Temperature-Programmed Desorption Study of the Reaction of Water and Epitaxial Graphene on Silicon Carbide

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Benefits of Nuclear Power

- Much less greenhouse gases released
- Produce more electric power for less cost
- Technology already understood

However....
Safety and Fuel-Cladding

Disaster at Fukushima 1 Nuclear Power Plant in March of 2011 has brought concern over the safety of nuclear power.

The tsunami waves destroyed cooling system and caused the reactors to overheat.

Zircaloy fuel-cladding oxidized and produced hydrogen gas.

Hydrogen gas led to explosions in reactors.

Need for longer-lasting materials for fuel-cladding that are able to hold up to loss-of-cooling accidents (LOCA).
Fuel-Cladding

Fuel-cladding is used to separate the radioactive fuel from the coolant so the neutrons, gamma rays and alpha and beta particles that are created during the fission process are not released into the environment.
There are several factors to consider when deciding on the type of Fuel-Cladding material but most notably:

- Transparency to neutrons
- High Thermal Conductivity
- Low Thermal Expansion Coefficient
- Chemical Compatibility
- Mechanical Strength
Current Materials for Fuel-Cladding

Meeting the aforementioned requirements Zirconium (Zr) and alloys of Zr (Zircaloy and ZIRLO) are the most used fuel-cladding materials.

However, at high temperatures and high energy radiation exposure (like LOCA) exothermic reactions of Zr with water/steam yield hydrogen in great amounts which could lead to

1) Hydrogen Embrittlement
2) Production of hydrogen gas

Hydrogen Gas explosion at Fukushima Dai-ichi
Additional Materials for Fuel-Cladding

Promising research into Silicon Carbide (SiC) as a replacement for fuel-cladding

Epitaxial Graphene (EG) on SiC meets the thermal and mechanical needs for fuel-cladding and since graphene has a hydrophobic nature it is thought to be less reactive to steam and therefore less likely to produce hydrogen.

Single-layered graphene has not been fully tested under extremely high temperatures or under conditions that mimic LOCA.
Epitaxial Graphene

Mono-layered graphene in a 2-D honeycomb arrangement of carbon atoms

Studied for its electrical, chemical, thermal and mechanical properties.

Epitaxial means that the graphene is grown on a surface of a crystal material and mimics the arrangement of that substrate.

Sample from Dr. de Heer’s group using a “Confinement Controlled Sublimation” to grow high quality layers of EG on SiC.
Raman Spectroscopy

Used to detect low frequency modes in a sample.
Inelastic scattering (KE is not conserved)

Principle of Raman Spectroscopy

http://www_che.engr.ccny.cuny.edu/courses/che5535/lecsld002.htm
Raman Data of EG on SiC

The sample of EG on SiC showed the characteristic G peak (1500 – 1620 cm$^{-1}$) and 2D peak (2700-2750 cm$^{-1}$) of SiC was subtracted.
Monitoring Desorbed molecules of EG on SiC

To test the thermal stability of the sample the desorption defects were measured with a special interest in hydrogen gas.

Temperature Programmed Desorption (TPD): the technique of observing desorbed molecules from a surface as the surface temperature is increased via a mass analyzer. A quadrupole analyzer commonly used.

As desorption occurs ions are released into the chamber and guided to the quadrupole.

The ions move through the electric field formed by the rods. The strength and frequency of the field dictates if an ion of a particular mass will pass through the rods and therefore counted by the detector or collides into a nearby surface.
TPD

Designed and created by Orlando Group

Ultra High Vacuum chamber (1.4 E-8 Torr)

Sample stage with thermocouple attached to a thin tantalum wire acting as a spring clip, allowing direct contact with the sample.

Nitrogen chamber to keep out pump oil and other impurities.

Temperature range 500°C to 1000°C (5 minute intervals)

Temperature controlled by adjusting current 7 A – 19/20 A

Empty chamber, SiC, EG on SiC (with and without water – dose set for 2.5 E -10 A.

Quadrupole analyzer

Quadera software
Thermal Programmed Desorption
TPD Data of H₂
TPD Data of CO$_2$

![Graph](image)

- Run 1 Empty Chamber_H2O
- Run 1 SiC_H2O
- Run 1 EG on SiC_No_H2O
- Run 1 EG on SiC_H2O

**Axes:**
- **Y-axis:** Ion Current Intensity (A)
- **X-axis:** Time (s)
TPD Data of CO
TPD overlay of CO and CO$_2$
FINAL THOUGHTS

Graphene appears to not react with water at high temperatures to produce hydrogen gas.

More research on EG on SiC in LOCA conditions.

Further quantify and characterize the TPD tested sample.

1) High Resolution Transmission Electron Microscopy
2) Raman spectroscopy

Future Direction

1) Wrapped EG around Zircaloy
2) Grow Chemical Vapor Deposited (CVD) on Zircaloy

Testing both in TPD chamber to analyze H2 desorption.
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References


References


References


