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Thermally induced outdiffusion studies of deuterium in ceramic breeder blanket materials

E.Carella[a,b], M.González[a], A.Moroño[a], M.H.H.Kolb[c], R.Knitter[c]

[a] *LNF-CIEMAT, Materials for Fusion Group, Madrid, Spain*

[b] *Fundación UNED, Madrid, Spain*

[c] *Karlsruhe Institute of Technology, Institute for Applied Materials (IAM-WPT), Karlsruhe, Germany*

Candidate materials for the ITER EU TBM are tested for their outgassing behavior simulating reactor operational conditions (Temp, radiation..).

Lithium orthosilicate based pebbles, with different metatitanate contents, and pellets of the individual oxide components were exposed to a deuterium atmosphere at RT while being gamma-irradiated up to 4MGy. Then the thermally induced release rate of D_2 gas was registered up to 800 C.

The rate desorption curves reveal differences in the D_2 sorption/desorption behavior depending on the composition and the radiation-induced electronic modifications of the trapping centers.

Reference material for ITER Tritium breeder blanket:

Lithium orthosilicate **pebbles** (**o-LiSi**) + 2.5 wt.% of silica fabricated by the established melt-spraying process at SCHOTT AG (Mainz, Germany) (95-96% D_{th}; close porosity of about 1%). To enhance the mechanical properties, presently **o-LiSi** pebbles with lithium metatitanate (m-LiTi) as a second phase are fabricated by a modified melt-based process.

Pellets of the individual components (o-LiSi and m-LiTi) were isostatically pressed startin from powders manufactered by different methods, and finally sintered in air up to 1150ºC/2hs achieving an open porosity of 22 to 27% with pore sizes <1μm.

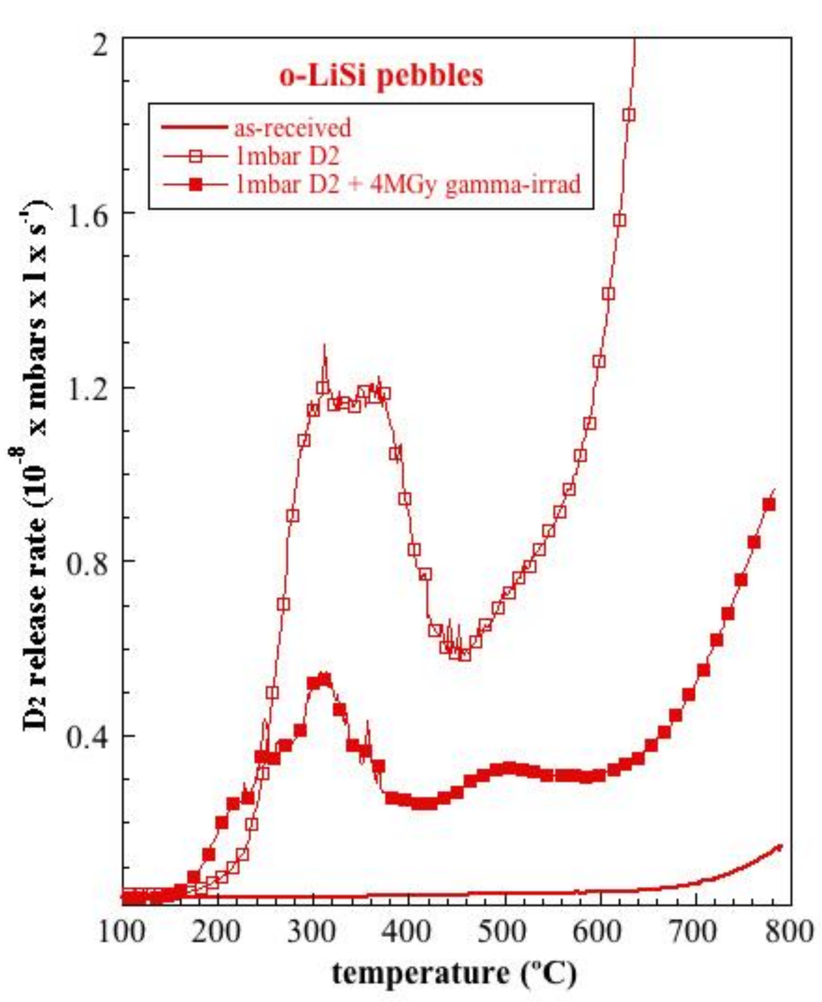
Thermally induced desorption (TID) measurements.

Samples were:

1.- dehydrated at 400ºC/2hs in vacuum;

2.- exposed to a D_2 gas atmosphere at RT inside a pressurized steel capsule (1mbar) (***sorption process of unirradiated condition***) and while being γ-irradiated up to a total dose of 4 MGy and a dose rate of 1.8 Gy/s, inside a ^{60}Co pool facility (CIEMAT, Madrid, Spain) (***sorption process under irradiated condition***).

3.- ***Desorption process*** --- the D_2 release rate was followed up to 800 ºC at 10 C/min using a mass spectrometer (Pfeiffer Smart Test leak detector. Mass selected: 4 amu; Detection limit of 10^{-12} mbar x l x s^{-1}; Sensitivity better than $5x10^{-12}$ mbar x l x s^{-1} which is equivalent to approx 10^8 D_2 x s^{-1})



m-LiTi content mol%	Ea (eV) < 300ºC unirradiated	Ea (eV) < 300ºC gamma-irradiated
0	0.19	0.12

Two D_2 desorption processes:

* A low temperature, due to adsorbed D_2 in defects or impurities already present in the as-received condition.

* A high temperature process, represented by a continuous gas release of trapped gas in deep centers.

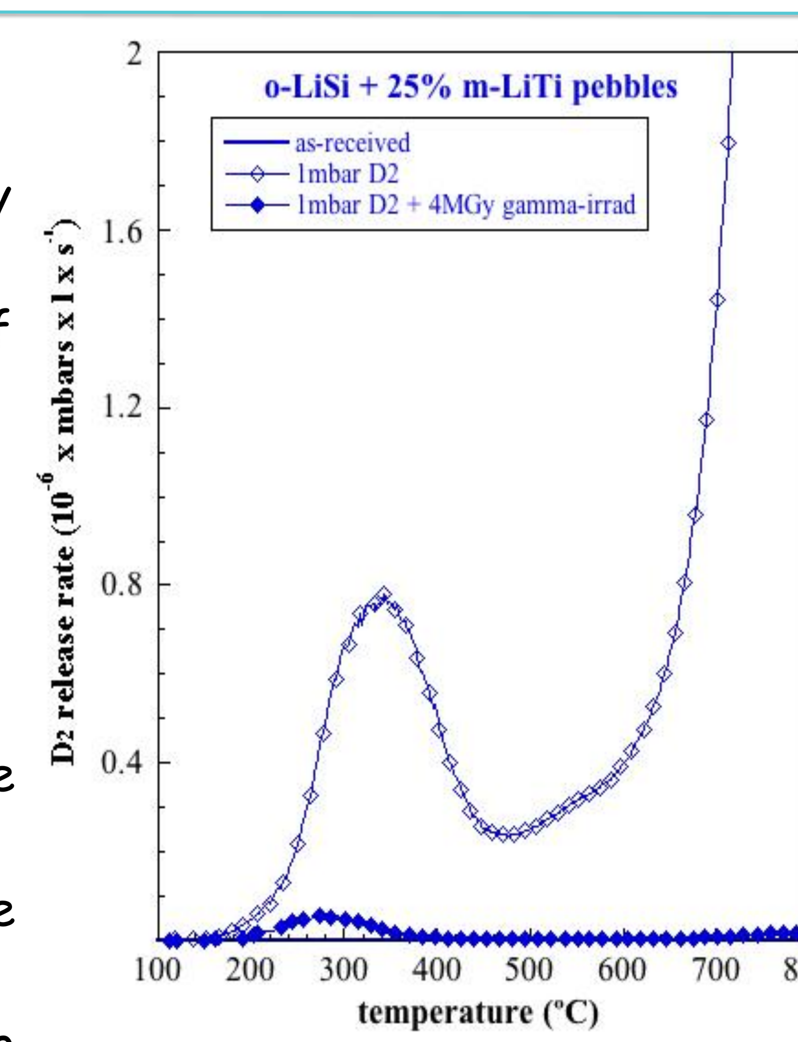
Lithium vacancies (V^-_{Li}), oxygen vacancies (F^+) … could be potential centers for ionic deuterium (D^+, D^-) to be trapped.

The D_2 release rate is notably <u>decreased</u> in the irradiated condition, which could be explained as:

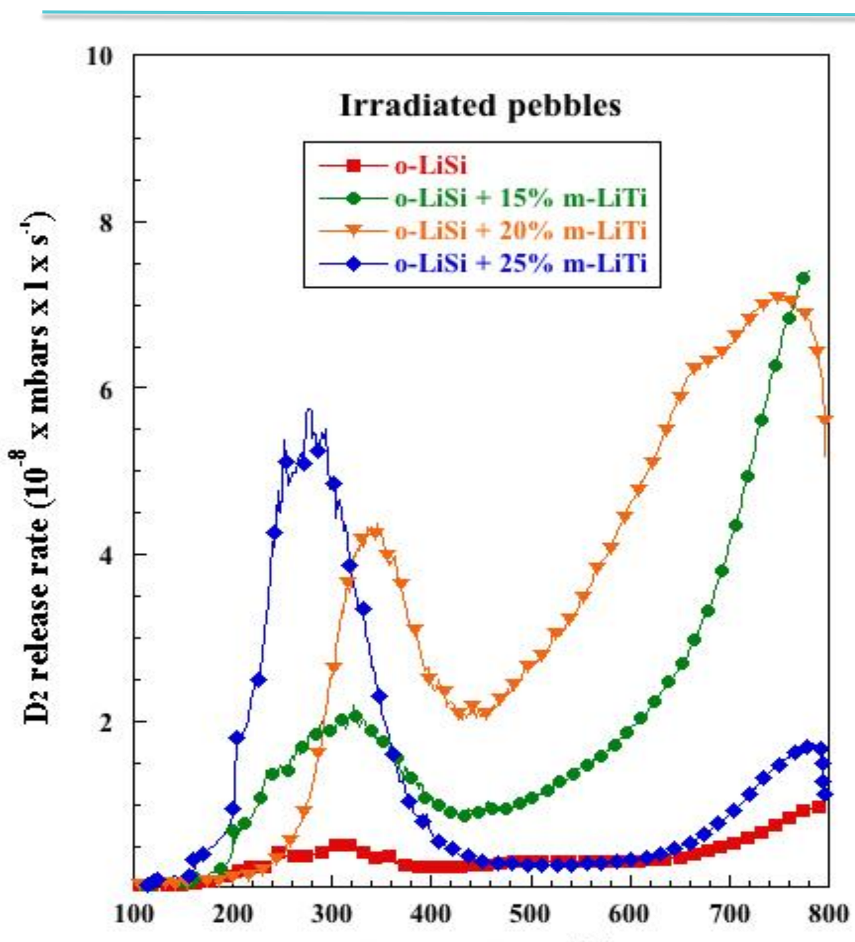
a) Mobile electrical charges during ionizing irradiation reduce the active centers for D_2 trapping;

b) D_2 is mainly trapped in deeper centers during irradiation. Consequently the desorption would occur at temperatures above the experimental range;

c) ^{60}Co gamma-radiation sputters oxygen from material surface, giving rise to D^+-$O^=$ association in the gas phase, decreasing the effective D to be sorbed and trapped.



m-LiTi content mol%	Ea (eV) < 300ºC unirradiated	Ea (eV) < 300ºC gamma-irradiated
25	0.18	0.13



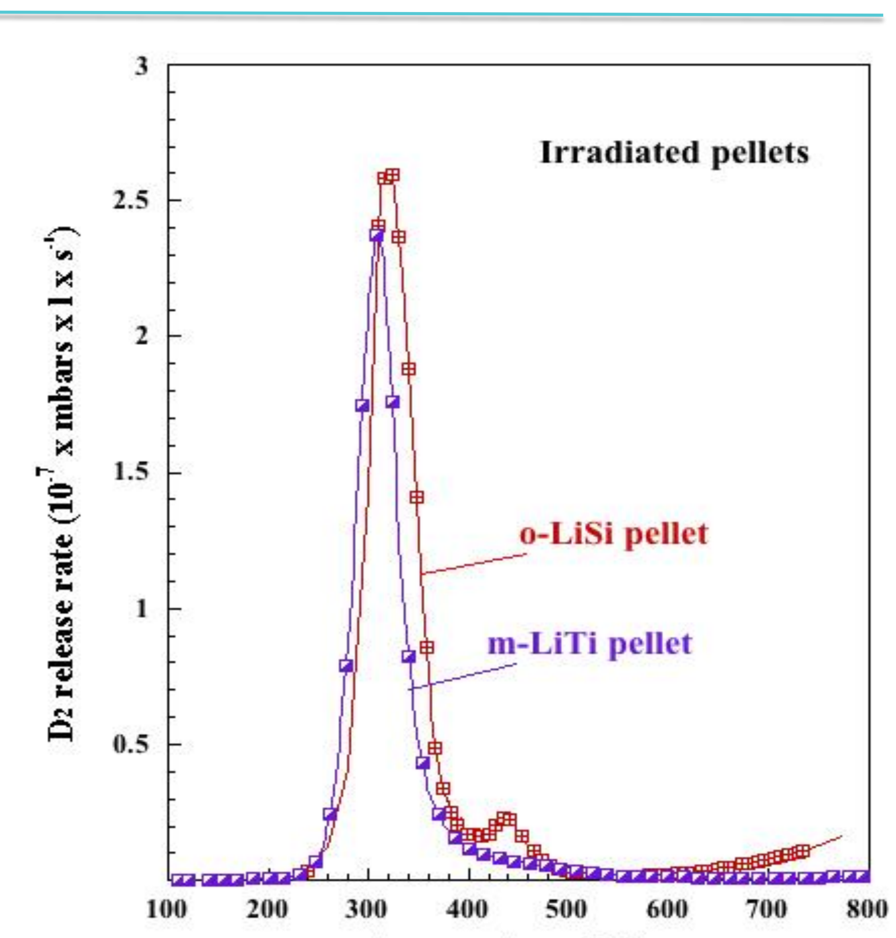
Varying m-LiTi proportion

The presence of the m-LiTi second phase does not provide new or different trapping centers involved in the sorption/desorption processes of D_2 gas.

These D_2 trapping centers are common to silicate and titanate crystalline structures BUT a greater number is introduced with the increase of m-LiTi content.

Pellets behavior

Desorption in pellet samples exhibits the same thermally-induced processes for D_2 release. Contrary to expectations, the ceramic characteristics (fabrication route, density, porosity) do not have any influence on the main desorption mechanisms of D_2.



CONCLUSIONS

= A low temperature process of low activation energy occurs below 300 ºC, suggesting that surface defects are acting as trapping centers for D_2.

= D_2 is also trapped in deeper centers, being released at temperatures above approx 500 ºC.

= Ionizing radiation (up to 4 MGy) modifies the electronic structure of intrinsic defects affecting the sorption and desorption processes.

= The involved trapping centers are common o-LiSi and m-LiTi crystalline structures. The presence of this second phase increases the number of active centers for D_2 sorption.

The desorption of deuterium, and by extension also the release of the tritium produced in the breeder, will be effective at fusion operational temperatures.

Contact: elisabettacarella@gmail.com