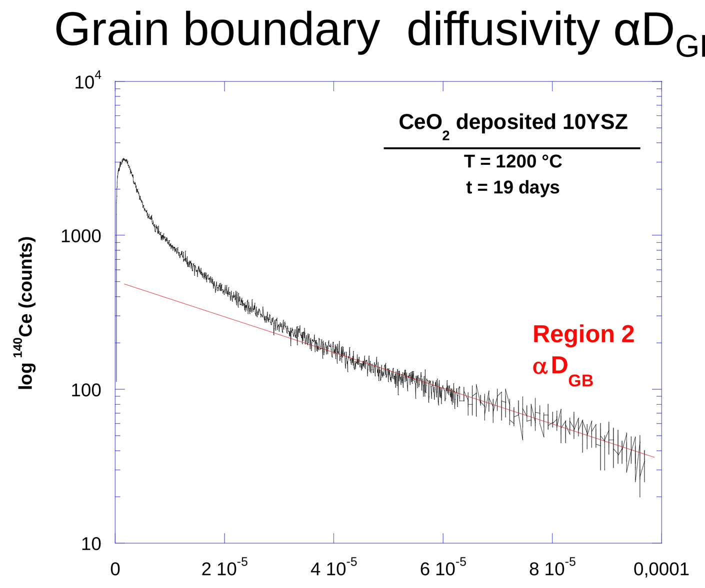
$\phi = 5.10^{-4}$ cm ; $\delta = 10^{-7}$ cm

$$f = \frac{3\delta}{\phi}$$

Solution of 2nd Fick law for thin films

$$\frac{C(X)}{C_0} = \exp\left(-\frac{X^2}{4 D_{eff}\, t}\right)$$

Grain boundary diffusivity αD_{GB}



Hart equation

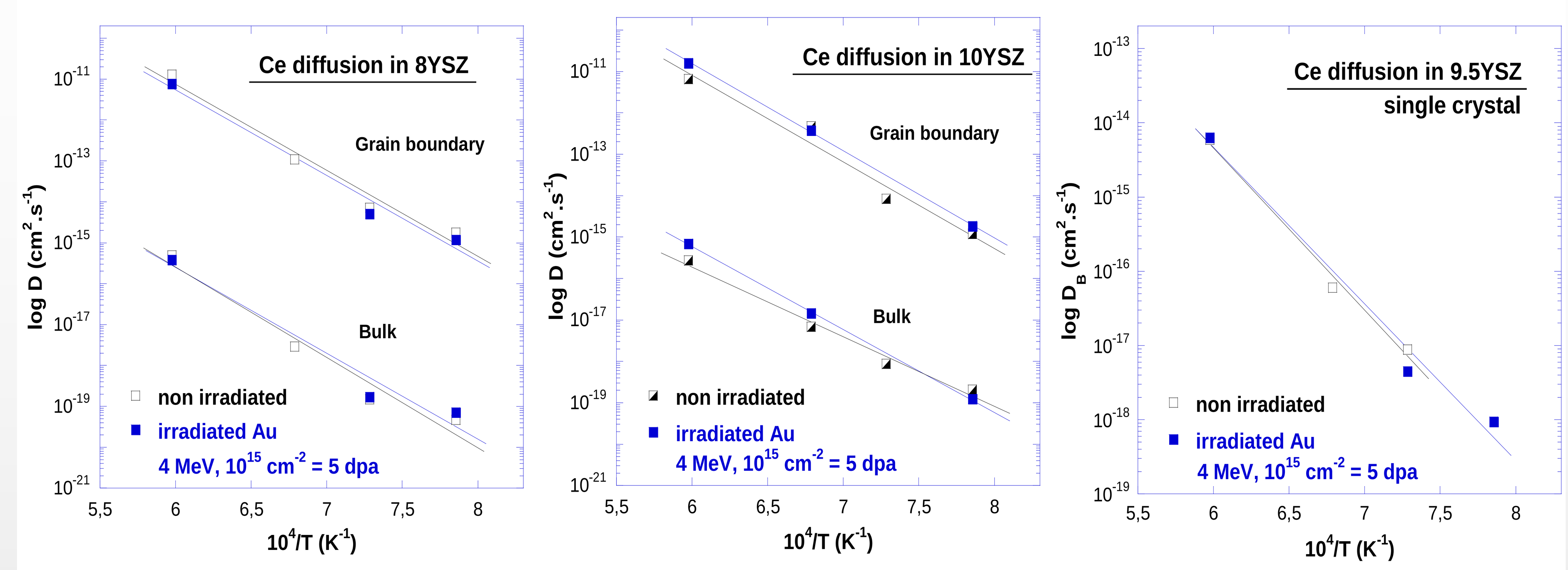
$$D_{eff} = (1-f) D_B + f\,\alpha\, D_{GB}$$

Whipple / Le Claire model

$$\delta\,\alpha\, D_{GB} = 0{,}66 \left(\frac{4 D_B}{t}\right)^{1/2} \left(-\frac{\partial \ln C(X)}{\partial X^{6/5}}\right)^{-5/3}$$

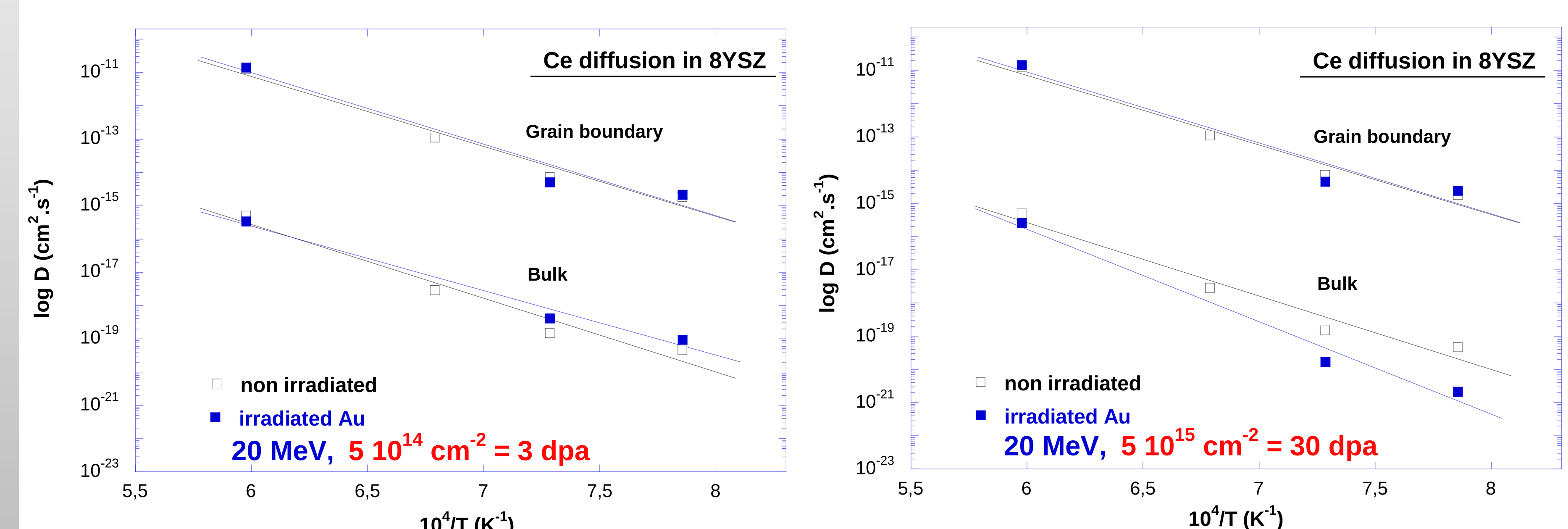
RESULTS

Irradiation with Au ions of low energy 4 MeV 10^{15} cm^{-2}



➢ No influence of low energy irradiation on D_B and αD_{GB} of Ce in polycrystalline and single crystal YSZ

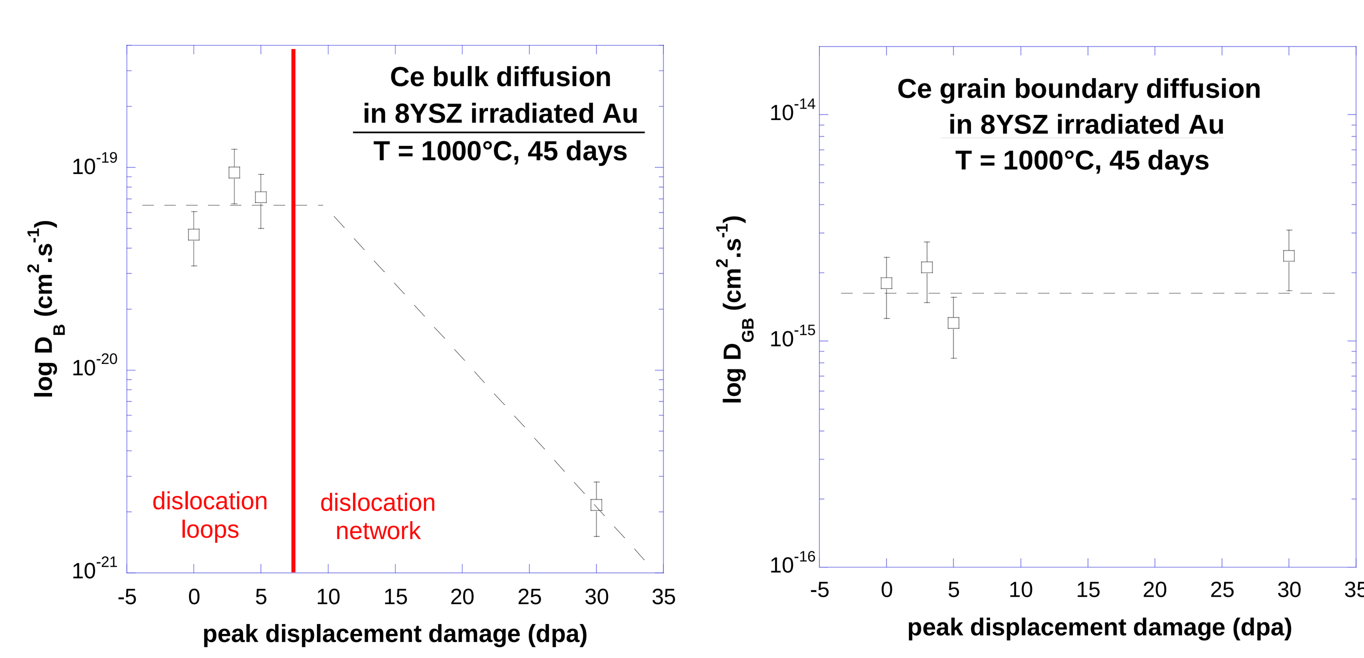
Irradiation with Au ions of high energy 20 MeV



➢ For the lowest fluence ($5\,10^{14}$ cm^{-2}), bulk diffusion D_B and grain boundary diffusion αD_{GB} parameters do not change with irradiation damage.

➢ For the highest fluence ($5\,10^{15}$ cm^{-2}), at low temperatures, D_B Is smaller in irradiated materials.

Influence of the radiation damage



➢ only D_B is affected by high radiation damage

dpa = number of displacement per atom

CONCLUSIONS

The study of Ce diffusion in irradiated and non-irradiated single and polycrystalline yttria stabilized zirconia in the temperature range 1000°C – 1400°C in air has shown that :

- For low damage level (dpa < 5), D_B is quite the same in both irradiated and non-irradiated materials,
- For high damage level (~30 dpa), D_B is slown down probably due to trapping by irradiation defects,
- Irradiation damage has no influence on grain boundary diffusion.

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