Deuterium retention via highly oxygenated lithium coatings

PP8.34

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Motivation

Background

- Lithium dramatically improves plasma performance in fusion devices, in part, by reducing deuterium recycling.
- This was initially suspected to be a result of LiD bonding.
- However, lithium attracts oxygen, and then *oxygen is the dominant mechanism for binding deuterium* in lithiated graphite.

Objectives

- Provide experimental and theoretical evidence demonstrating that oxygen is the dominant mechanism for binding deuterium in lithiated graphite.
- Identify all potential contributing sources of oxygen in this system, and qualify how much each oxygen source contributes towards oxygen enhancement.

Primary results

- Oxygen content dramatically increases during ion bombardment, but also increases during quiescent periods via gettering.
- Lithium deposit conjectured to contribute more strongly than other oxygen sources.

1 Surface Characterization Methods

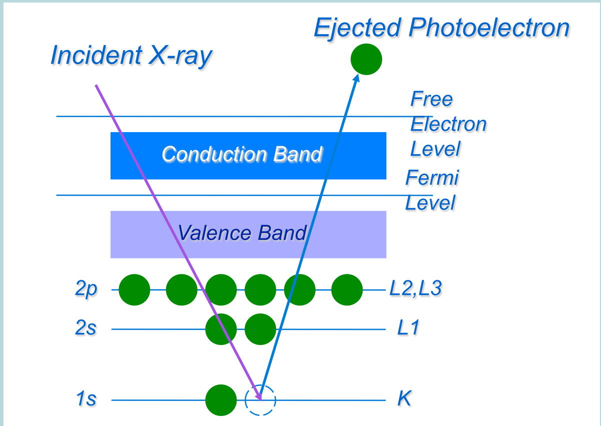
Photoelectron spectroscopy: The photoelectric process

- *XPS spectral lines are identified by the shell from which the electron was ejected (1s, 2s, 2p, etc.).*
- *The ejected photoelectron has kinetic energy:*

KE = hv – Binding E – Work Function

- *Energy of ejected photoelectron reveals characteristic information regarding the elemental composition of the substrate.*

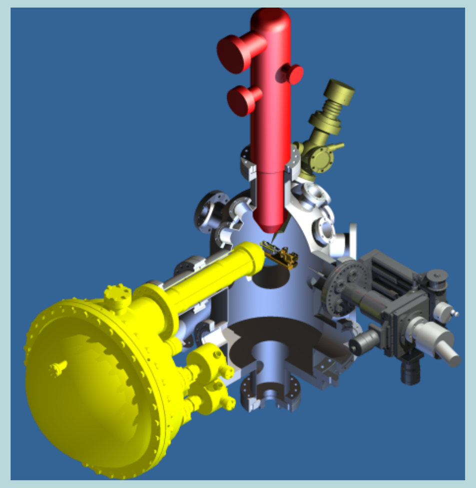
X-ray Photoelectron Spectroscopy

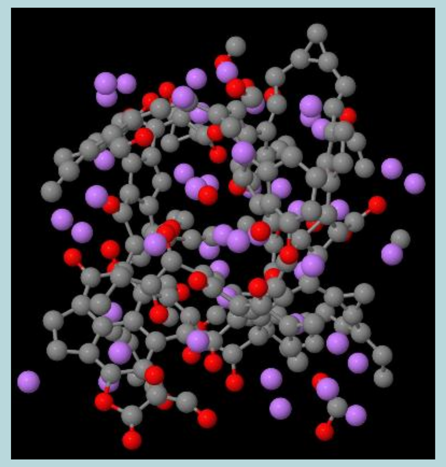


- Binding energy of ejected photoelectron is characteristic of the element from which it came.
- New peaks and peak shifts indicate new chemistry.

Facilities

Particle and Radiation Interaction with Hard and Soft Matter (PRIHSM) experiment is designed to be a versatile facility able to measure in-situ the surface evolution during energetic particle modification.





Atomistic simulations

- Self consistent charge-tight binding density functional theory (SCC-DFTB) is used to model the C-Li-O-D system.
- Performed on ORNL Kraken super computer.
- 'Nearest neighbors' analysis looks at the final location of implanted D+ and provides a qualitative indication of likely binding pairs.

2 Experiment: oxygen content increases

XPS reveals dynamic nature of oxygen

Virgin Graphite

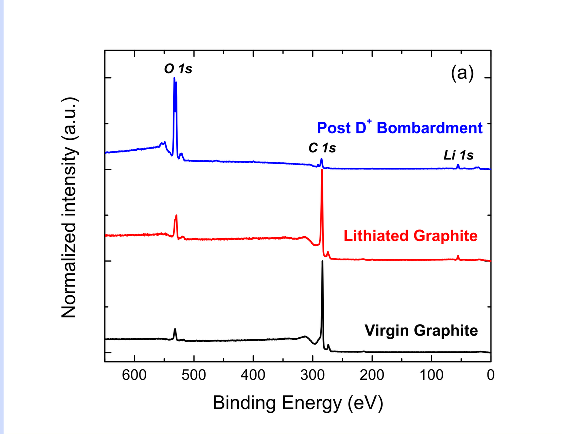
- Virgin ATJ graphite contains ~5.4% surface oxygen concentration.

Lithiated Graphite

- Depositing lithium general increases oxygen concentration by 2x.

Post D Bombardment

- Deuterium irradiation further increases oxygen content to 20-45%.



X-ray photoelectron spectra show oxygen surface concentration increases from ~5 to 8% immediately after depositing lithium. After deuterium bombardment, the oxygen content increases significantly to ~36%.

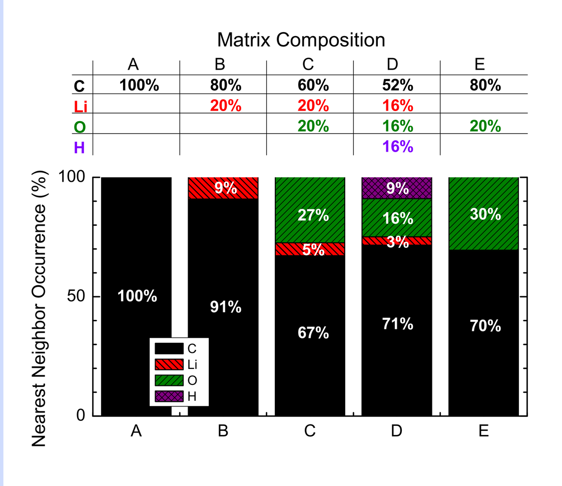
3 Model: deuterium prefers oxygen

DFTB simulations show that deuterium prefers to be near oxygen.

- Five different matrices prepared (A-E) and bombarded by 1000s of D ions. Final rest location of ions indicates possible binding pairs.

Results

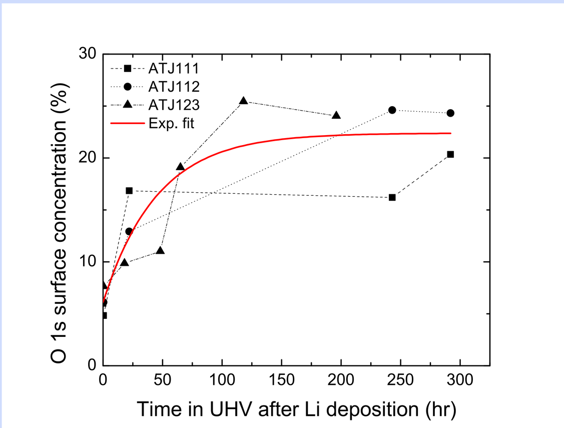
- 20% lithium in carbon leads to *9% deuterium-lithium* nearest neighbors.
- 20% oxygen in carbon leads to *30% deuterium-oxygen* nearest neighbors.
- When lithium and oxygen are present in equal quantities, oxygen dominates.



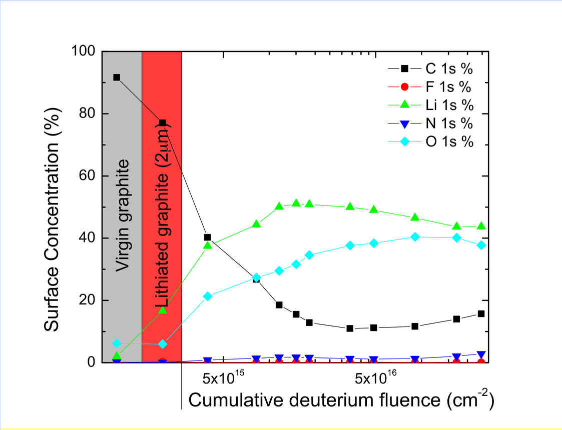
Integrated distribution of deuterium nearest neighbors after deuterium ion bombardment for various matrix compositions. When oxygen is present in the matrix, it becomes the predominant nearest neighbor, an indication of binding pairs, to deuterium.

4 Trend of increasing oxygen

1. **The oxygen content increases as the sample sits in UHV (10^{-10} Torr) for long periods of time.**
   - Gettering is a slow, thermodynamic process (10-100s of hours).
   - Possible curve fit suggests an ultimate oxygen surface concentration of ~20%.



The oxygen surface concentration of lithiated graphite slowly increases from ~5% to more than 20% as the samples sits in 10^{-10} Torr UHV over the course of 100s of hours.



Data points in the left-hand gray panel represent the graphite sample in its virgin state; data in the adjacent red panel are taken after depositing lithium. Next, a 1 minute irradiation commences (3.9×10^{15} cm^{-2}) and increases the oxygen content from 6.0% to 21.3%. Subsequent irradiation steps further increase the oxygen content to 40.4%.

2. **The oxygen content increases as the deuterium fluence increases.**
   - Pause D+ irradiation frequently to investigate oxygen concentration at low fluences.
   - After a *1 min deuterium* irradiation (~10^{15} cm^{-2}), the oxygen content *more than triples*!
   - Oxygen concentration plateaus at ~40% (for this sample).

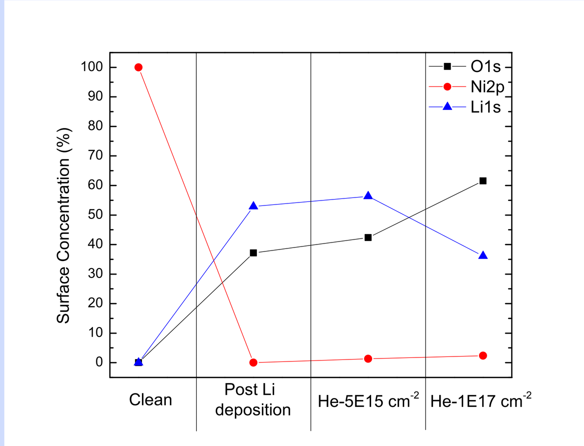
5 Three potential oxygen sources exist

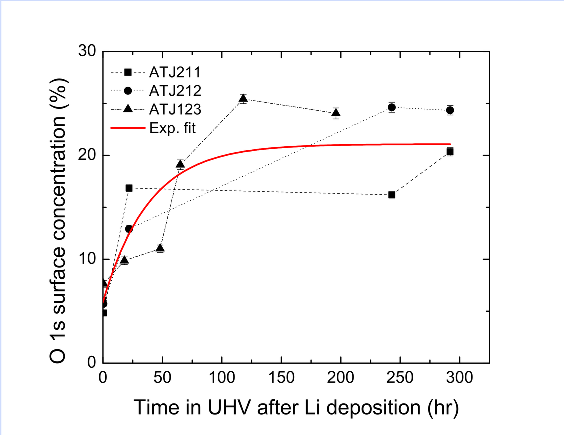
1. ***Oxygen from within the bulk graphite***

To test, use a different substrate:

- Deposit lithium on nickel.
- Bombard with helium ions.
- Quantify surface concentrations.

Results: Clean nickel has no detectable surface oxygen. Ion bombardment of lithiated sample shows large increase in oxygen!
Conclusion: Oxygen from within bulk sample is not a significant contributor.





1. ***Oxygen gettered from ambience (partial pressure, walls, sources, etc.)***

To test, monitor oxygen gettering during quiescence:

- Deposit lithium on graphite.
- Periodically perform XPS over long UHV (10^{-10} torr) waiting period.
- Quantify surface concentrations.

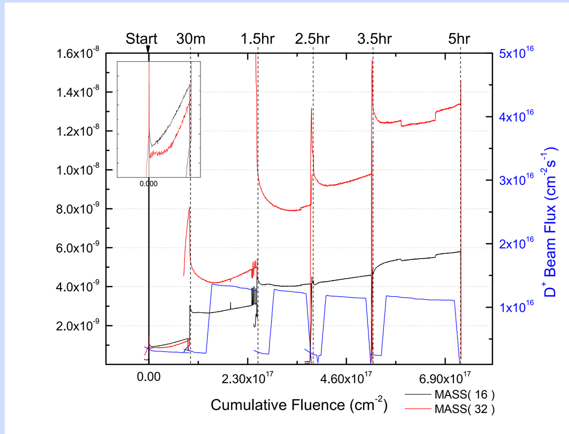
Results: Over the course of 100+ hours, the oxygen concentration increases to ~20%.
Conclusion: Gettering contributes substantially, however on long time scales irrelevant to ion bombardment.

3. ***Oxygen pre-trapped within lithium***

To test, monitor oxygen emission/consumption during ion bombardment.

- Negative slope indicates consumption from ambience
- Positive slope indicates oxygen liberation.

Results: Oxygen liberation lowest during period when surface oxygen concentration increases by 26.6%. Need further experiments to link to source of oxygen.
Hypothesis: Oxygen pre-trapped in lithium deposits contribute more significantly than other sources.



6 Conclusions

- ❑ Oxygen is the dominant channel for retaining deuterium in lithiated graphite.
- ❑ Oxygen content in lithiated graphite increases as:
  - ❑ The samples sits in UHV (10^{-10} Torr) for 100s of hours.
  - ❑ Deuterium irradiation fluence increases.

Discussion

- ❑ Oxygen comes from three possible sources:
  - ❑ Bulk sample – not a significant contributor in these experiments.
  - ❑ Ambience – not a significant contributor.
  - ❑ Lithium deposit – conjectured to contribute most.

Future work

- ❑ Run more detailed residual gas analysis to conclusively determine the contribution of oxygen liberated from lithium deposit.

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