Revisiting the use of SiC as a Post Irradiation Temperature Monitor

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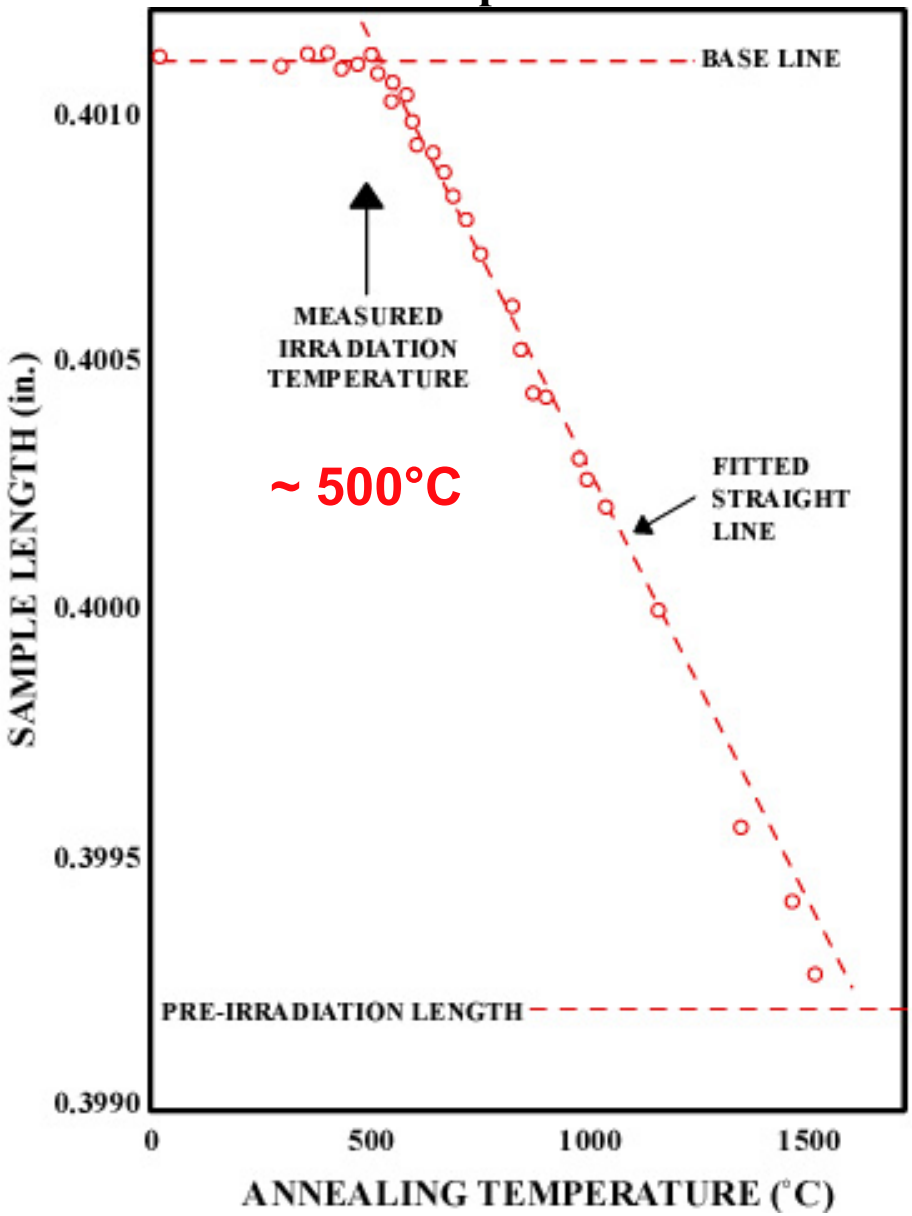
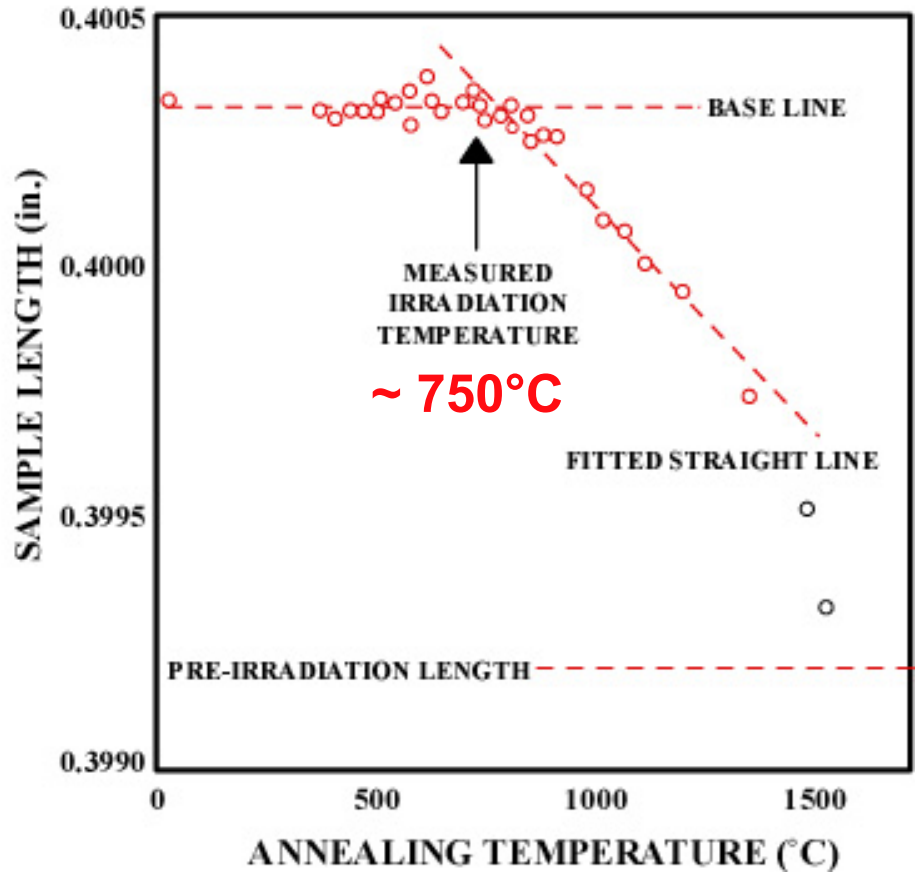
Presented at the
International Symposium on Materials Test Reactors
Idaho National Laboratory
September 28,2009

Introduction

- In 1961 Pravdyuk reported that neutron irradiation-induced lattice expansion of silicon carbide annealed out when post -irradiation annealing temperature exceeds the irradiation temperature.



- SiC has been used since as an ex-situ temperature monitor in fission irradiation experiments, mostly at high dose, and mostly by isochronal annealing of length change.

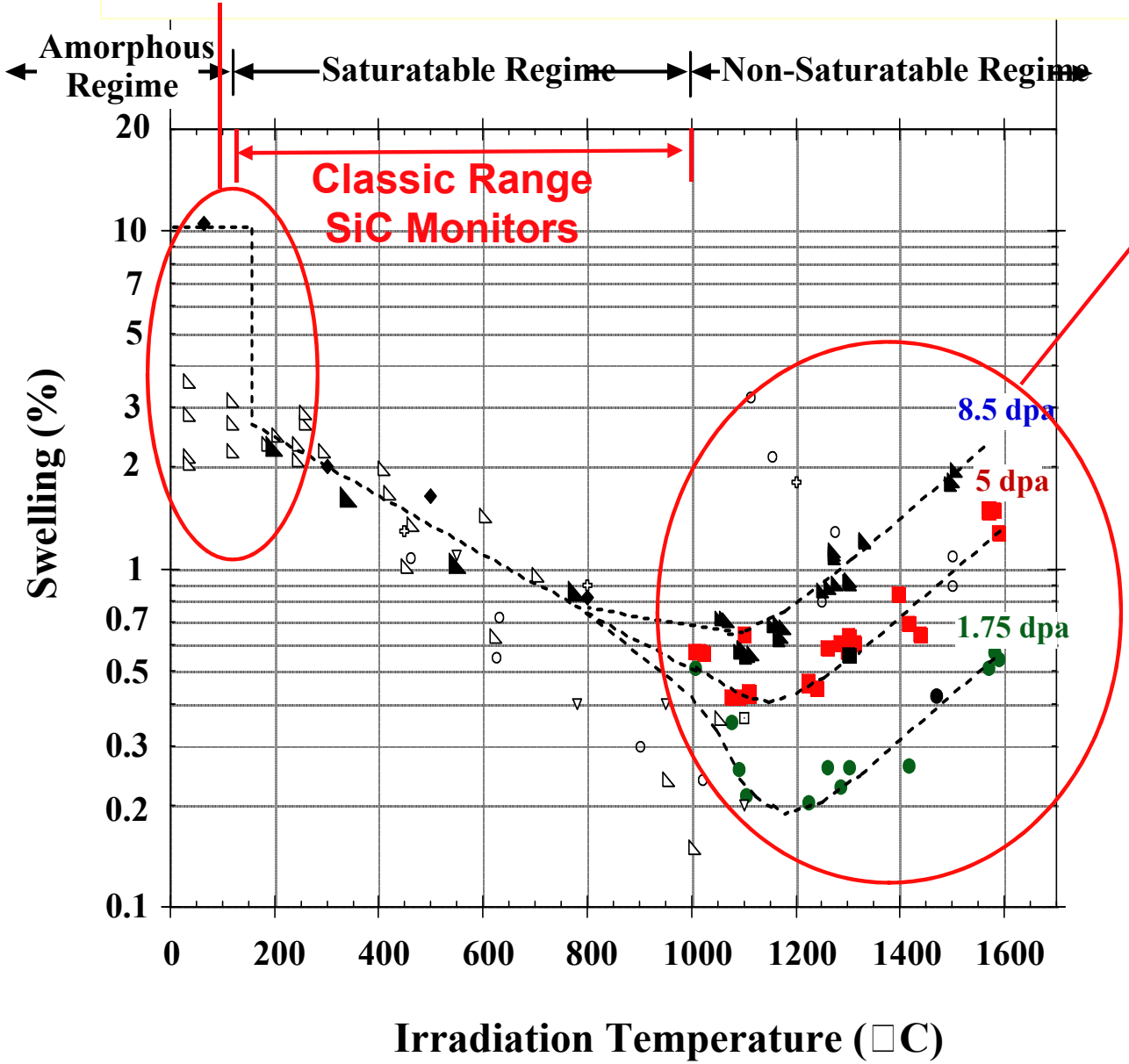


- Other measurement techniques: lattice parameter (Miyazaki 92), thermal and electrical conductivity (Price 72) have been studied, though showed no improvement over length change.

This paper revisits the use of SiC temperature monitors in light of new materials, improved measurement techniques, and additional information regarding SiC irradiation damage.

Amorphization causes transition from ~1% to ~3.5% linear swelling.

Question: Can monitors be used in amorphization regime?



Complex defects cause non-saturated void swelling

Question: Can monitors be used in void-swelling regime?

	CVD SiC Circa 1970	CVD SiC Circa 2000
Density (g/cc)	3.2	3.2
Thermal Conductivity (W/m-K)	~70	70-400
Electrical Resisitivity* (ohm-cm)	~10^5	1-->1000

* Dependent on dopant levels

Part 1 : Techniques

Dimensional, Density, Thermal and Electrical Conductivity

- Palentine (1976, 1980) performed extensive analysis to determine uncertainty in measured temperature using dimensional measurements:

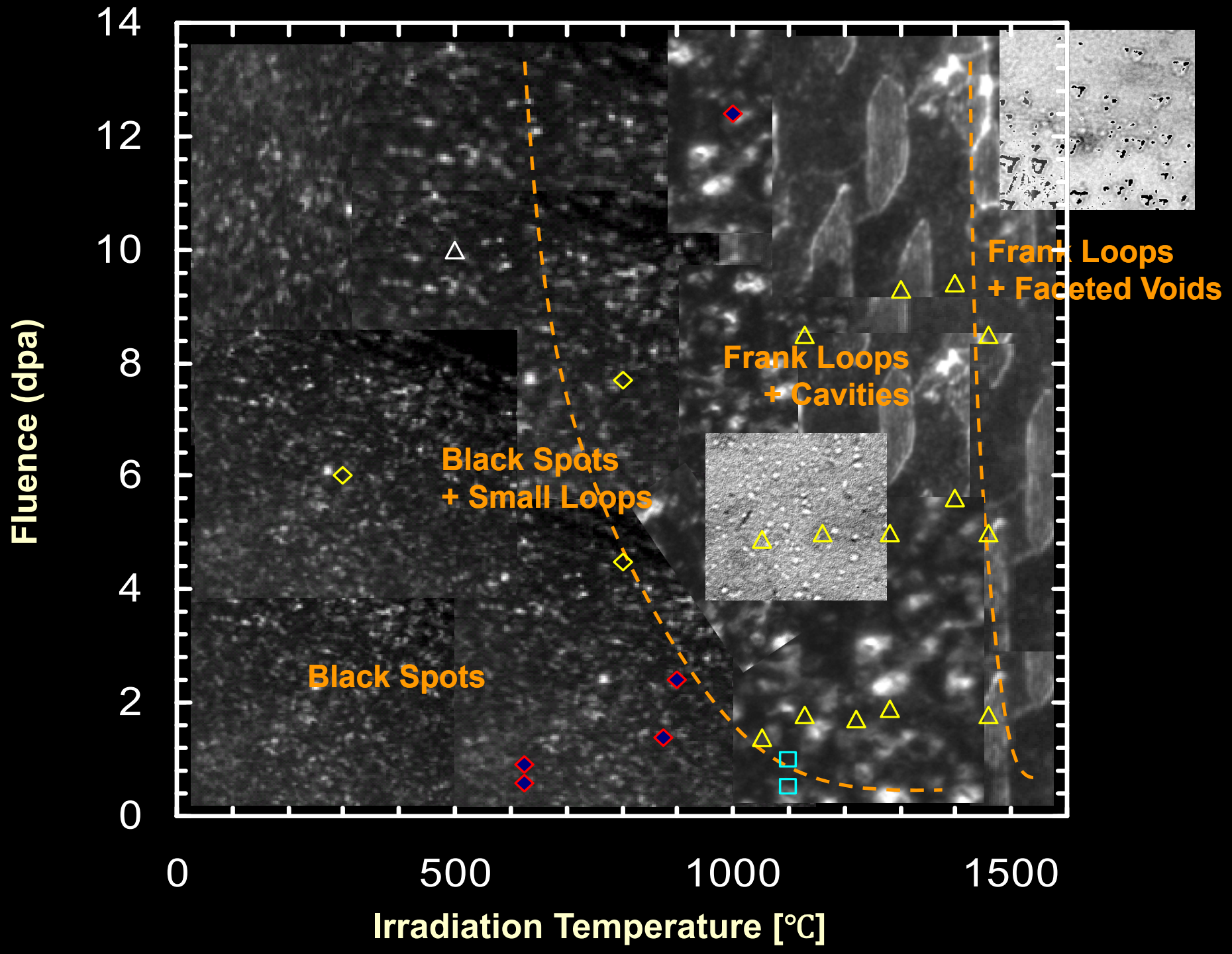
  12.7 mm long, 3.17 mm diameter.
  extreme precision in flatness and parallelism
  sets of 12 measurements
  rigorous statistical treatment

  -- > for 450°C irradiation, 95% confidence band was 12°C
  700°C " 76°C

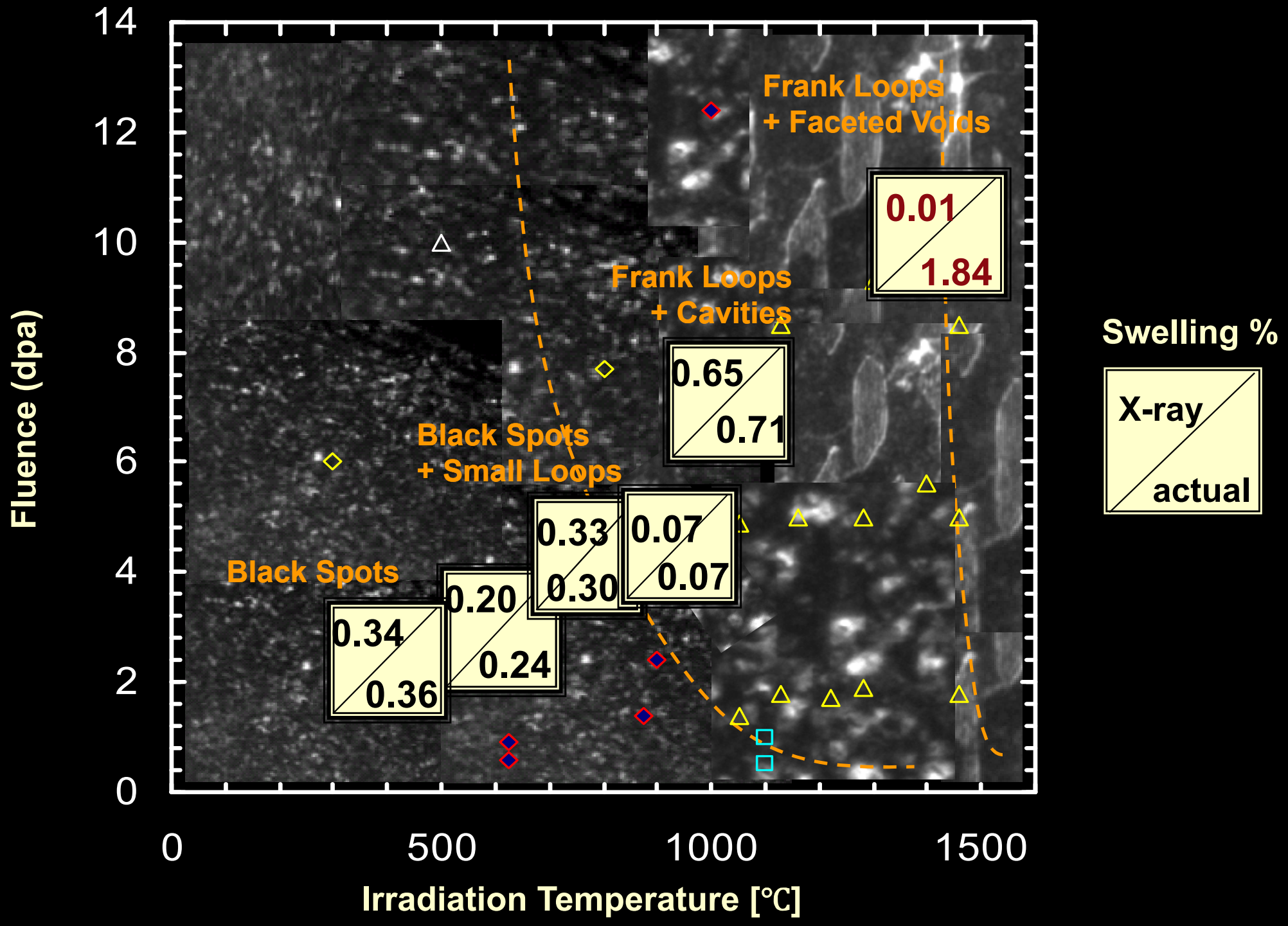
  --> furthermore: Palentine(76) and Maruyama(99) conclude that SiC temperature monitors, using the length change methadologly, overestimated irradiation temperature by 100°C !!!

However, in both cases, SiC used was not theoretically dense and not stoichiometric. It is recommended that high-quality CVD SiC be used for temperature monitors.

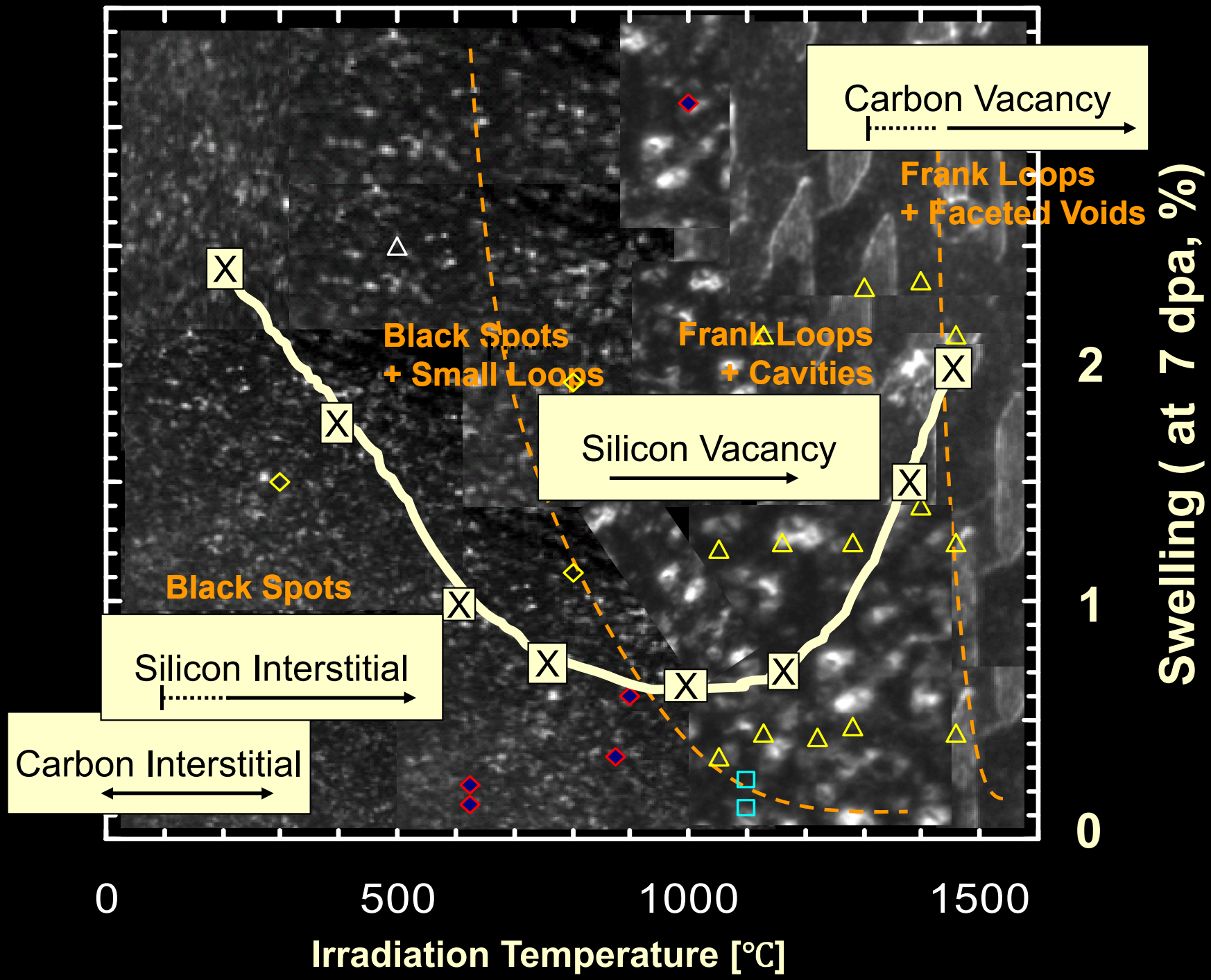
Microstructure of SiC Under Irradiation



SiC Density by x-ray and Gradient Column



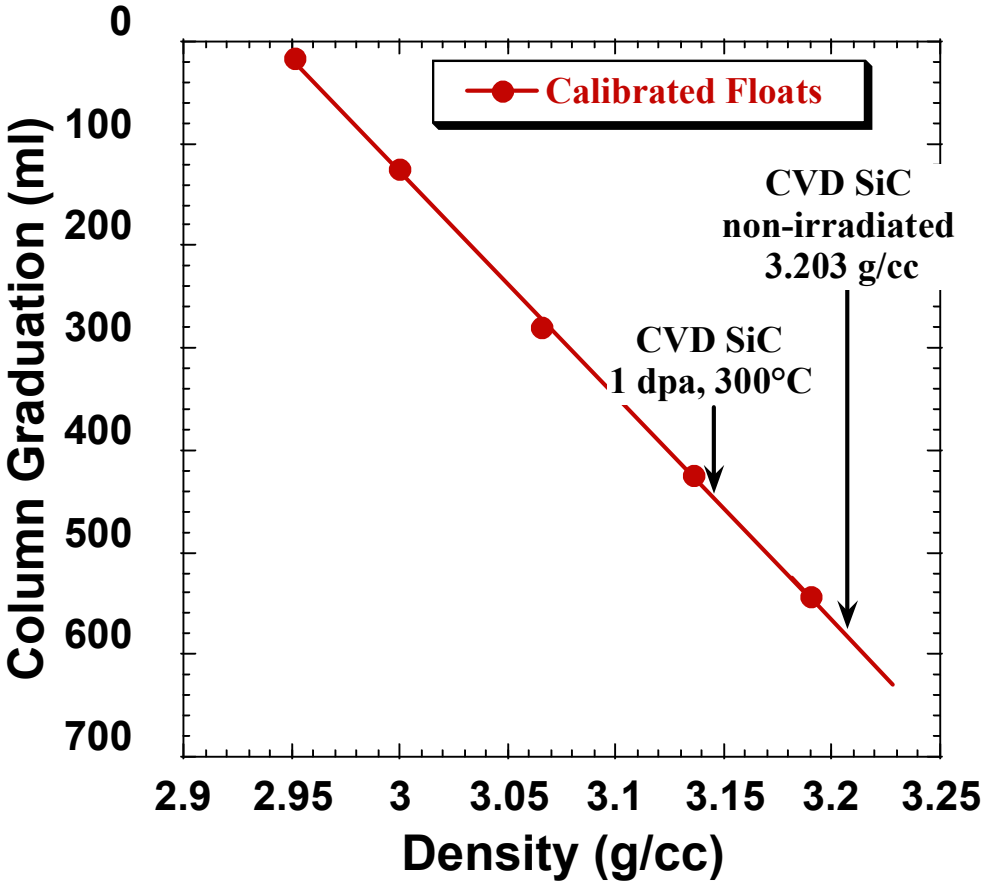
Density at ~ 7 dpa

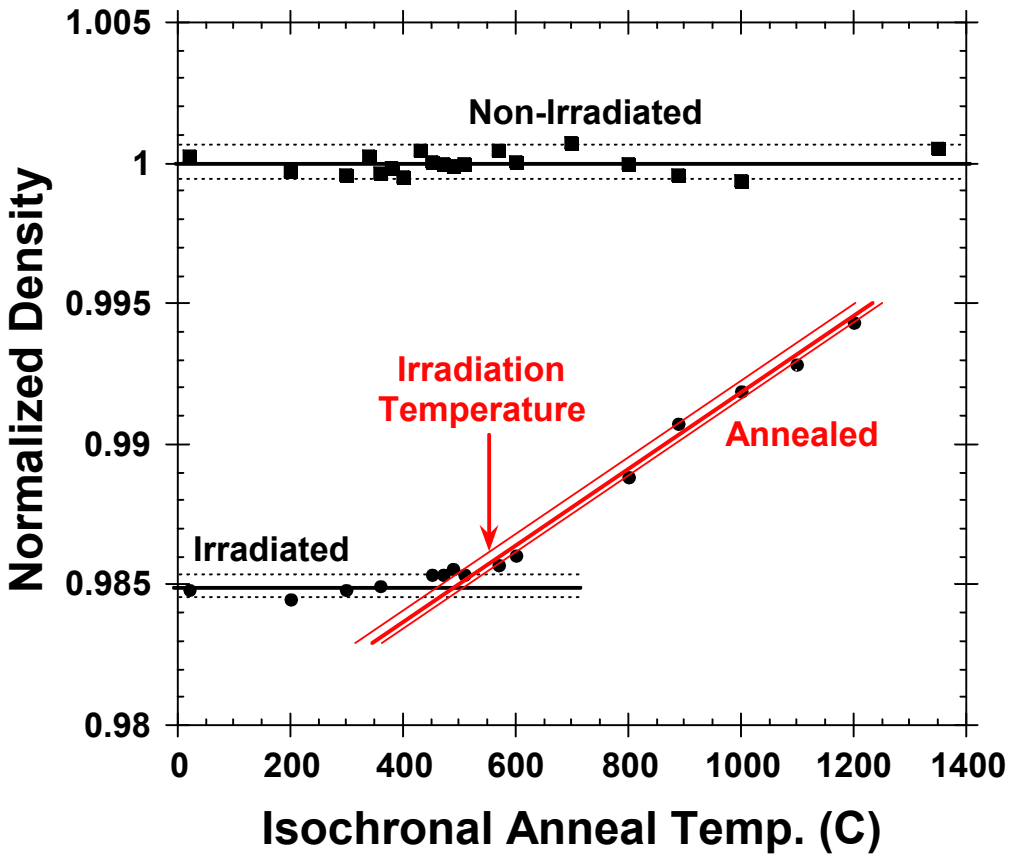


Techniques : Density

- Direct density measurements allow the ability to measure small, irregular monitors.

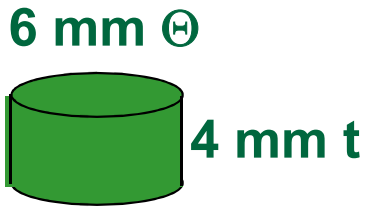
- Methods : Archimedes and other displacement techniques do not rival dimensional measurement for accuracy. However, the density gradient column technique gives exceptional accuracy. Standard measurement accuracy 0.003 g/cc, or 0.1%.

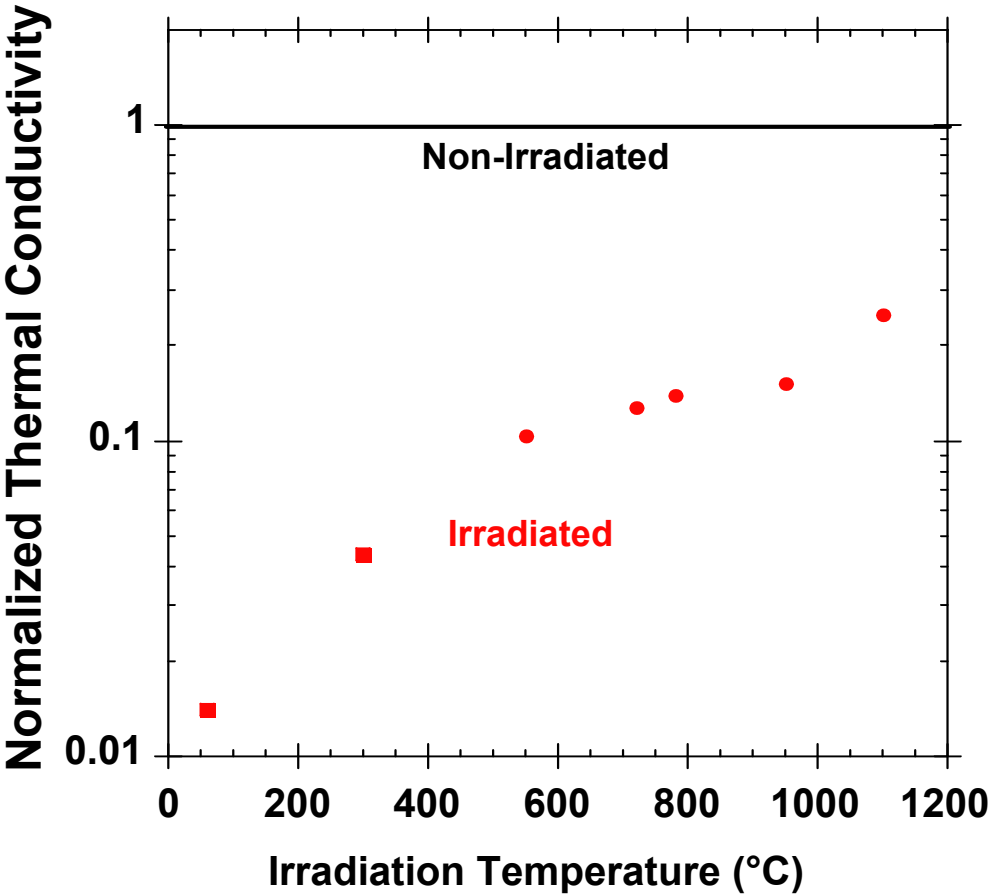


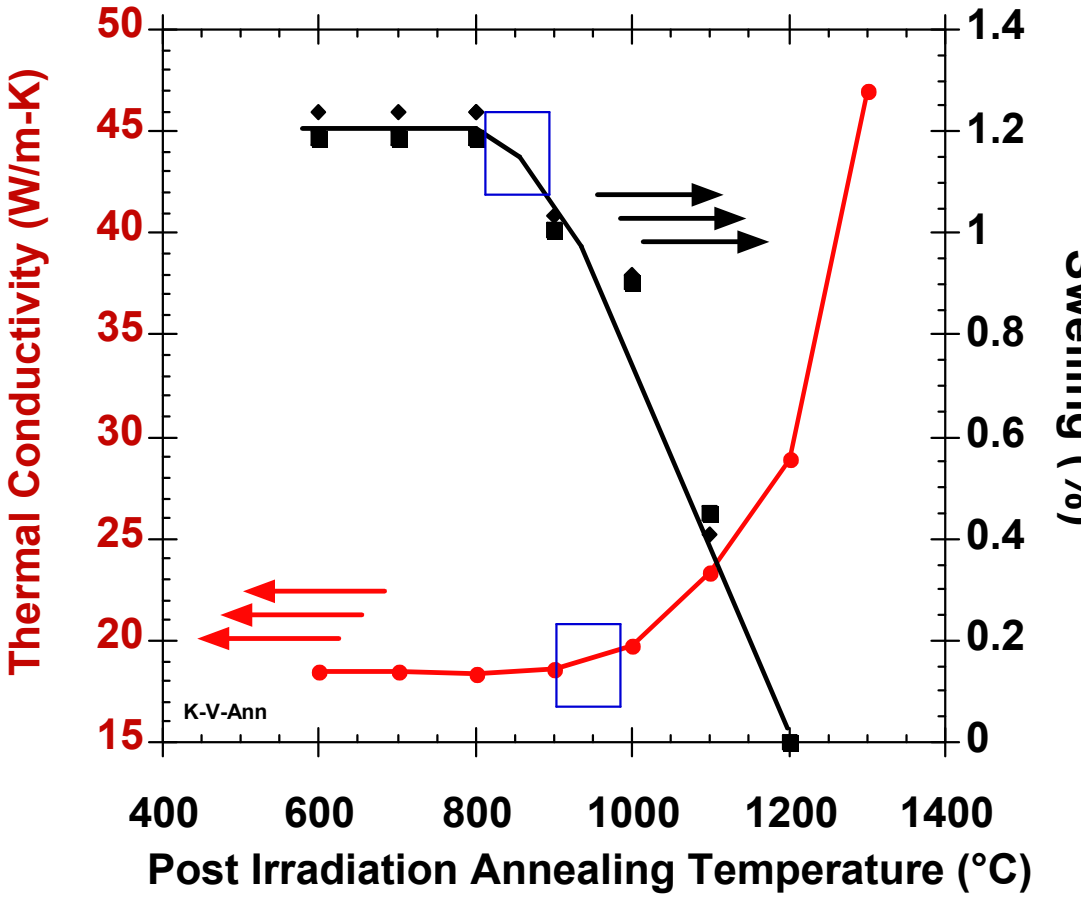


Techniques : Thermal Conductivity

- Thermal conductivity is drastically effected by the creation and annihilation of vacancies and vacancy clusters. The relative change is much greater than that the irradiation-induced swelling.

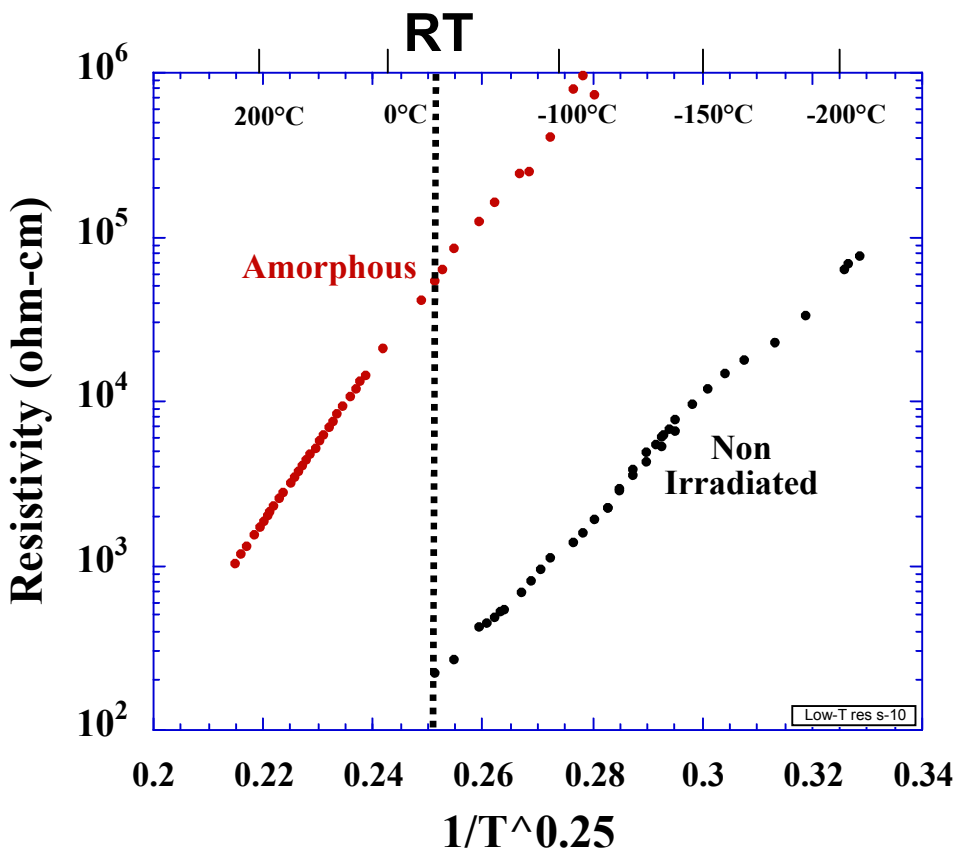


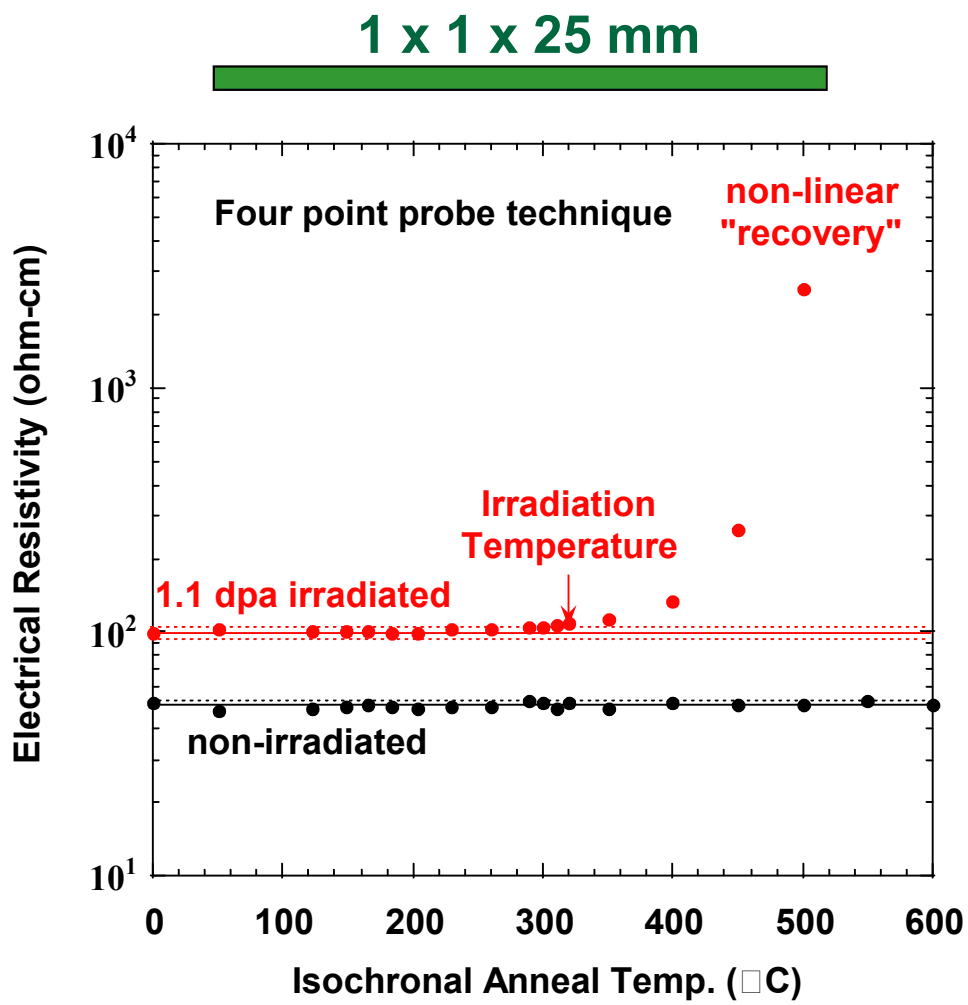




Techniques : Electrical Conductivity

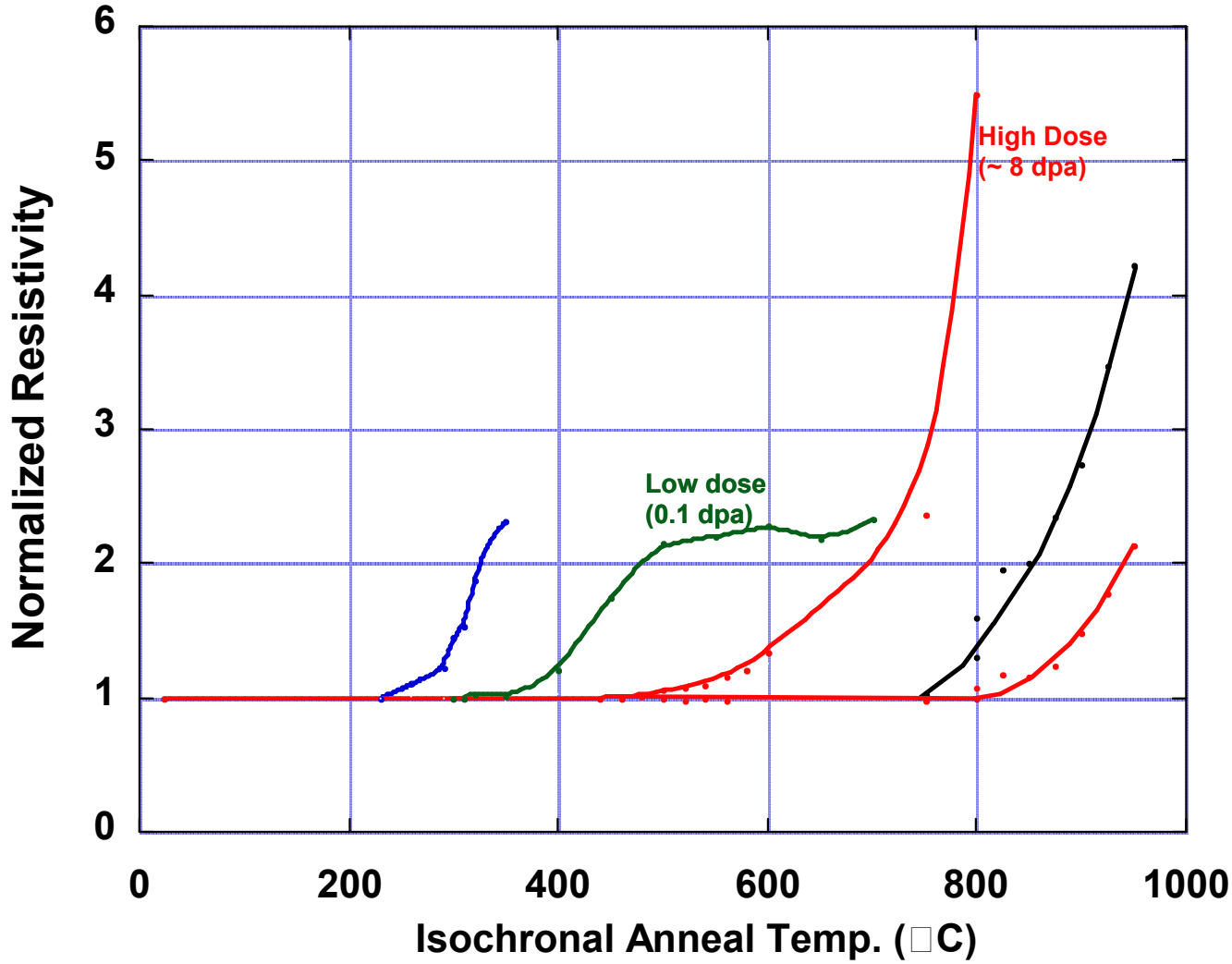
- As irradiated electrical conductivity in SiC is change by a combination of effects:
  - --> Nuclear transmutation doping: $^{30}Si(n,\gamma)^{31}Si \rightarrow ^{31}P$ (n-type dopant)
  - --> Boron (p-type acceptor) burn-up
  - --> Increased dangling bond density due to elastic collision (increase conductivity)



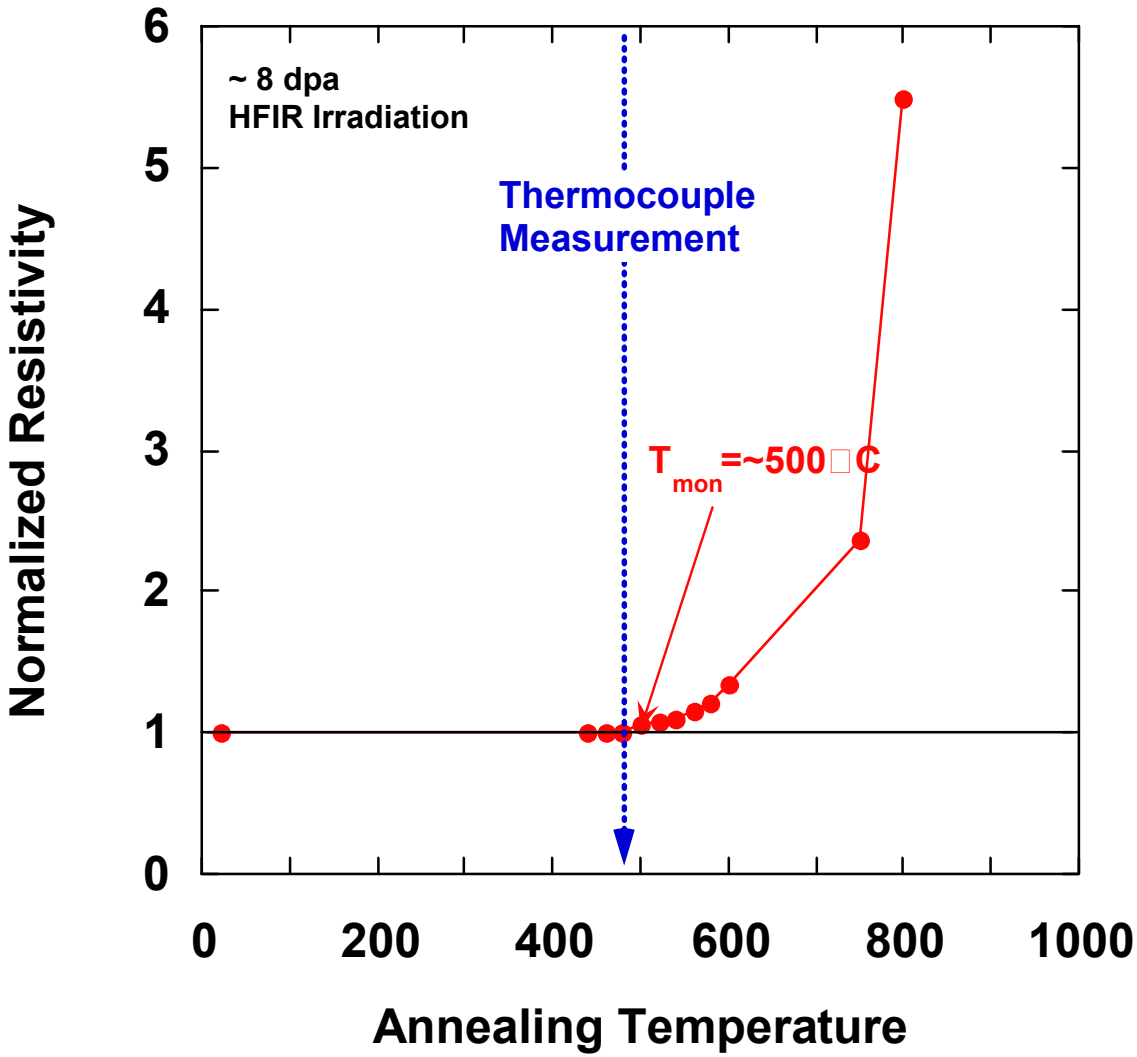


- Extreme care must be taken during measurement as SiC conduction is very temperature (3.6% / °C) and position sensitive.

Electrical conductivity increases or decreases with irradiation depending on the type and concentration of donors, and the neutron dose. However, it appears that crystalline SiC can be used as a temperature monitor over a wide temperature and dose range.



Heads-up Comparison to thermocouple ~ 8 dpa, HFIR



Direct comparisons have been carried out for 1 dpa, ~ 300°C (HFBR at BNL) and 8 dpa, ~ 500°C (HFIR at ORNL.) The discrepancy between thermocouple and SiC temperature monitor is within the technique error (~20°C)

Summary of Experience

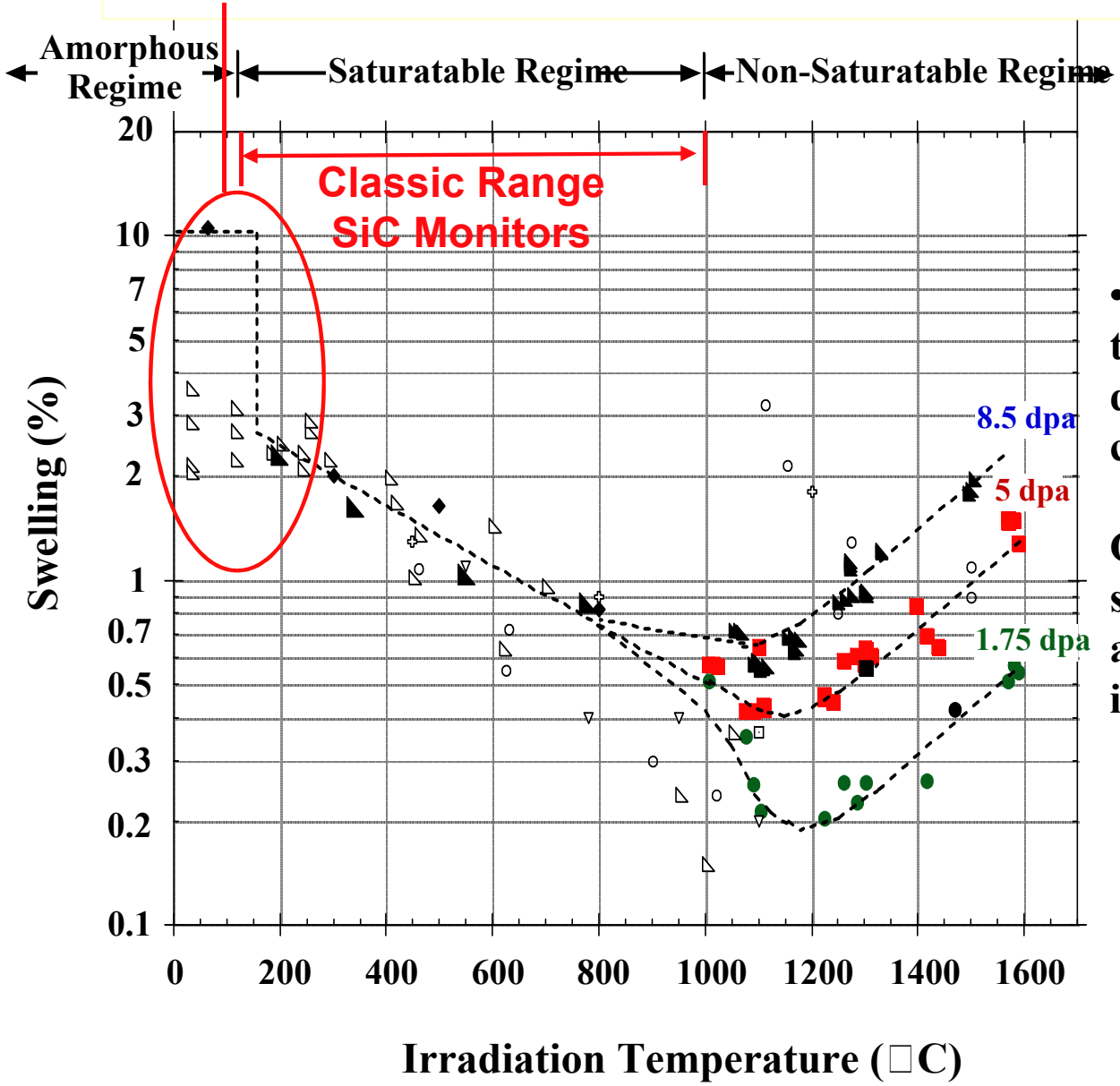
	Measurement Accuracy %	Property Change @ 500°C Irradiation %	Recovery Upon Anneal (% / 10°C)	Recovery / Accuracy	Recommended Sample Type	Technique Accuracy and comments
Dimensional Change	0.05	0.5	~ 0.007	~0.14	12.7 mm L, 3.17 mm Diam Extreme || Extreme Flatness	100°C overestimate? Accuracy 12°C at T_{irr} 450°C 70°C at T_{irr} 700°C
Electrical Resistivity	0.01-1	N/A*	~ 0.3	~.6	Bar 0.75 x 1 x Length	Within 10°C
Thermal Conductivity	3 - 5	90	~ 0.08	~ 0.02	2-4 mm L, 6-10 mm diam	Within 25°C Good for when length is limited
Density Gradient Column	~ 0.02	0.5	~ 0.02	~ 1	random, small	100°C overestimate? Within 20°C Time consuming unless multiple samples irradiated. Good for small sample
Lattice Spacing						Accuracy uncertain

* Extreme variability

Part 2 : Measurement at below 150°C

Amorphization causes transition from ~1% to ~3.5% linear swelling.

Question: Can monitors be used in amorphization regime?



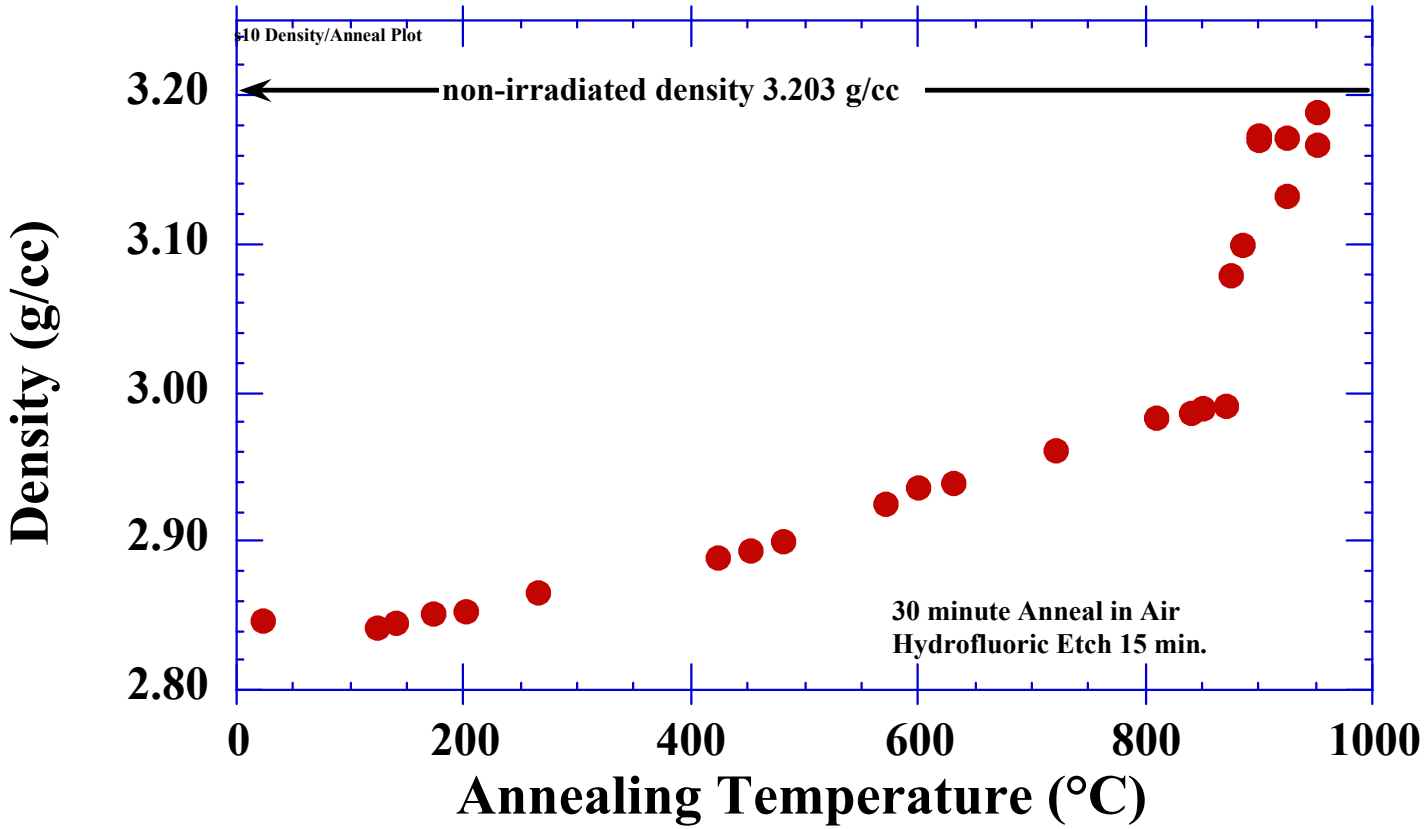
- **The previous techniques have relied on the annealing of simple point defects, or defect clusters into a more ordered crystalline system.**

Question : How will the amorphous structure and properties behave for annealing temperatures above the irradiation temperature?

Recovery of ~ 80-100°C amorphized SiC

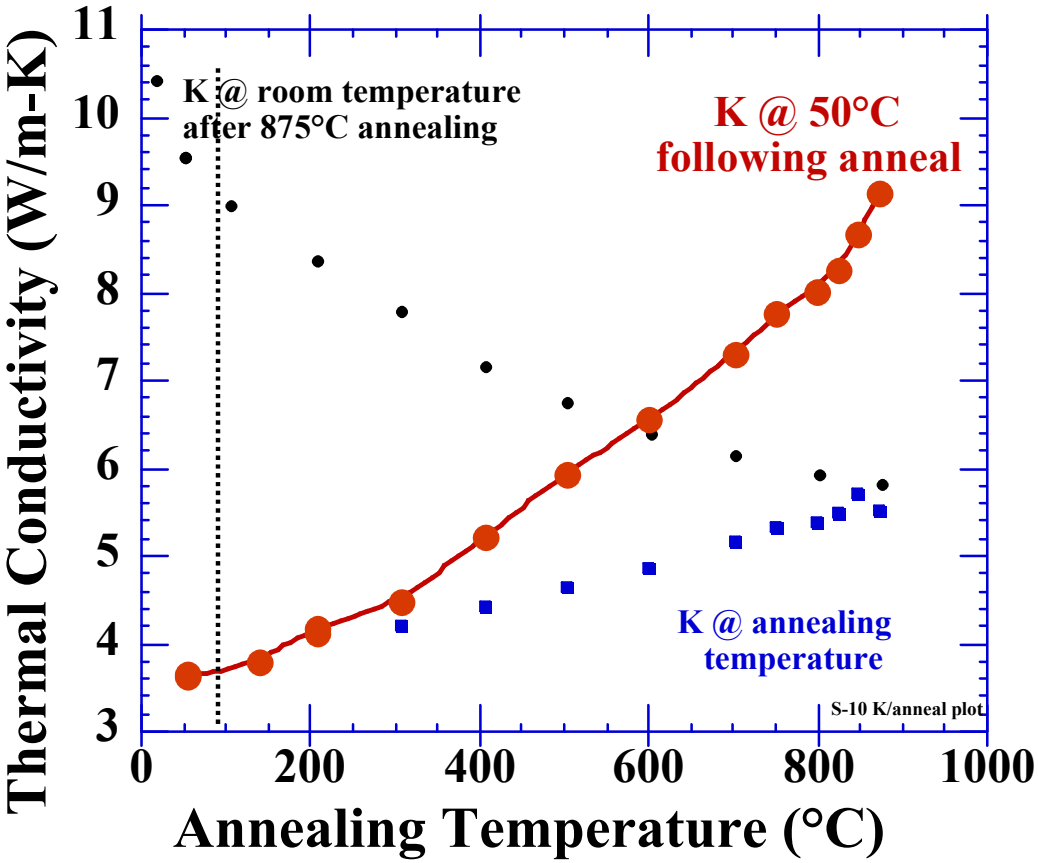
For amorphous SiC, structural relaxation (short range reordering) occurs from irradiation temperature to ~875°C yielding ~4.8% increase in density.

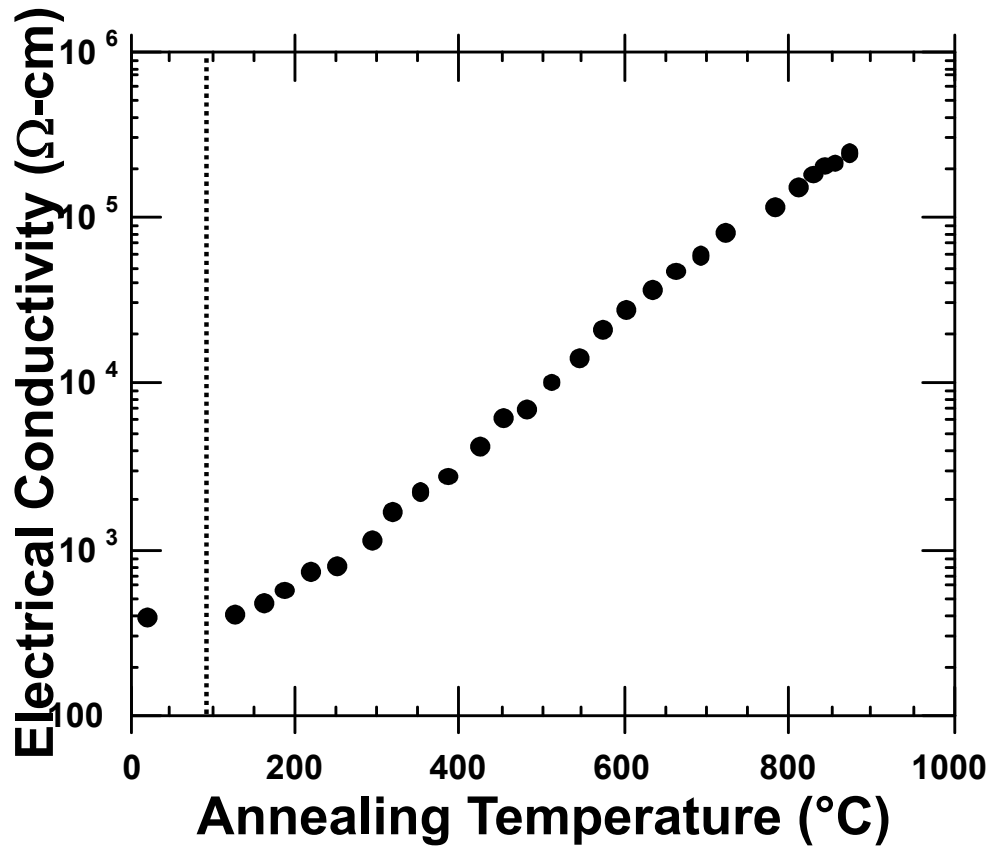
- Explosive crystallization occurs at 875-885°C, reaching theoretical density by 950°C.



- Though the mechanism is different, SiC can be used as a temperature monitor in the amorphous condition using the density gradient column technique.

Recovery of ~ 80-100°C amorphized SiC



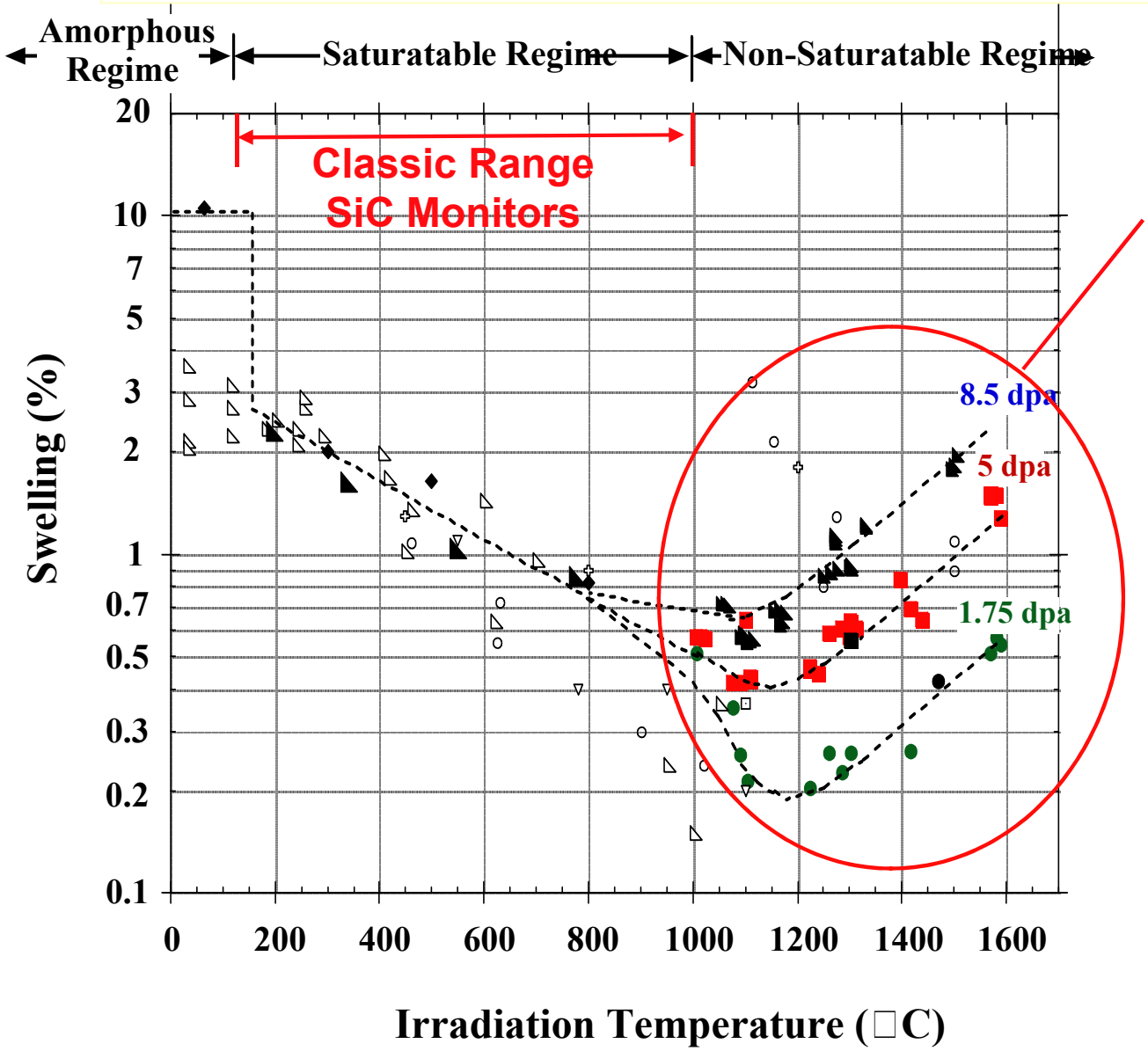


- **Local strain is reduced upon annealing leading to lower phonon scattering and dangling bond density. Both thermal conductivity and electrical resistivity recover due to relaxation of the amorphous structure.**

Part 2 : Measurement at above 1000°C

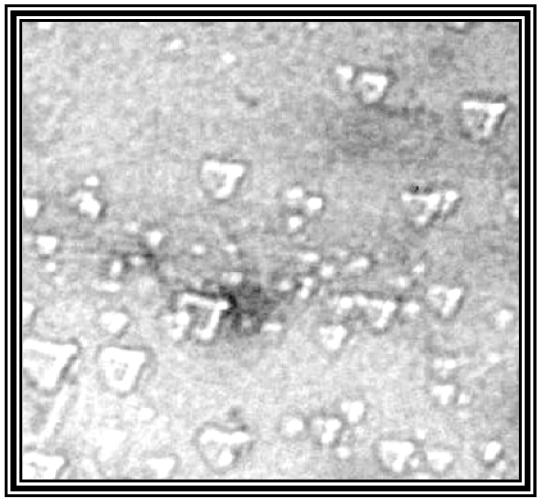
- The previous techniques have relied on the annealing of simple point defects, or defect clusters into a more ordered crystalline system. These simple defects are not created above 1000°C.

Question : Can annealing of complex defects and voids be used to determine irradiation temperature?



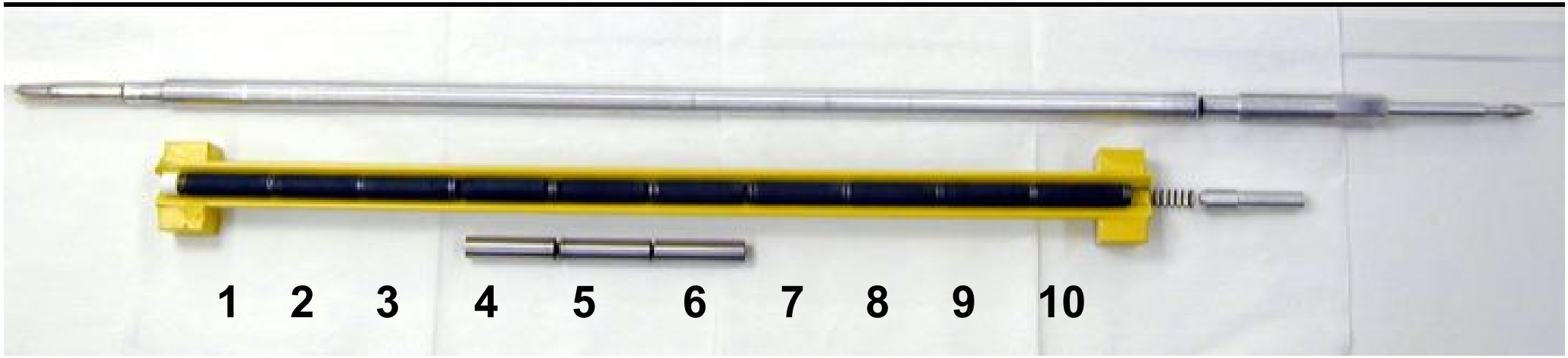
Complex defects cause non-saturated void swelling

Question: Can monitors be used in void-swelling regime?

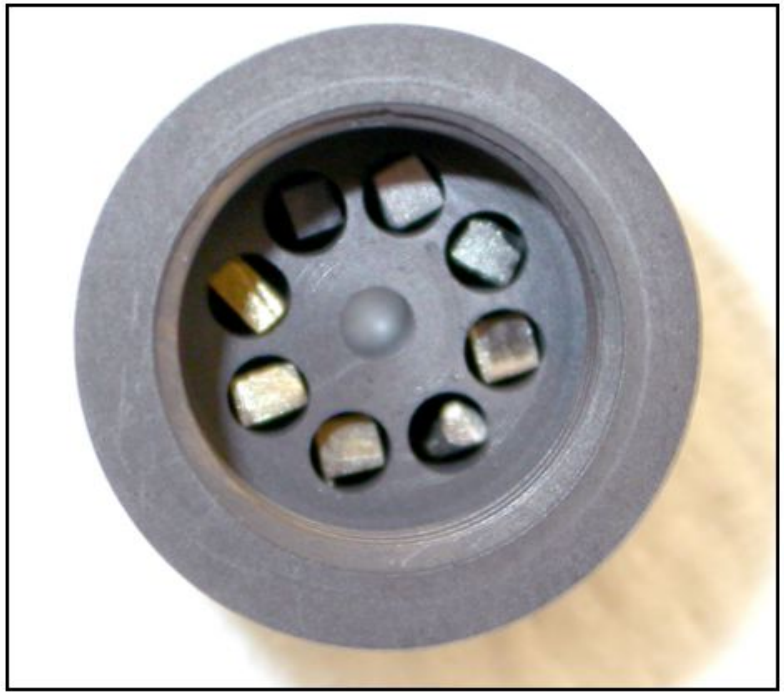


METS Experiment

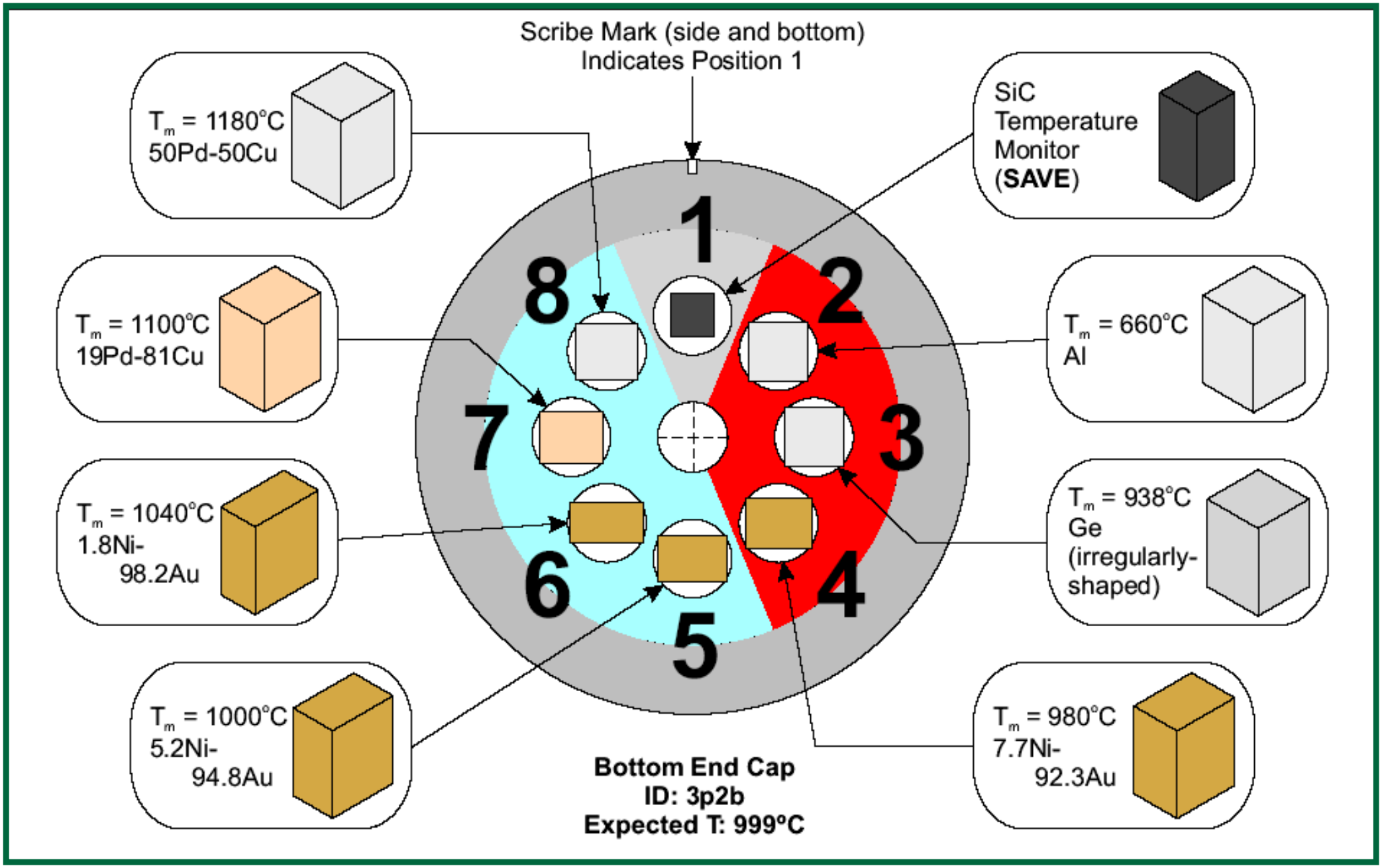
10 Subcapsules, up to 1600°C



Application of Melt Monitors



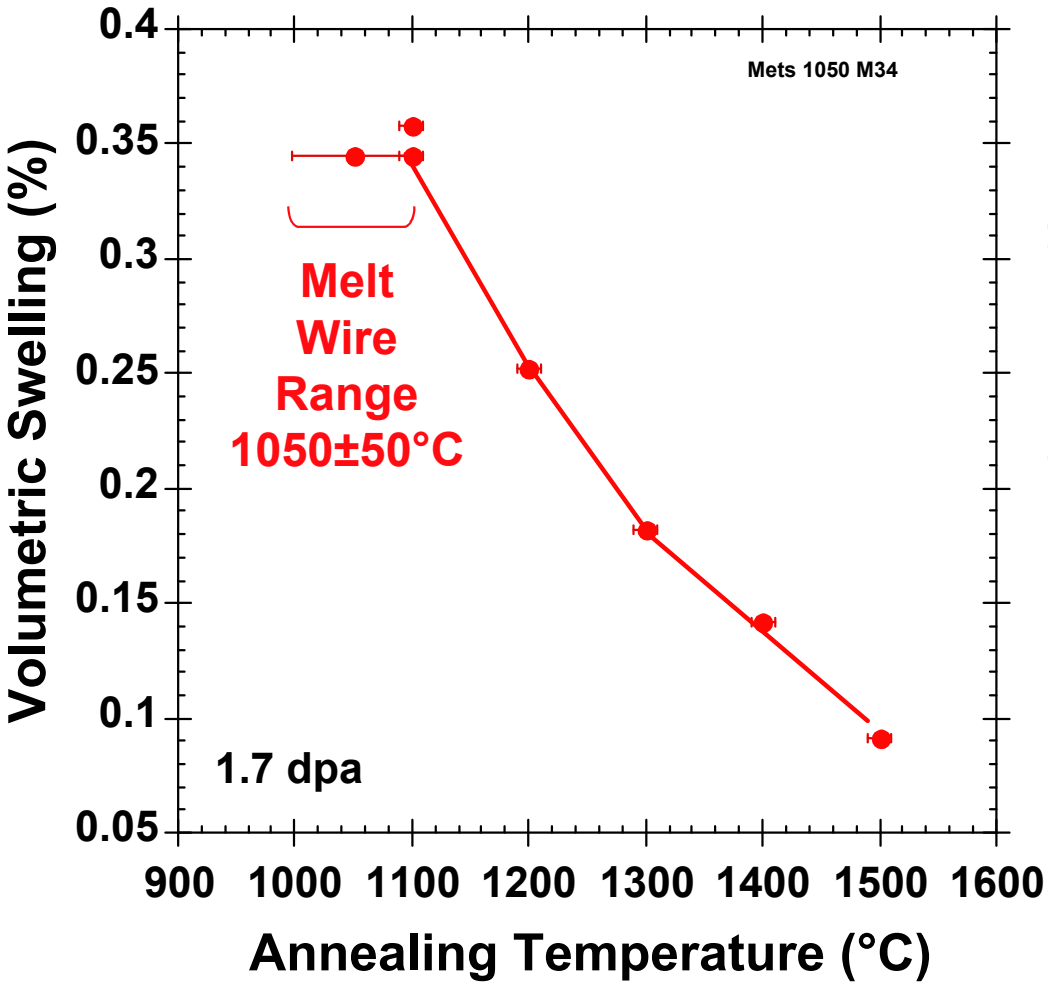
Melt wires : a combination of pure metals and alloys

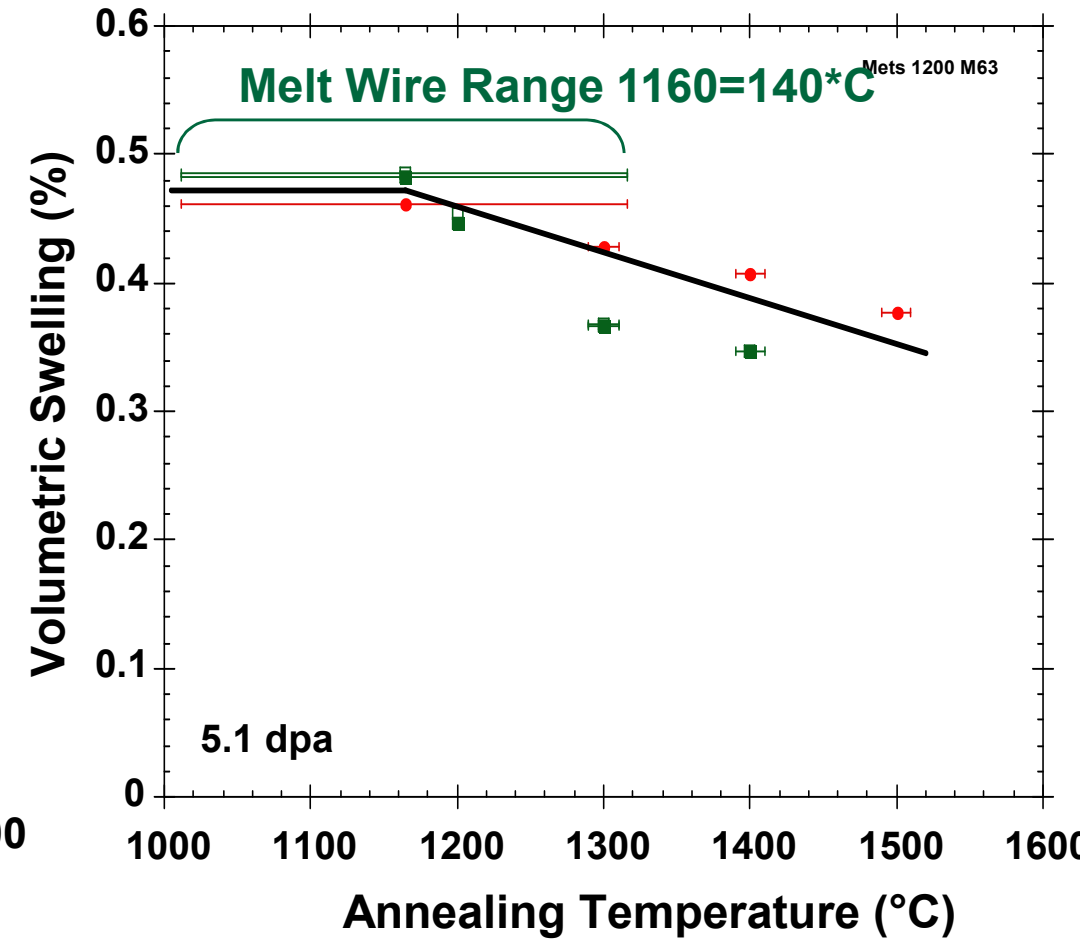


QuickTime™ and a
TIFF (LZW) decompressor
are needed to see this picture.

looking at these subcapsules

Part 2 : Measurement at above 1000°C





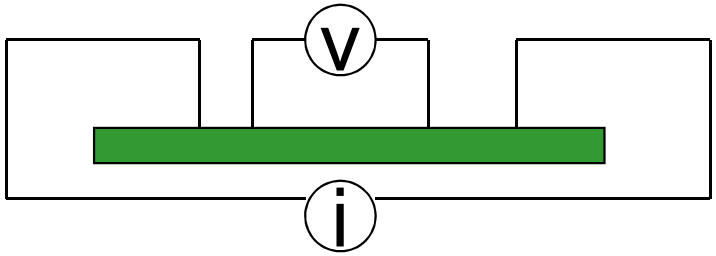
- At ~1050-1160°C, SiC is near its minimum irradiation-induced swelling. However, annealing above T_{irr} shows clear indication of recovery.

Concluding Remarks

- Based on work of Palentine(76) and Maruyama(99) the dimensional change technique yields fairly large discrepancy with thermocouple measurements (up to 100°C.) This discrepancy is not seen using electrical conductivity measurement (error is < 20°C) at 300 and 500°C irradiation.

- Based on large change in electrical resistivity upon annealing above the irradiation temperature, combined with the accuracy inherent in resistivity measurement, this technique is preferred.

- The following variables greatly effect accuracy of measurement of electrical resistivity
  - temperature (~ 3.6% error/°C)
  - inhomogenous electrical properties of material (spatial dependence)
  - constant inner electrode spacing
  - surface silica



- SiC appears to be an adequate temperature monitor in the amorphous regime