PennState College of Engineering

NucE 497: Reactor Fuel Performance

Lecture 34: CRUD

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Most material taken from slides by Dr. Michael Short at MIT

Today we will discuss CRUD formation on the cladding

- Module 1: Fuel basics
- Module 2: Heat transport
- Module 3: Mechanical behavior
- Module 4: Materials issues in the fuel
- Module 5: Materials issues in the cladding
    - Zirconium alloys and fabrication
    - Cladding creep and growth
    - Mechanical behavior
    - Oxidation
    - Hydride formation
    - **CRUD formation**
- Module 6: Accidents, used fuel, and fuel cycle

Here is some review from last time

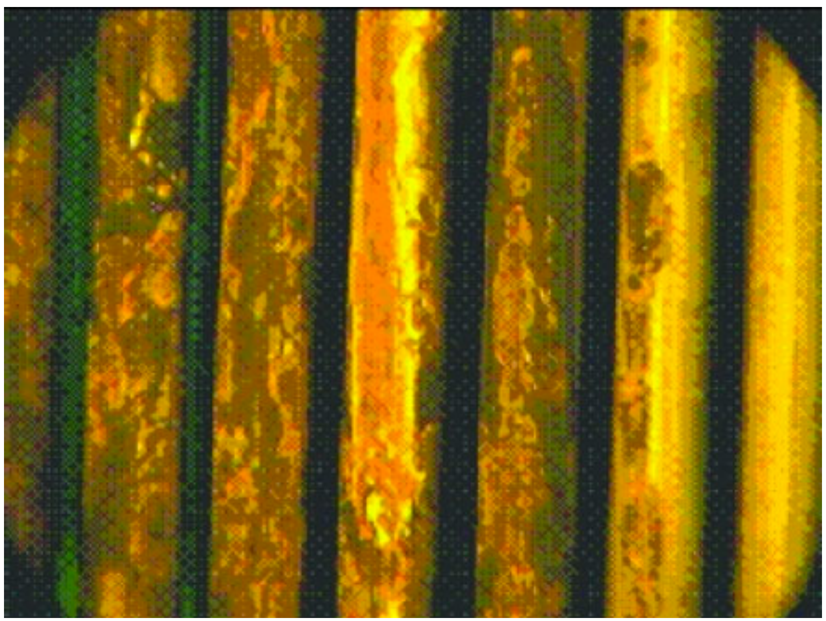
- Why does a hydride rim form in the cladding?
  a) Because of the slow diffusion of hydrogen
  b) Because hydrogen diffuses towards cooler areas
  c) Because the amount of hydrogen that can be in solid solution decreases with temperature
  d) Because the amount of hydrogen that can be in solid solution increases with temperature
- Why do hydrides form circumferentially sometimes, and radially others?
  a) Due to break away oxidation
  b) Radial hydrides form under a tensile hoop stress
  c) Circumferential hydrides form under a tensile hoop stress
  d) Hydride orientation depends on the type of zircaloy

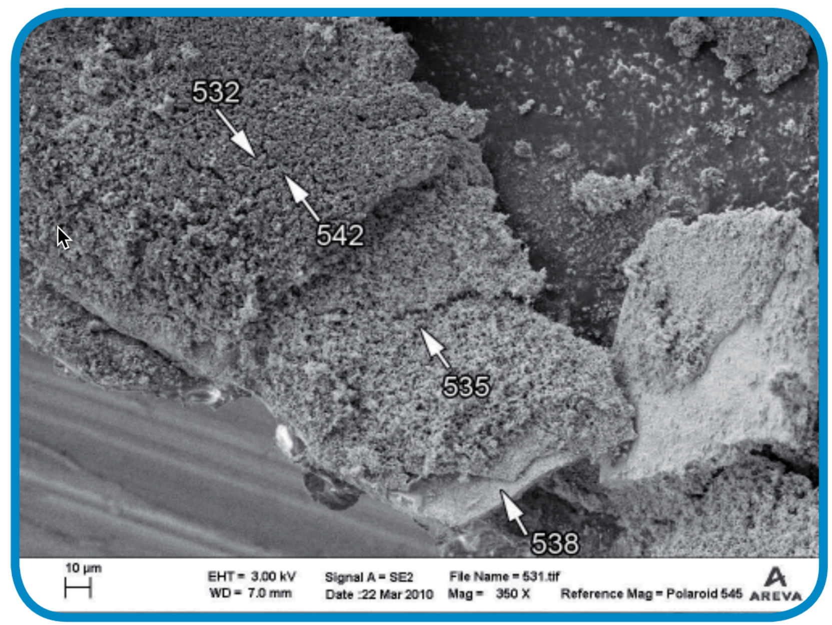
CRUD was first identified at Chalk River Laboratory in Canada

- In one of their first reactors, the scientists at Chalk River (now the Canadian Nuclear Laboratory) found their coolant system was clogged with a highly radioactive black substance.
- They dubbed it CRUD: Chalk River Unidentified Deposits
- CRUD is a large issue still today for high burnup fuel both in BWRs and PWRs

Fuel CRUD is a deposit that forms on the outside of the cladding, on top of the zirconium oxide layer

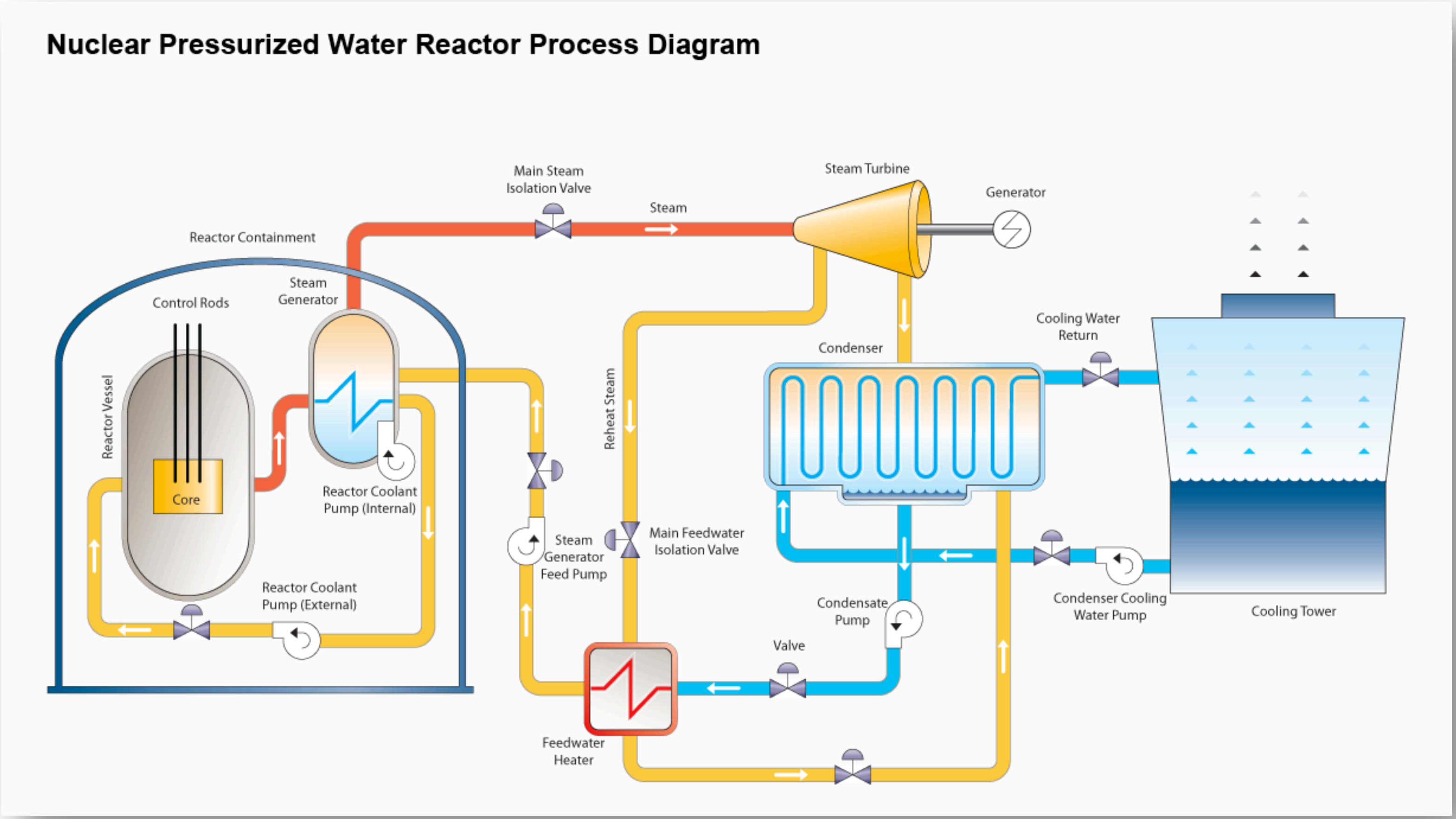
- CRUD is primarily composed of iron oxide, and a collection of other metal oxides.





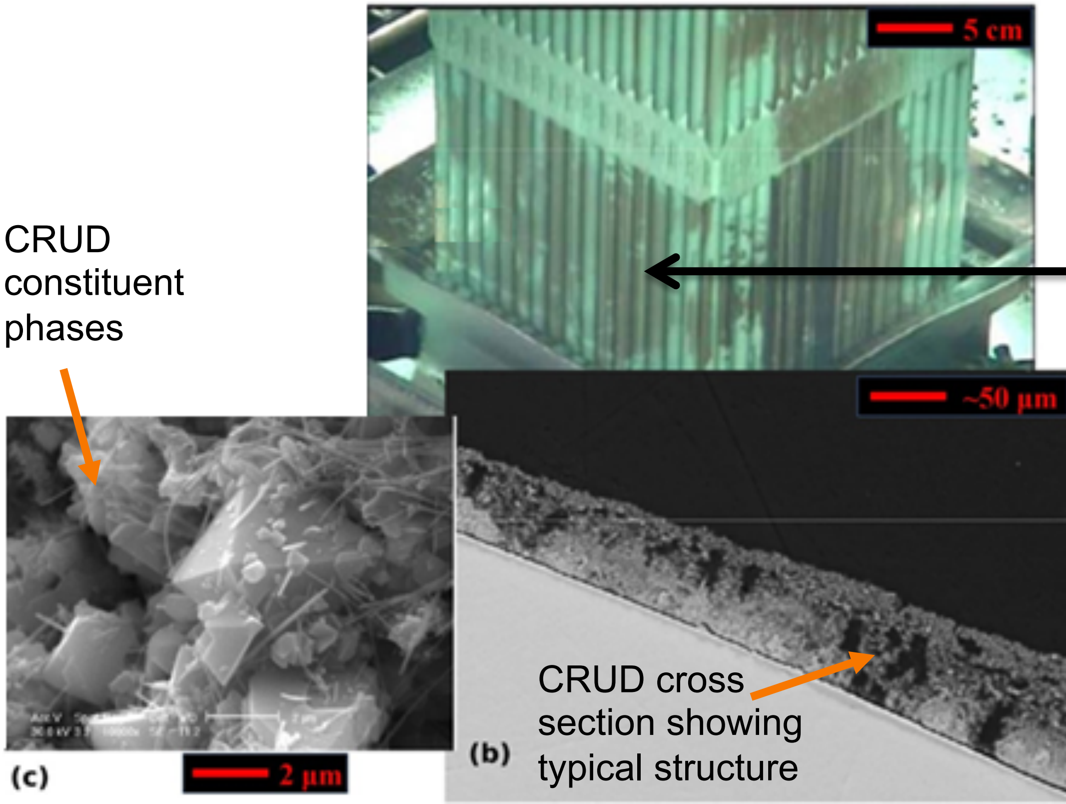
Where does the metal come from that are the primary parts of the oxide CRUD deposits?

- The metal comes from other components that the coolant travels through

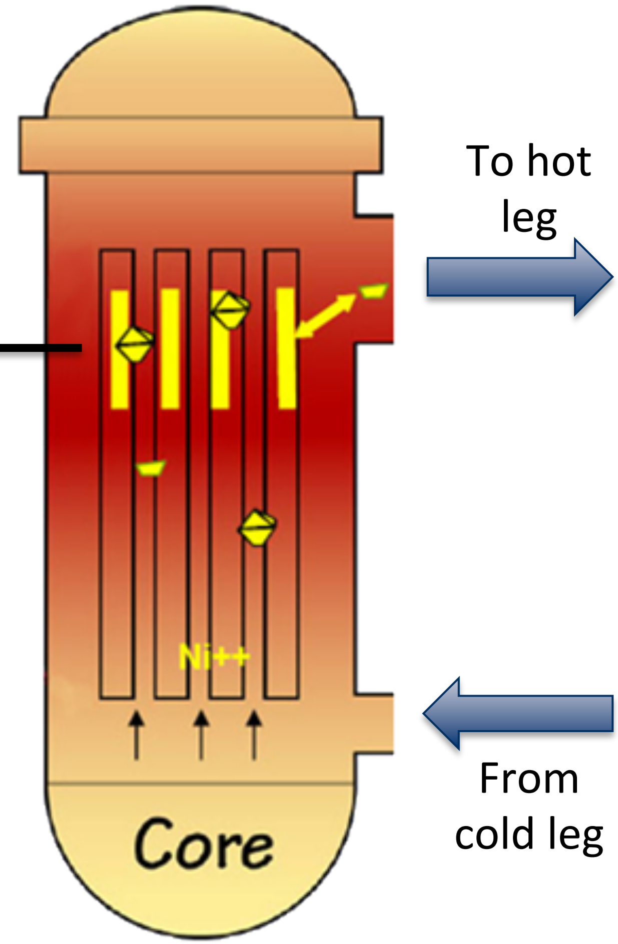


Corrosion products from the coolant deposit on the cladding, forming CRUD

CRUD constituent phases



CRUD cross section showing typical structure



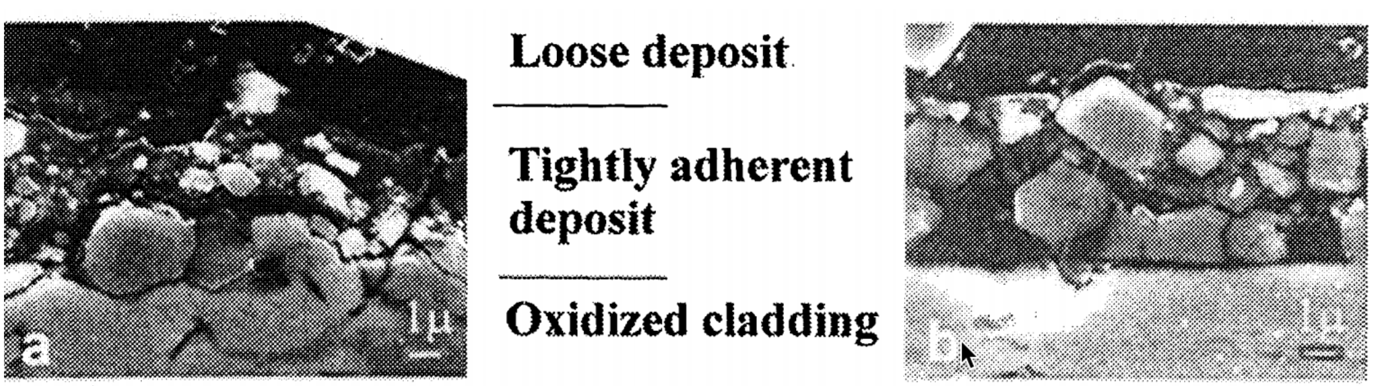
To hot leg

From cold leg

Short, M. P.,et al. *MRS-B* 39(1):71-77 (2014)

Fuel CRUD in BWRs has a red or black appearance and primarily comes from the feed water system

- The CRUD microstructure has two sections:
  - Outer loose deposit
  - Inner tighly adherent deposit



	Cr	Mn	Fe	Co	Ni	Cu	Zn
Forsmark 3	2.6%	0.9%	76%	0.6%	19.1%	0.2%	0.5%
Forsmark 2	2.7%	3.4%	72.7 %	0.4%	17.8%	0.1%	2.8%
Ringhals 1	0.9%	1.2%	84.1%	0.7%	12.7%	0.2%	0.3%

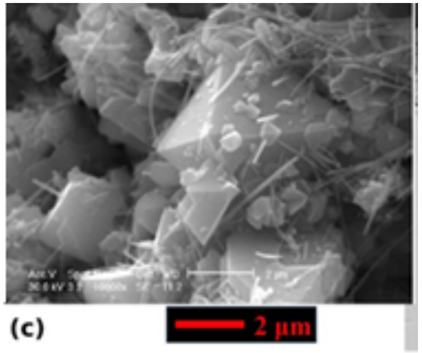
J Chen, SKI Report 00:5, January 2000

Fuel CRUD in PWRs is black with shades of grey.

- It is primarily composed of $NiFe_2O_4$ crystallites
- There are also some NiO and Fe_3O_4

	Cr	Fe	Co	Ni
Bergmann	0.8%	39%	0.11%	19%

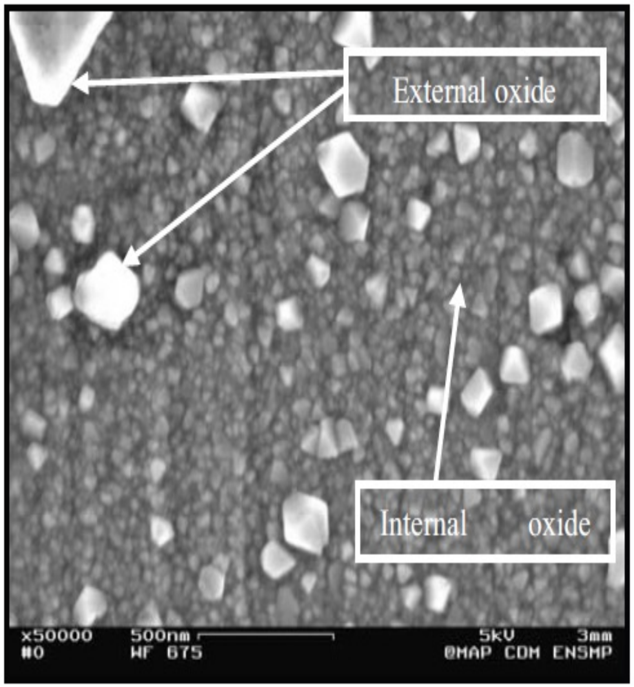
- The corrosion products appear to arrive to the coolant as particulates



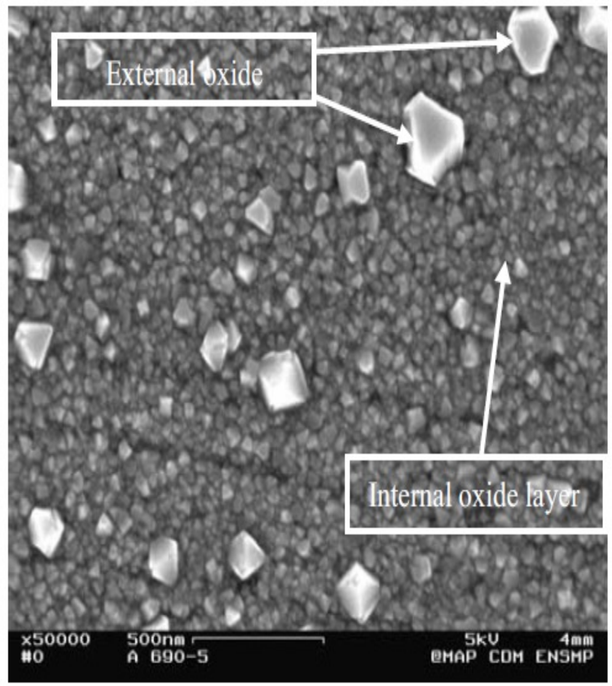
Short, M. P.,et al. *MRS-B* 39(1):71-77 (2014)

J Chen, SKI Report 00:5, January 2000

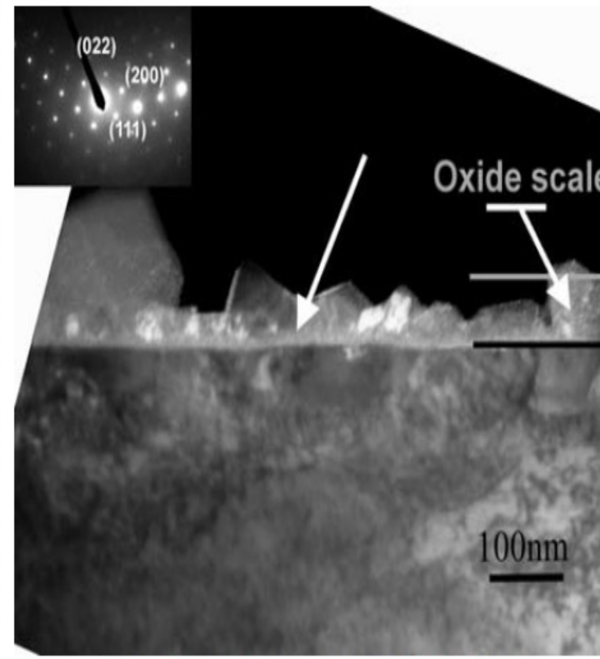
All reactor internals create $NiFe_2O_4$ outer oxide crystals, which are then carried to the cladding



Alloy 600 corrosion



Alloy 690 corrosion



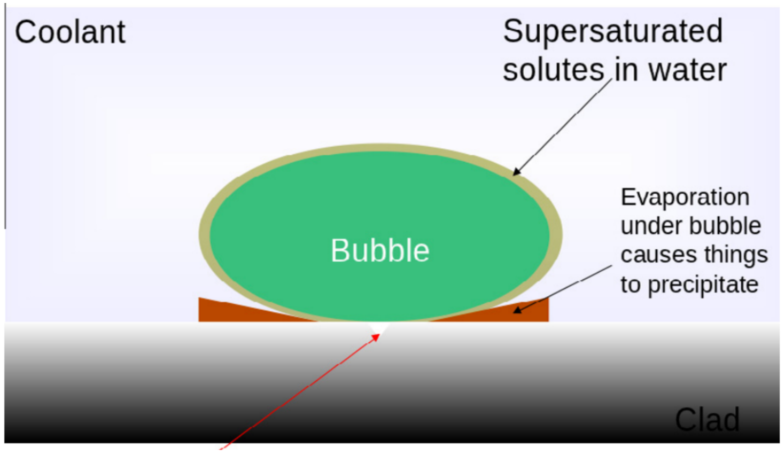
The white arrows point to the Cr_2O_3 layer

Alloy 600 oxide film

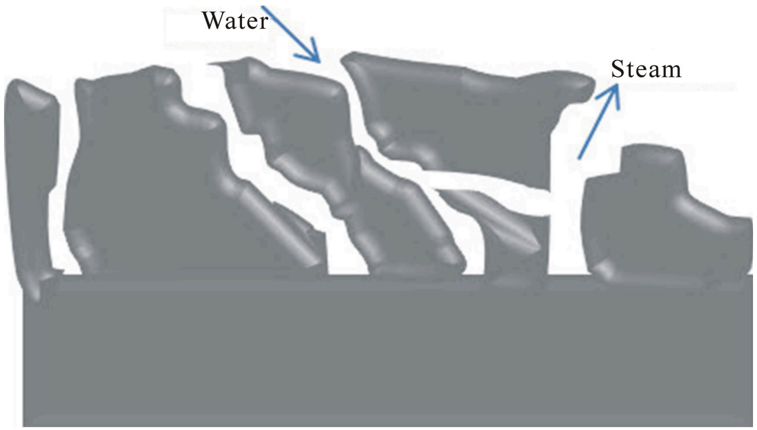
P. Combrade et al. "Oxidation Of Ni Base Alloys In PWR Water: Oxide Layers And Associated Damage To The Base Metal." Proceedings of the 12th International Conference on Environmental Degradation of Materials in Nuclear Power System – Water Reactors – *Edited by T.R. Allen, P.J. King, and L. Nelson* ***TMS (The Minerals, Metals & Materials Society), 2005***

CRUD forms due to small regions of coolant boiling

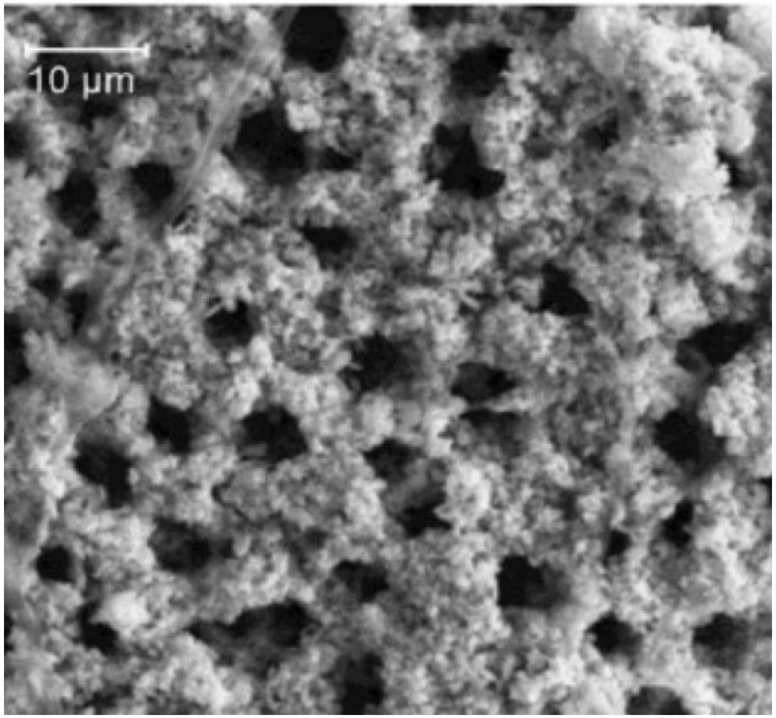
- Boiling causes the precipitation of impurities carried by the coolant

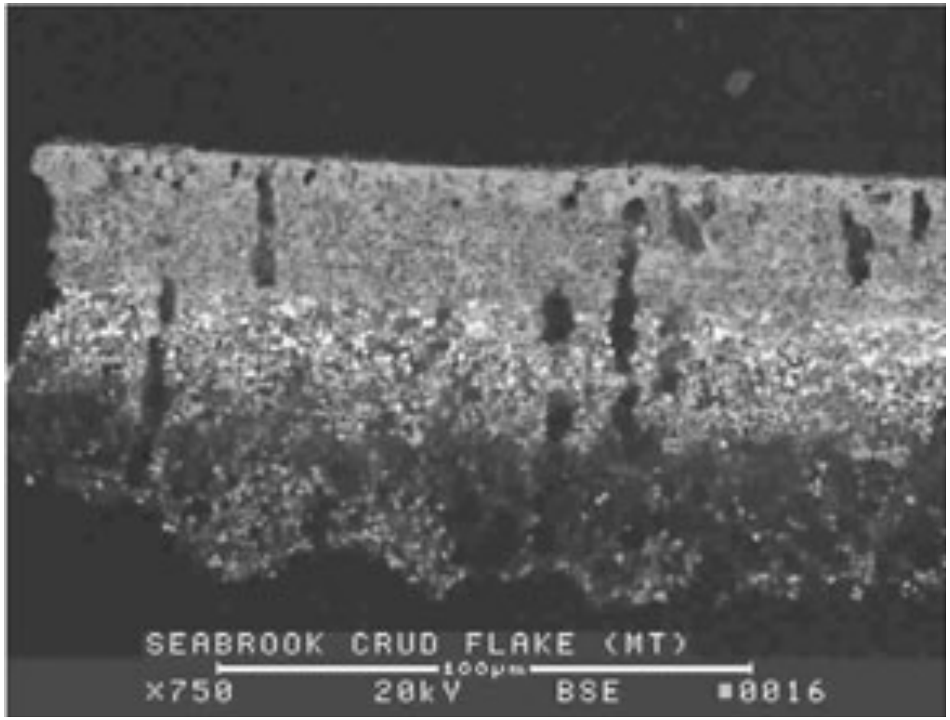


- Cladding forms network of boiling chimneys



The boiling chimneys are clearly evident in the structure of the CRUD





The IHTFP loop at MIT creates CRUD in the lab

1 Pressure Control

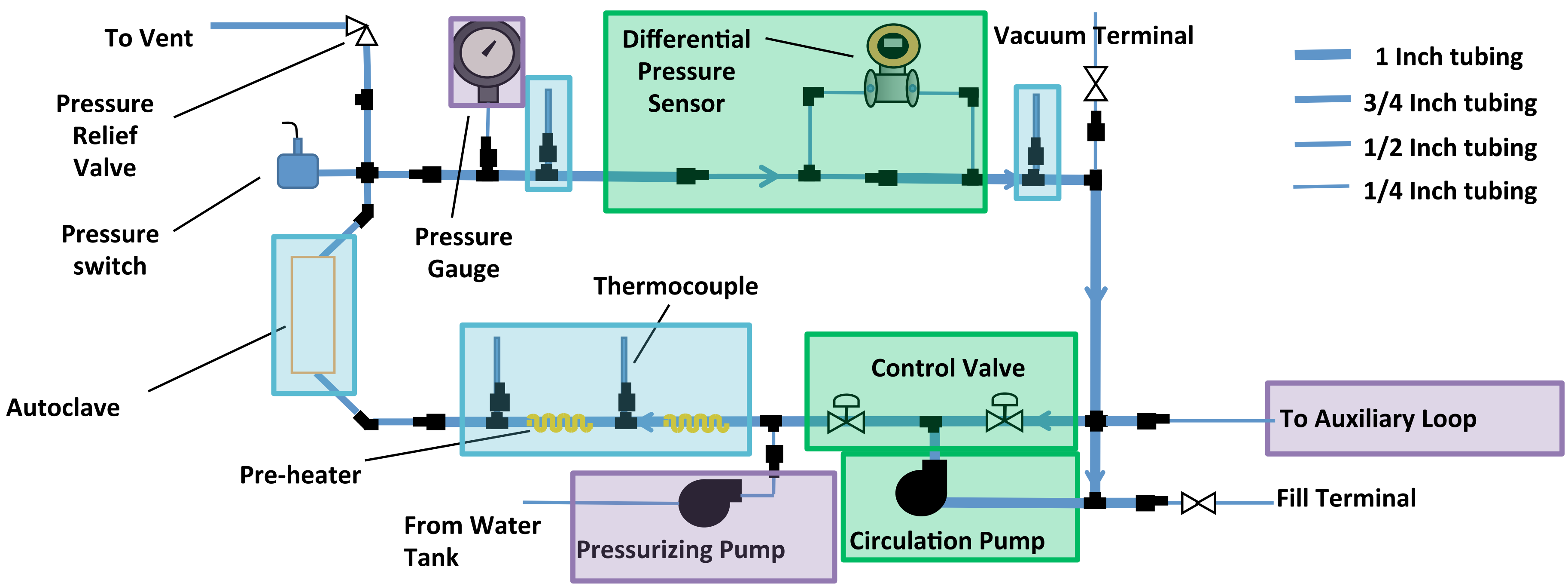
- Increased by pressurizing pump
- Measured by pressure transmitter and gauges
- Set by back-pressure regulator

2 Temperature Control

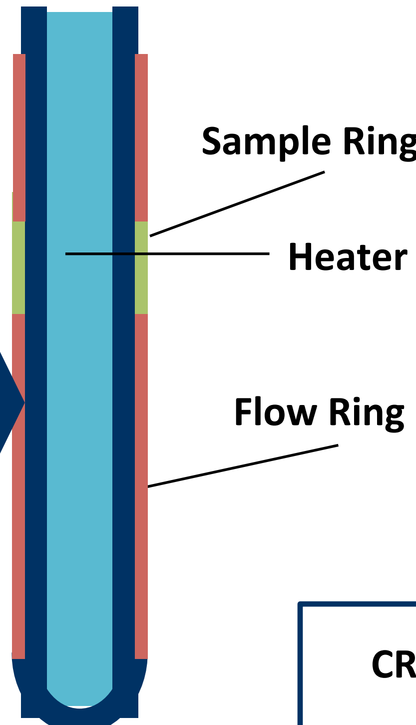
- Increased by autoclave heating rod and pre-heater
- Measured by thermocouples in autoclave and on the loop

3 Flow Control

- Increased by circulation pump
- Measured by differential pressure transmitter
- Moderated by control valve



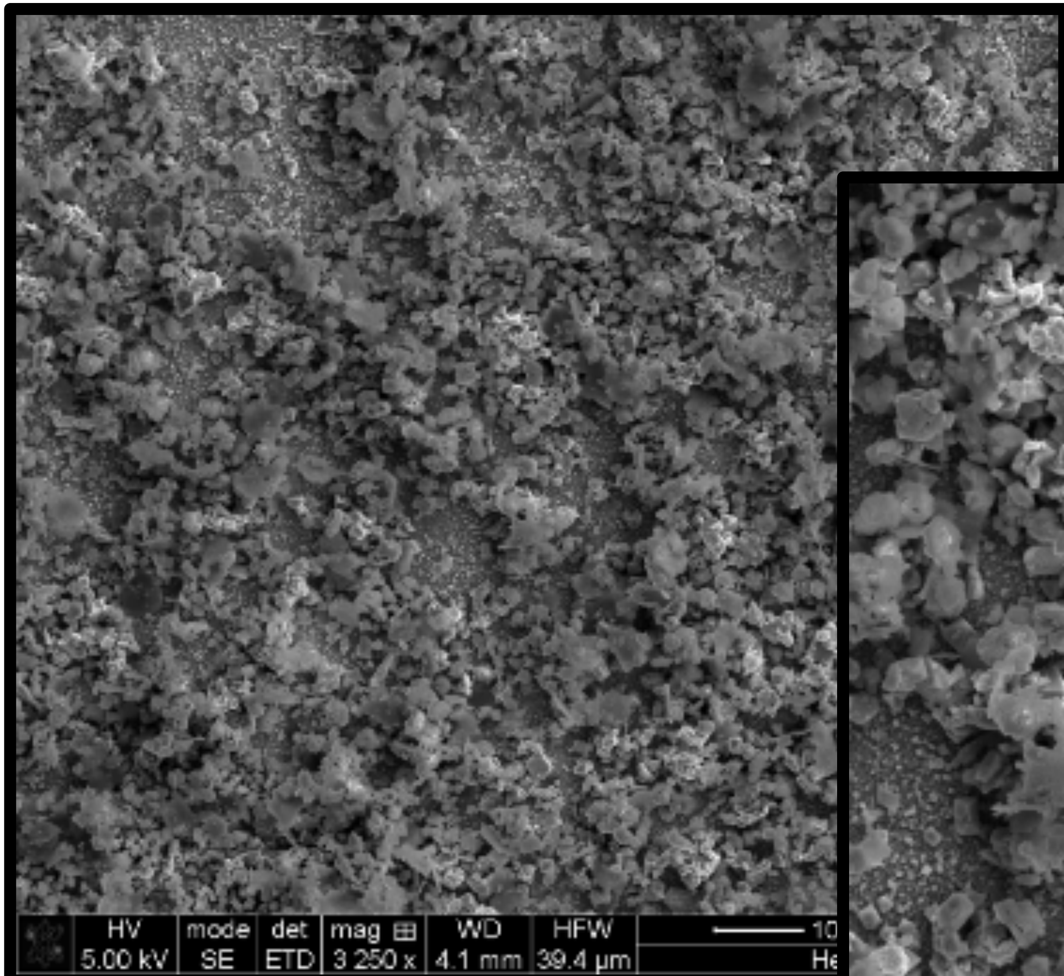
First CRUD Growth after One Week



CRUD grown:

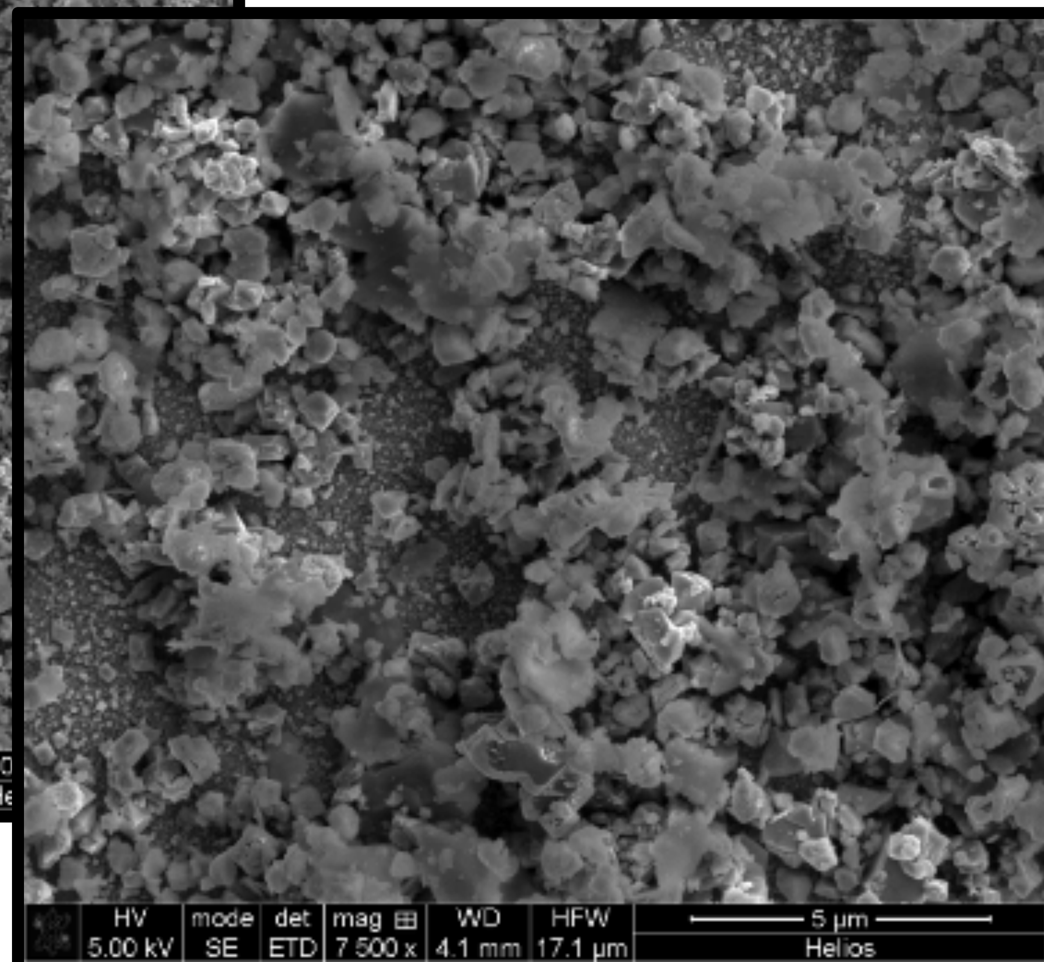
- Heater Temperature: 470 Celsius
- Water Temperature: 320 Celsius
- Pressure:155 bars
- 7 days

First CRUD Sample: SEM Images

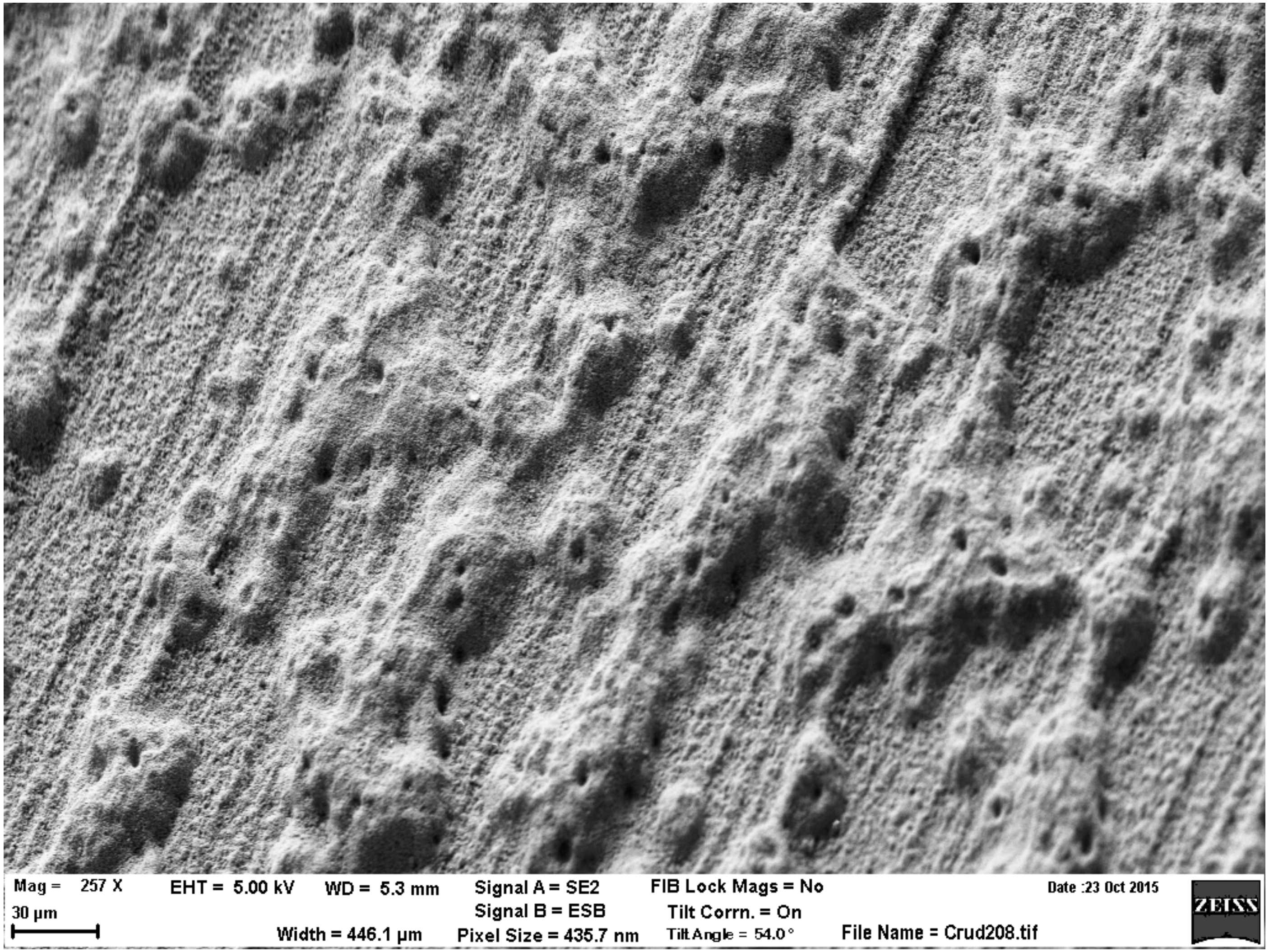


Magnification x3,250

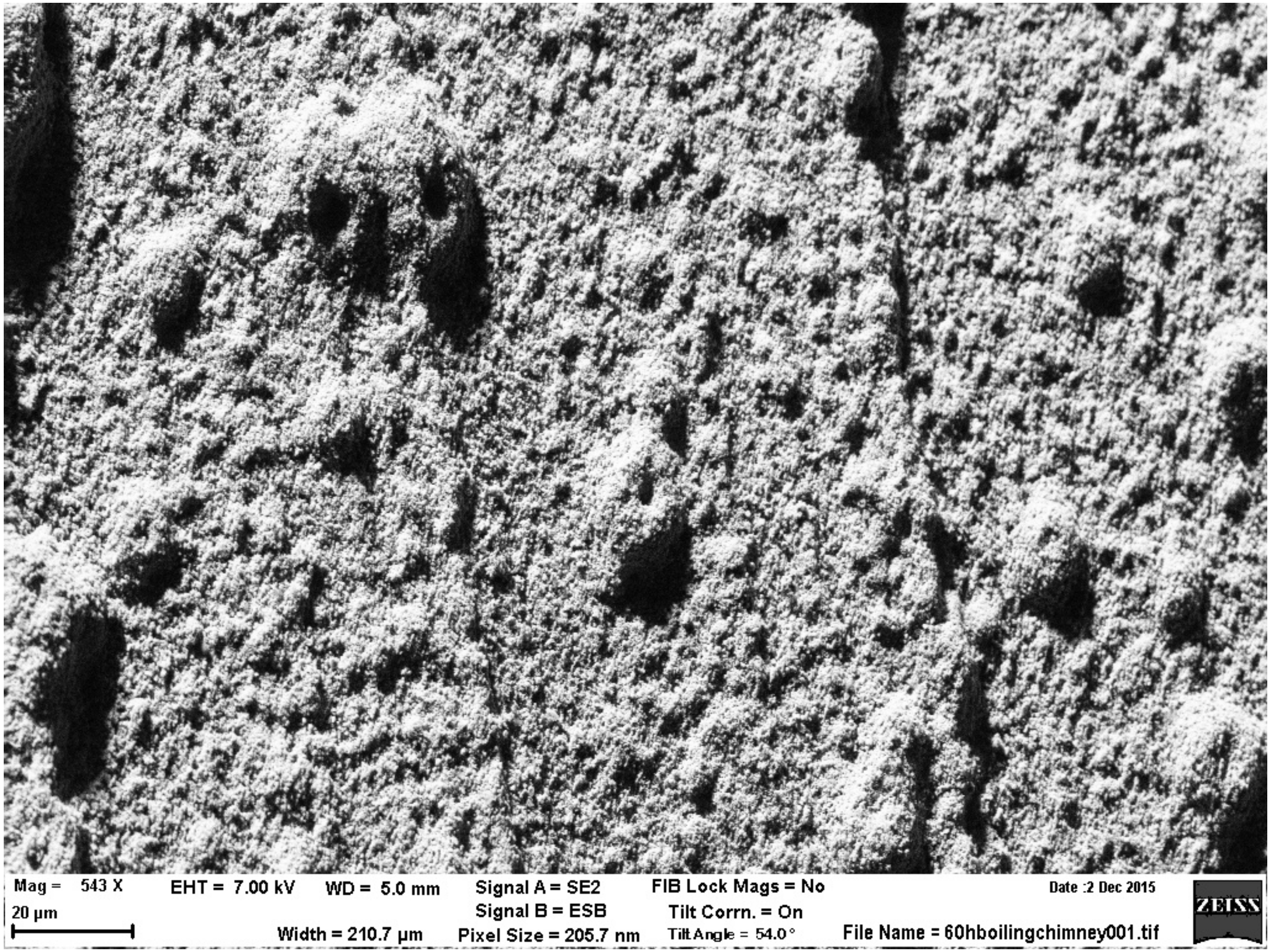
Magnification x7,500



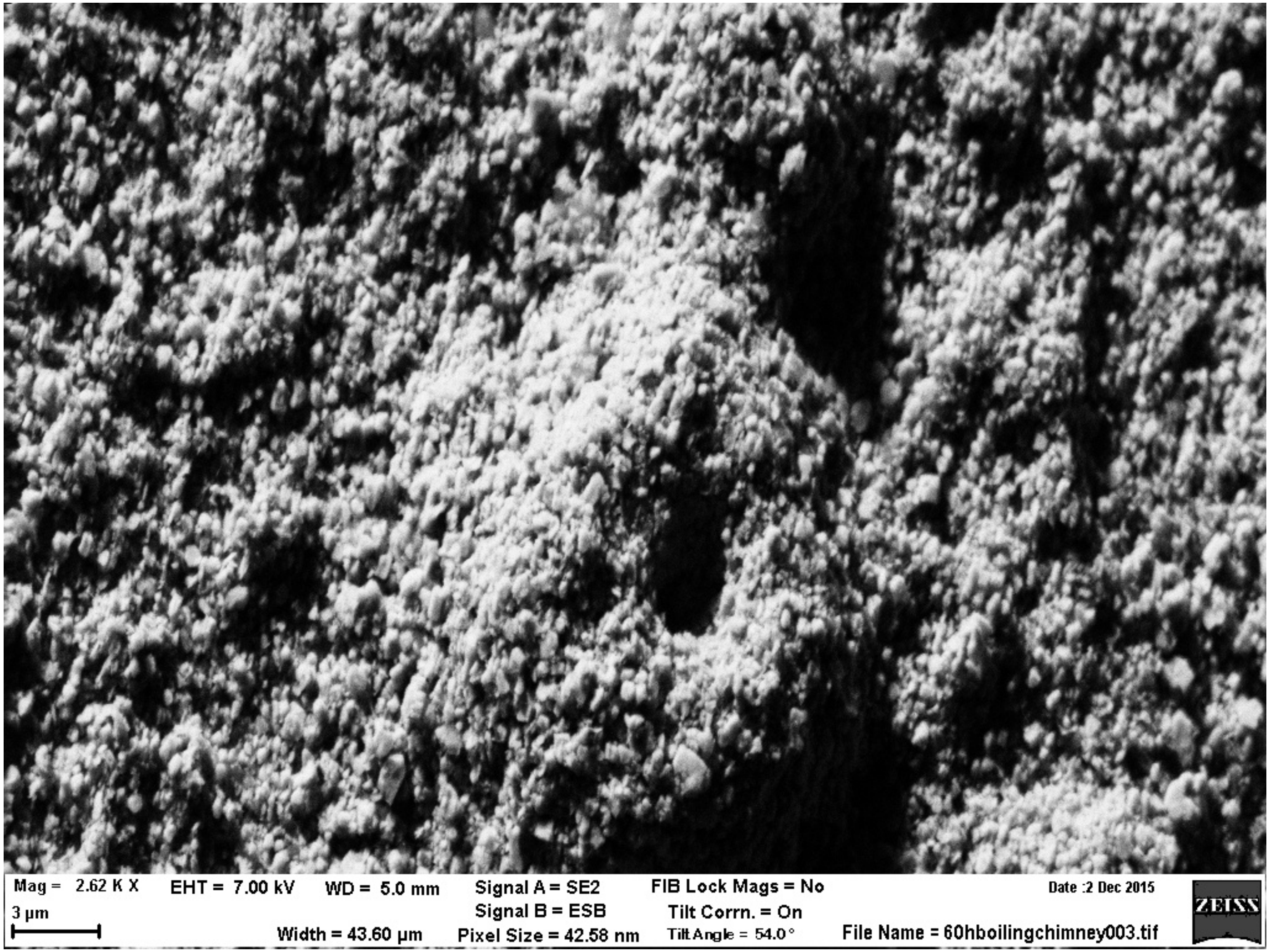
One Week of CRUD Growth



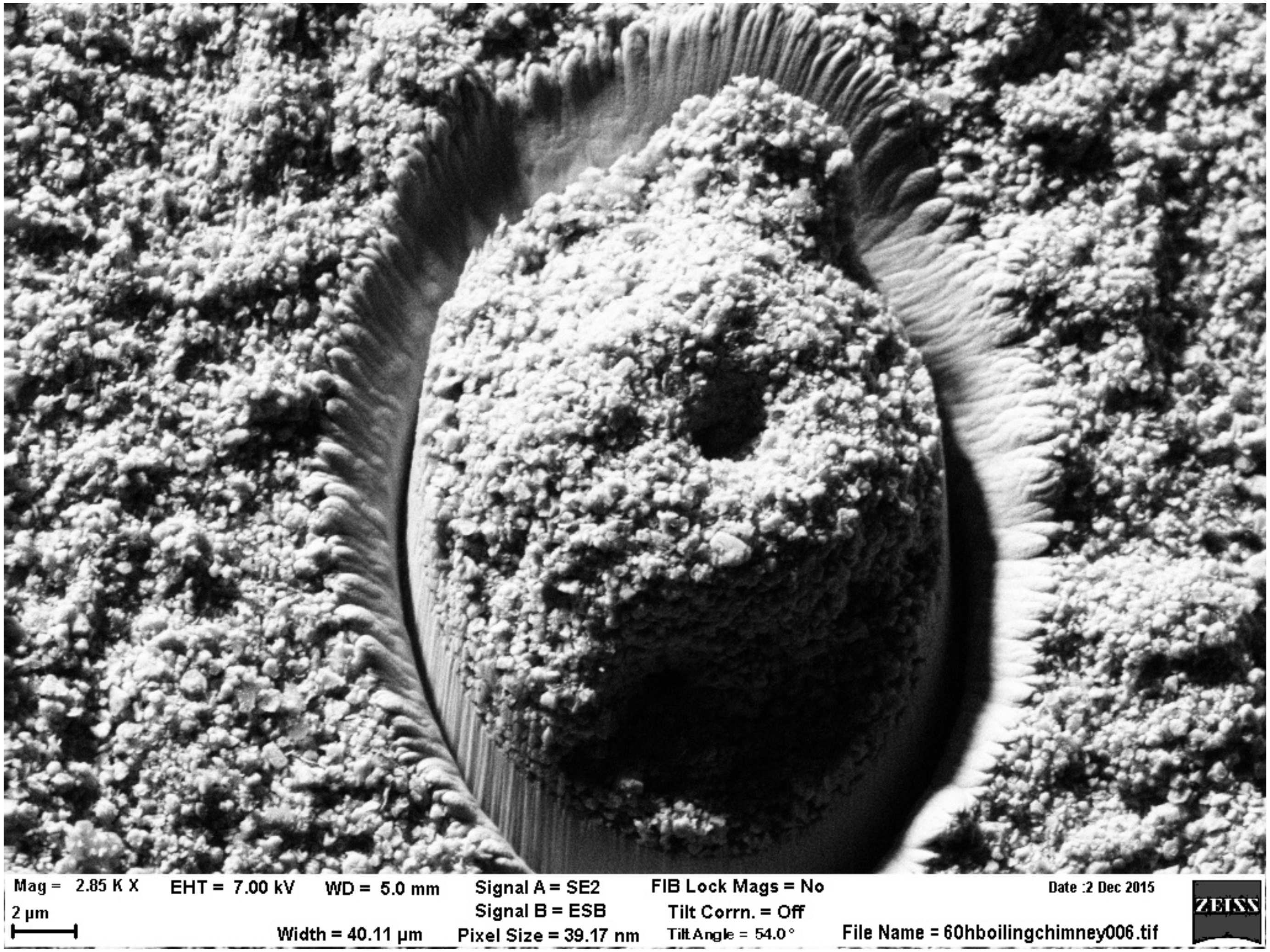
Isolating Boiling Chimneys



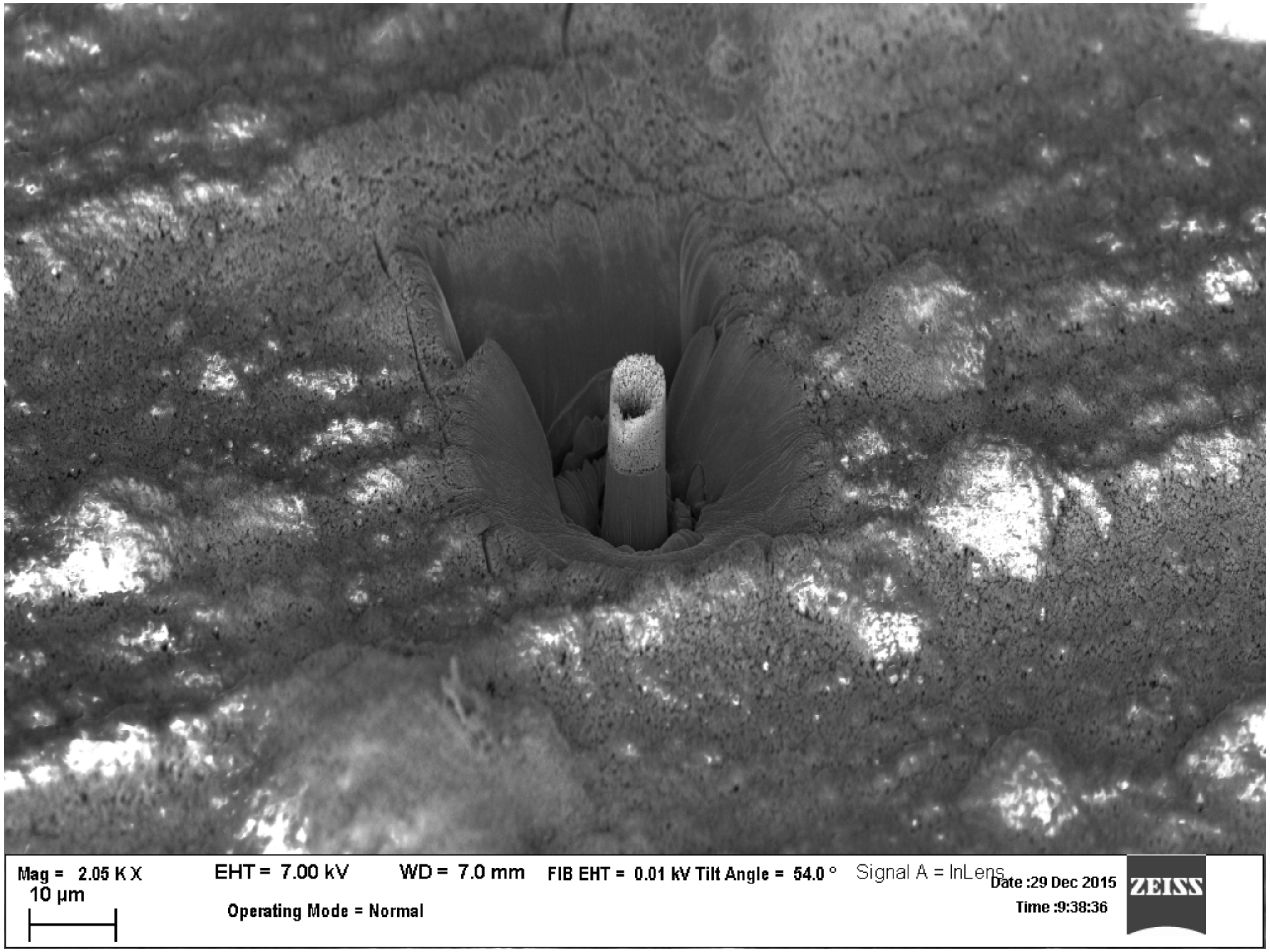
Isolating Boiling Chimneys



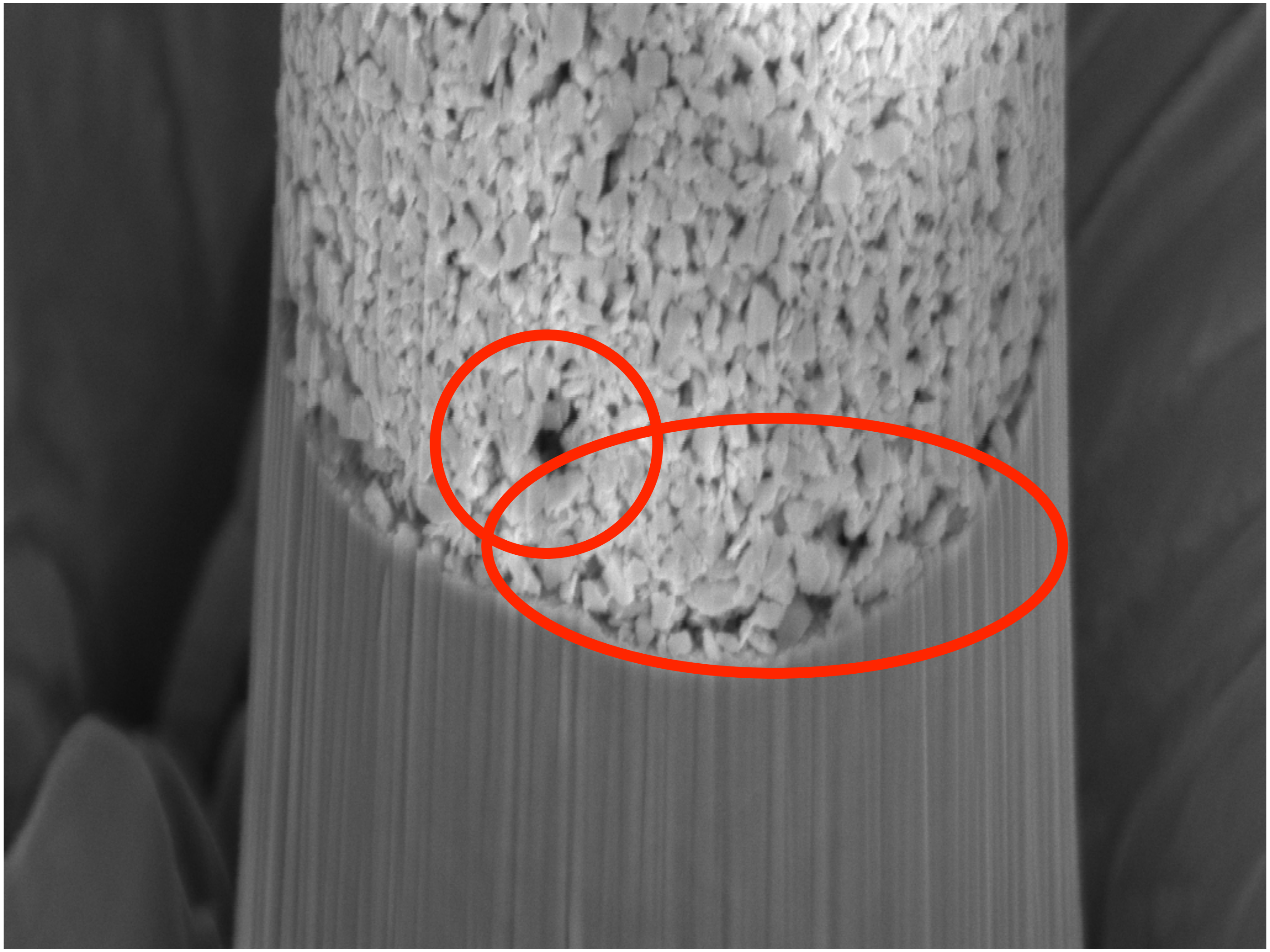
Isolating Boiling Chimneys



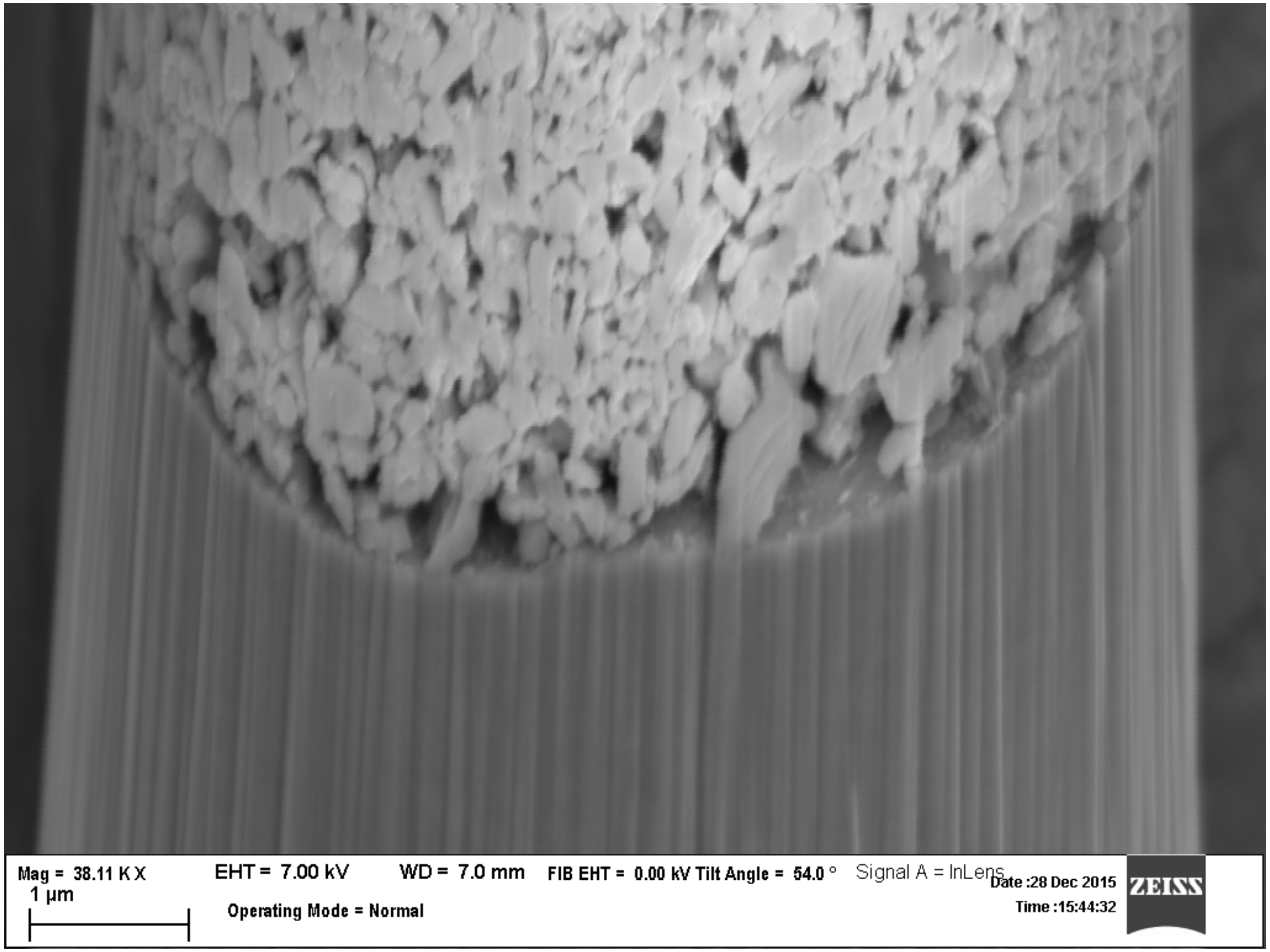
Going Deep into Boiling Chimneys



Going Deep into Boiling Chimneys



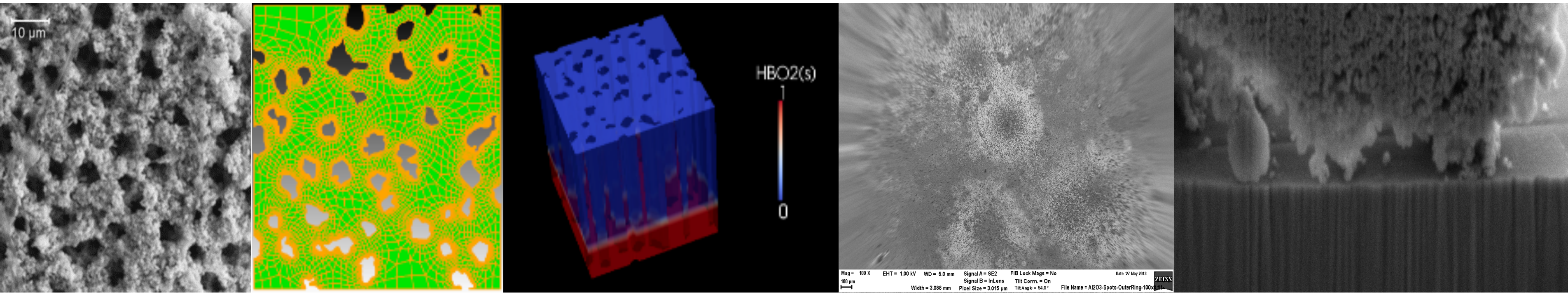
Going Deep into Boiling Chimneys



There are four negative consequences of CRUD

- Reduced heat transport
- Increased radioactivity of the fuel rods
- CRUD induced power shift (CIPS)
- CRUD induced localized corrosion (CILC)

Just like the ZrO_2 layer, the CRUD reduces heat transport through the cladding



- What would be the impact of this reduced heat transport on the fuel performance?

CRUD can be highly radioactive

- The corrosive products that are in the coolant and eventually form CRUD can be activated to form radionuclides.
- Thus, the CRUD becomes highly radioactive, which can complicate matters during reactor servicing.

Table 2.3 Radioisotopes commonly found in PWR and BWR fuel crud*.

Radio-isotope	Half-life (days)	GE reactors BWR ($\mu Ci/cm^2$) [16]	Point Beach 1 PWR ($\mu Ci/cm^2$) [16]
^{51}Cr	27.7	-	0.9-16
^{59}Fe	44.6	-	0.1-2.0
^{58}Co	70.8	40-50	0.7-13
^{65}Zn	244	-	-
^{54}Mn	312	15-90	0.1-2.0
^{60}Co	1924	110-180	0.1-2.0

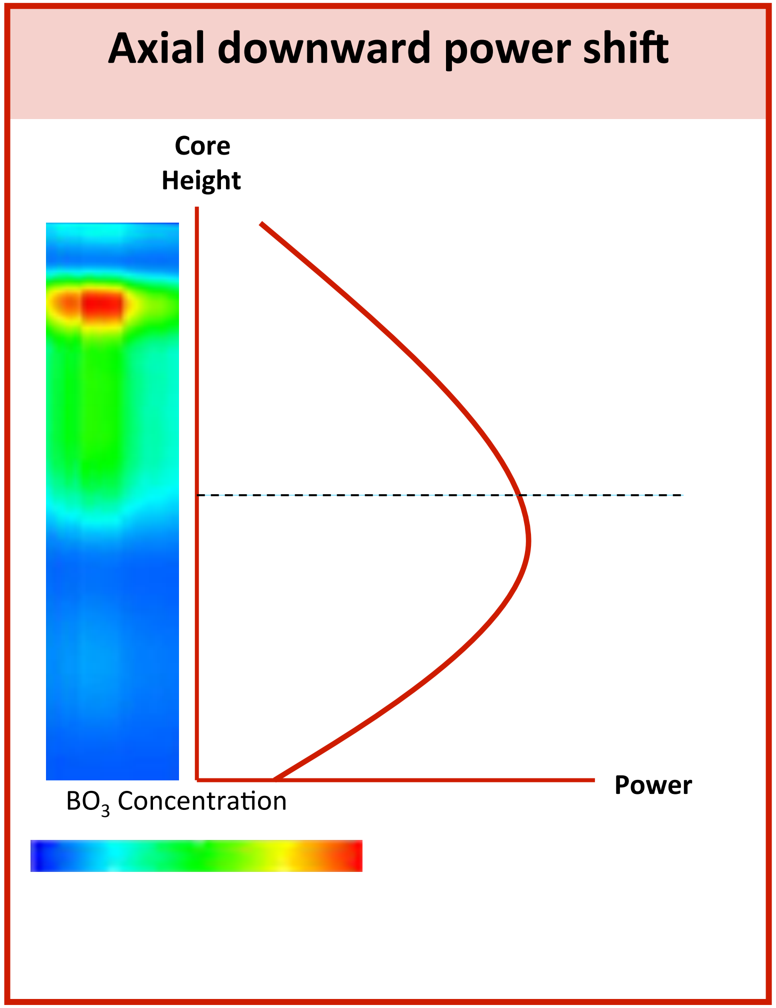
*The data is corrected to the reactor shutdown time.

J Chen, SKI Report 00:5, January 2000

What else is dissolved in the coolant that would enter the CRUD after boiling?

- What would be the impact of boron surrounding the cladding?

CRUD Induced Power Shift (CIPS) results in an axial shift of the power profile



In certain conditions, CRUD and result in CRUD-induced localized corrosion (CILC)

- In 1979 and the early 80's, localized fuel cladding corrosion failures occurred in some plants that had
  - Copper alloy (Admiralty brass) condenser tubes and filter-demineralizer condensate clean-up systems.
  - Such plants have higher levels of soluble copper in the water
- A few rods failed at burn-ups >15 GWd/MT.
  - Over 90% of the failed rods contained (U,Gd)O2 fuel (i.e. they were "gadolinia rods").
  - Most fuel reloads and fuel bundles were not affected, even in susceptible plants.

The CRUD composition was found to be quite different in these fuel rods

- The CRUD deposits had with high copper concentrations, rather than the typical fluffy Fe_2O_3 crud.

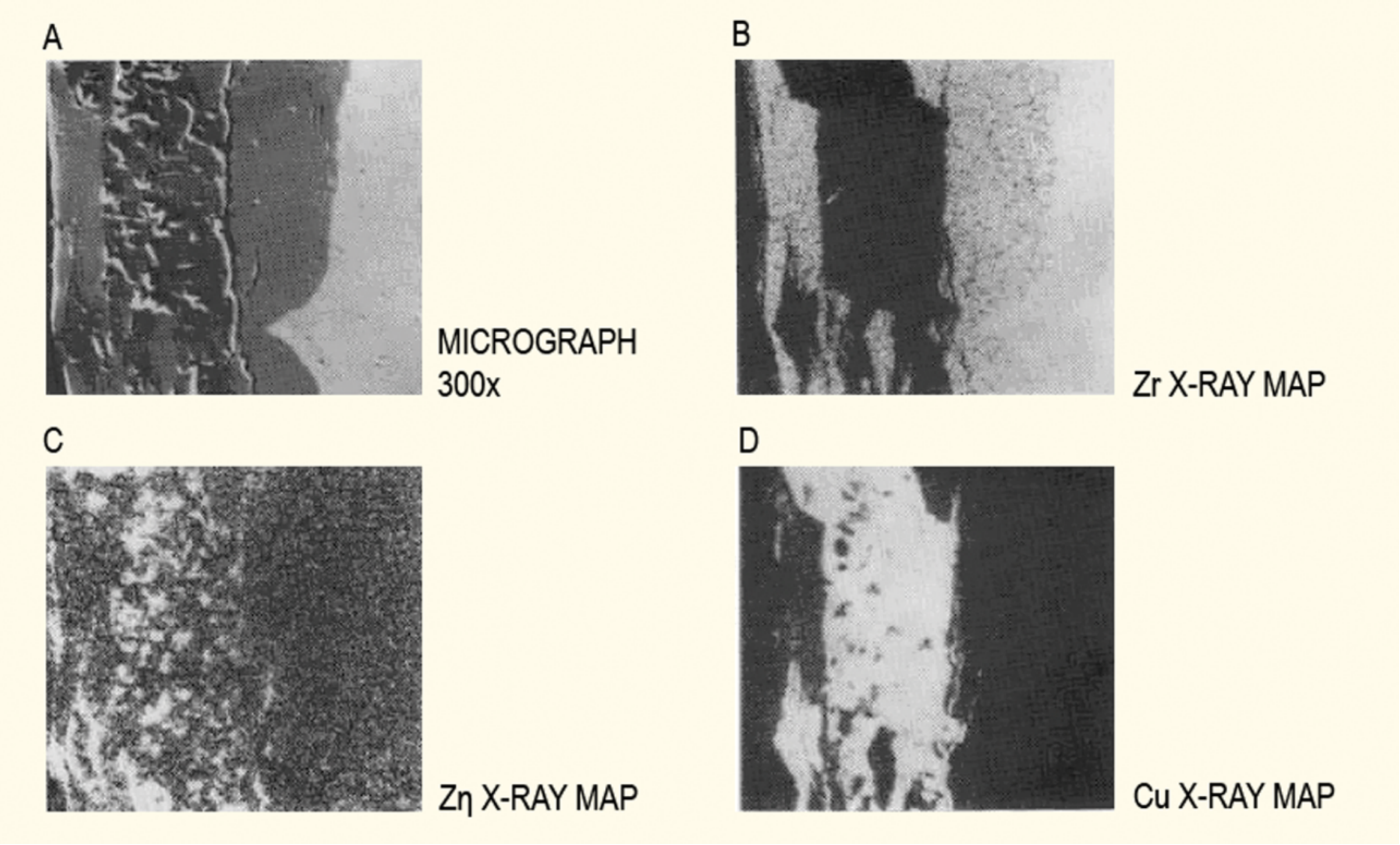
	Standard *CRUD*	*CILC CRUD*
Major Phase	Fe_2O_3	CuO
Iron	87%	21.1%
Copper	2.0	52.8
Zinc	4.4	11.1
Nickel	3.3	2.5
Manganese	2.2	3.3
Chromium	1.1	2.5
Cobalt	0.3	0.6

CILC occurs in two phases

- Incubation Phase (low to moderate power)
  - Extensive nodular oxidation occurs early in first cycle (90 – 100% coverage)
  - Copper deposits between the oxide nodules
  - This forms a thick sandwich structure (ZrO_2/CRUD/ZrO_2)
- Failure Phase (moderate to high power)
  - Cracks form within sandwich structure producing local, steam-insulated regions
  - Insulating effect increases temperature and accelerates cladding corrosion and hydriding
  - Cladding penetrations occur locally by formation of auto-catalytic corrosion pits or by cracking of hydrided Zircaloy in spalled regions

Copper-rich crud deposited in laminations (cracks) of zirconium oxide

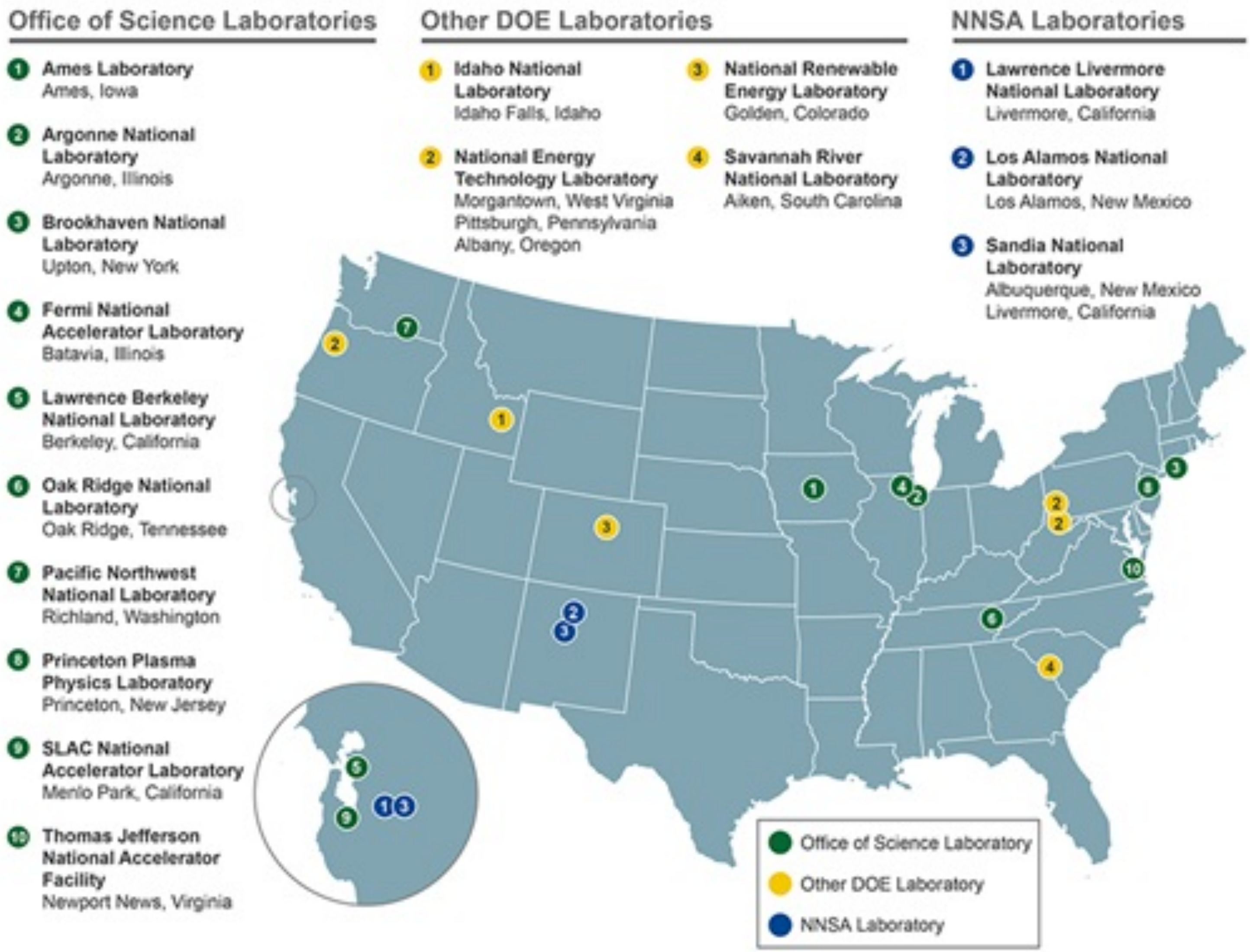
In the X-ray map, a light region indicates the presence of the element in question, Marlowe et al., 1985.



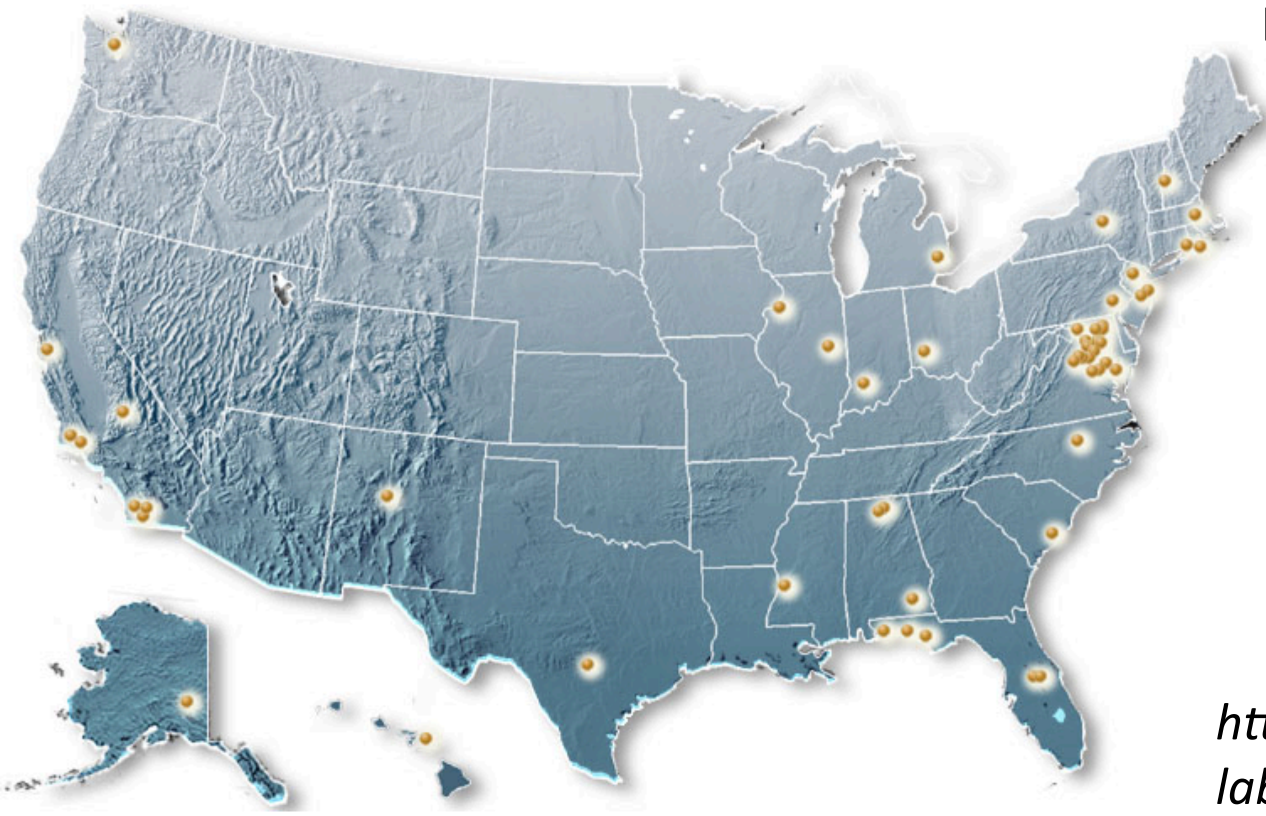
Summary

- CRUD results from particles in the coolant that come from the corrosion of other parts of the reactor coolant system
- It forms a porous layer outside of the Zr oxide layer on the cladding
- Localized boiling creates boiling chimneys that build up the CRUD
- The negative effects are
  - Reduced heat transport
  - Increased radioactivity
  - CIPS
  - CILC

The Department of Energy runs the most National Laboratories



The Department of Defense also runs National Laboratories

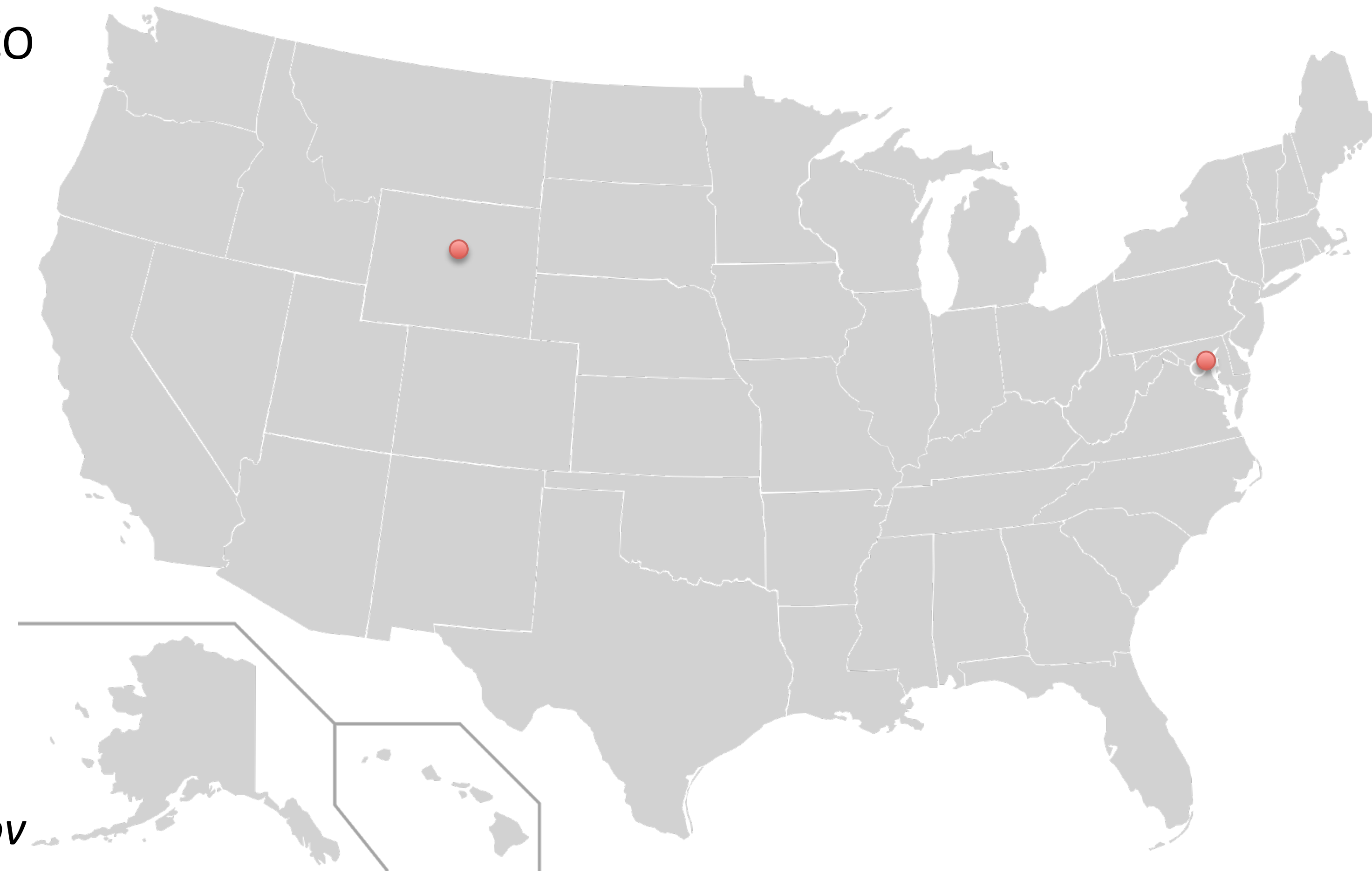


- The Department of Defense National Laboratories include
  - Army Research Lab
  - Navy Research Lab
  - Air Force Research Lab
  - Bettis
  - KAPL
  - Many more…

http://www.acq.osd.mil/rd/laboratories/labs/map.html

The National Institute of Standards and Technology has two main locations

- Gaithersburg, MD
- Boulder, CO



www.nist.gov

National Laboratories are excellent places to work if you want to focus on DOING research

- As research staff, you do your own research.
- You can also mentor postdocs and summer interns
- You may not need to find your own funding, but you have less freedom to choose what to work on
- The overhead costs are very high, limiting who will pay you to do work
  - An employee making $100K/year needs around $370K/year of funding
- Myths (not universally true)
  - You must be a US citizen
  - You will have to work on nuclear weapons
  - They only hire Ph.D.s