High Temperature Glass Seal

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Outline

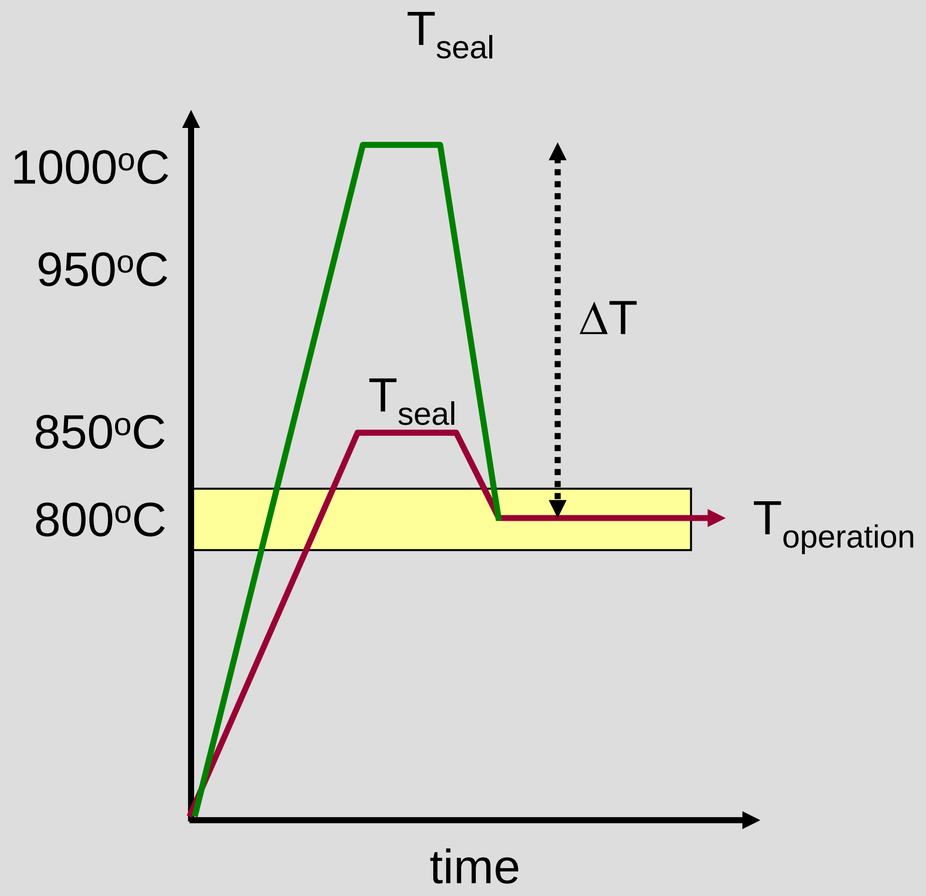
- Accomplishment of FY07
- Mechanical strength evaluation: plain crofer, aluminized, spinel-coated
- Electrical stability evaluation: aging under DC loading
- Summary
- Future work

FY07 Accomplishment

- Completed seal strength evaluation of high-temperature glass. Evaluated pre-oxidation, aging, coating, and environmental effects on strength.
- Without coating, strength would degrade if a thick Cr_2O_3 oxide layer present or aged in air. No strength reduction if aged in reducing gas. Cause for strength degradation was $SrCrO_4$ formation.
- Alumina coating is effective in blocking Cr; however, the deposition process needs to be optimized to minimize overdose.
- Spinel coating showed best results with minimum strength reduction even aging in air.
- Tested conventional and high-temperature sealing glasses in SOFC environment and 0.7 V DC loading. Conventional glass showed severe Fe diffusion and rapid increase in conductivity (830^{o}C/~80hr), while high-temperature glass showed excellent electrical stability over ~1200hr at 850^{o}C.

High-temperature sealing glass

1. Increase contact bonding strength
2. Increase thermal stability
3. decrease interfacial reactions



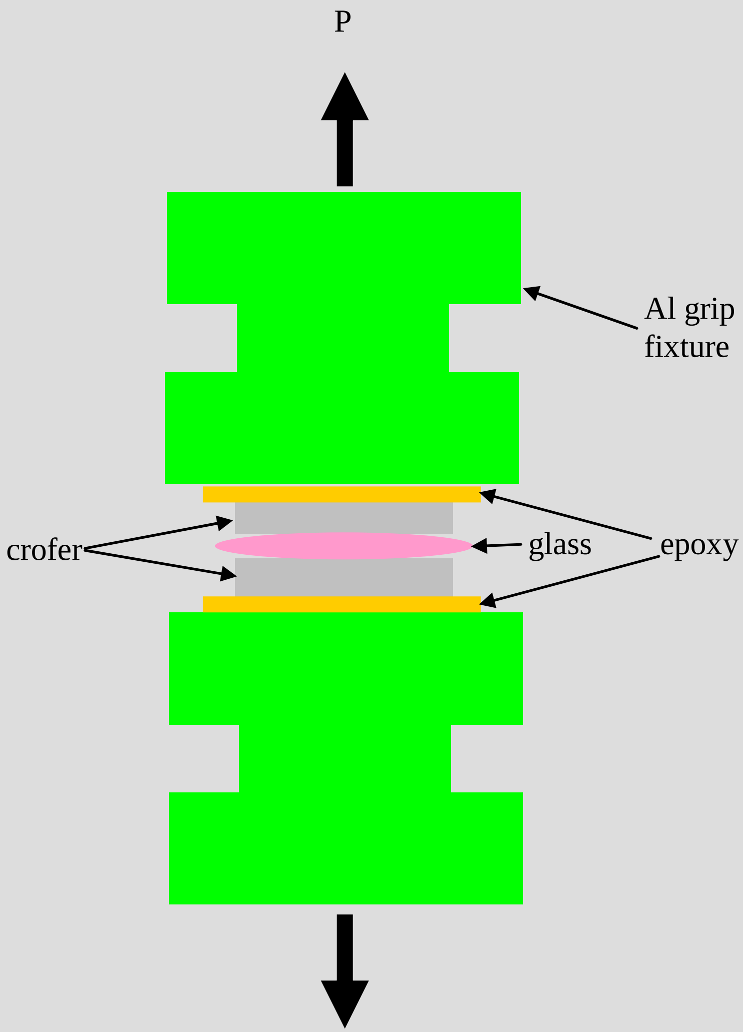
Battelle

Mechanical strength evaluation

1. Effect of pre-oxidation: Cr_2O_3 layer thickness
2. Effect of different protective coating: Al_2O_3, $(Mn,Co)_3O_4$
3. Effect of environment: oxidizing, reducing
4. Accelerated condition:

- 850^{o}C/500h in air
- 850^{o}C/250h in 30%H_2O, 70%(2.7%H_2/Ar)

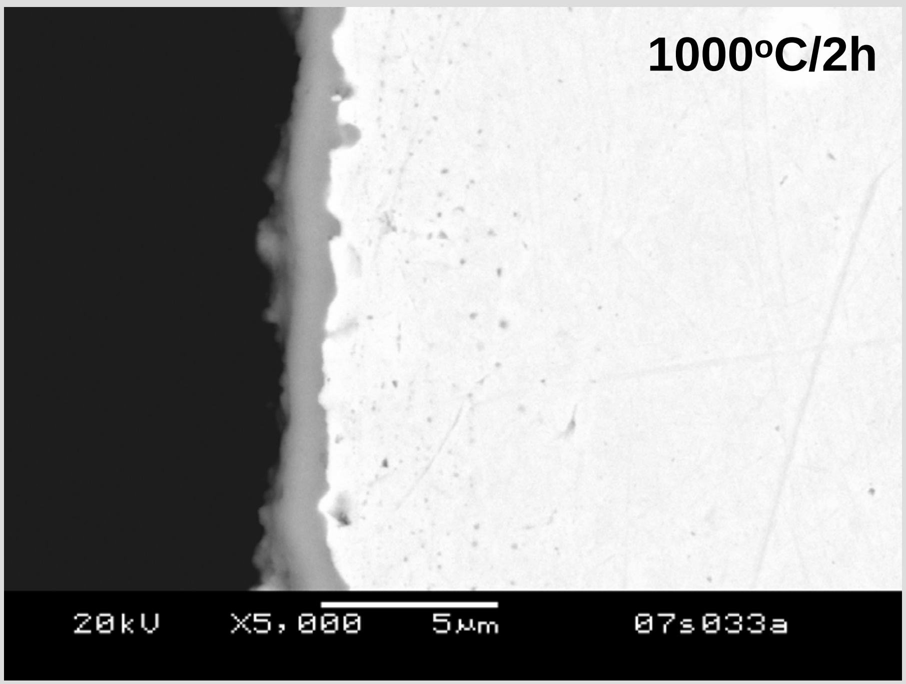
Interfacial tensile testing

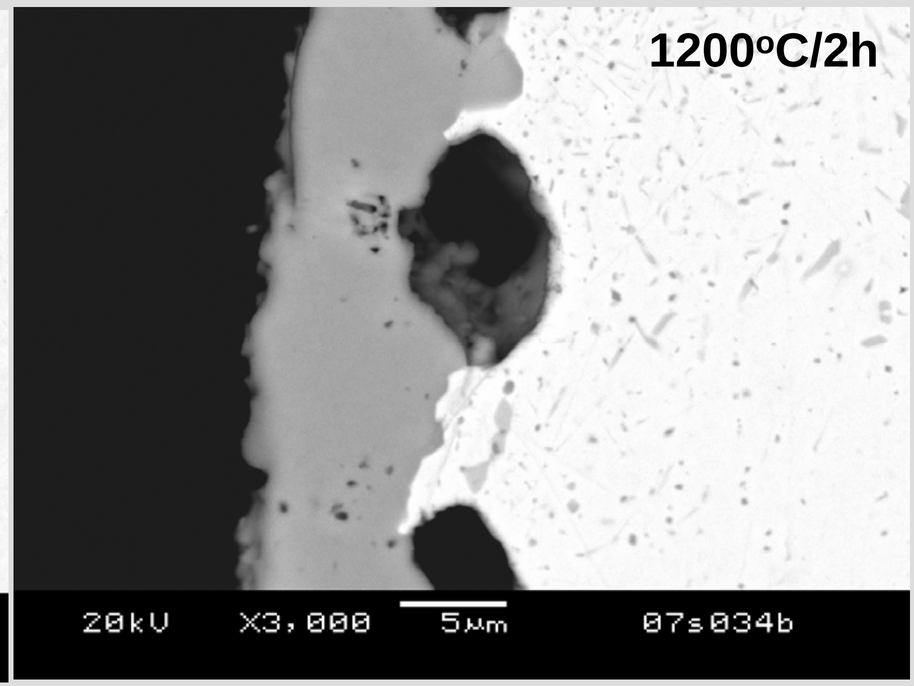


- R. T.
- 0.5 mm/min cross-head speed
- 5-7 samples tested for each condition
- Self-aligned grip fixture
- ½”x½” sample size

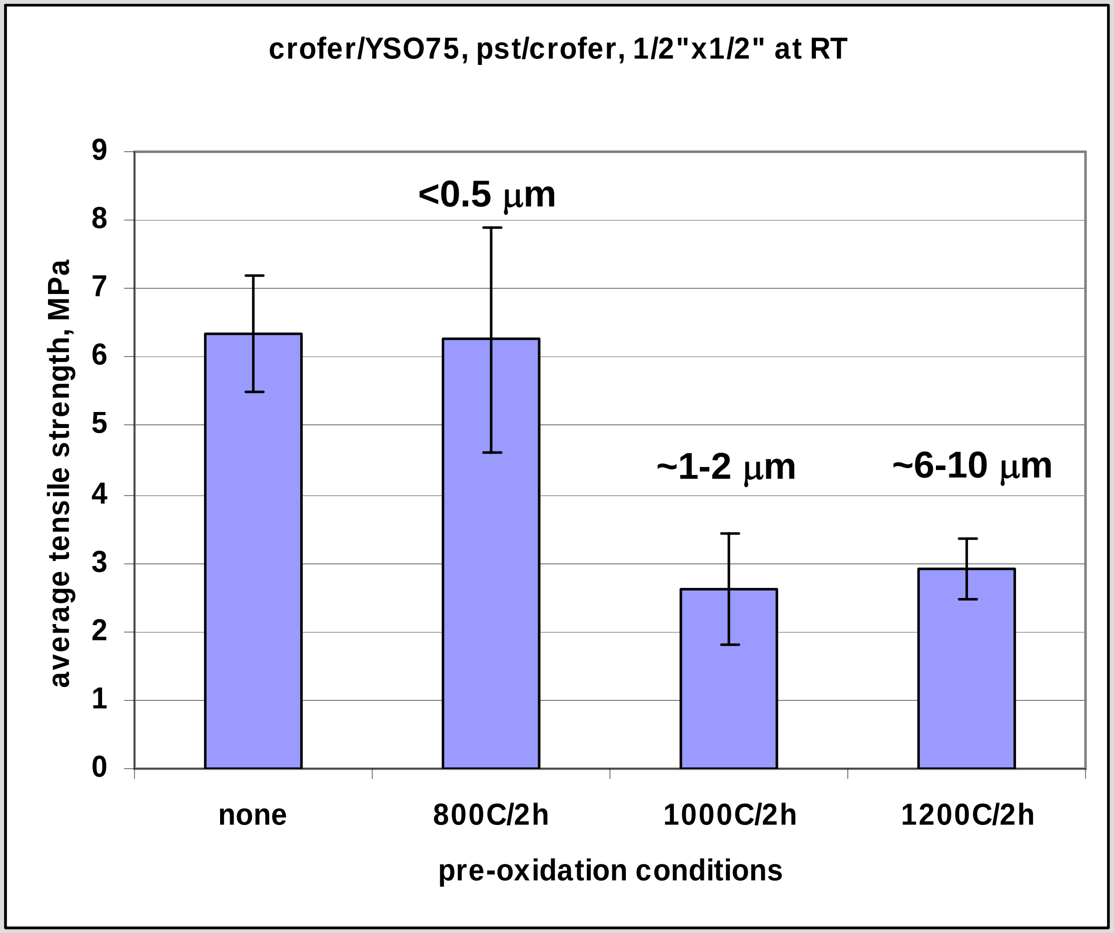
Pre-oxidized crofer

To mimic long-term oxidation
1200°C resulted in pores underneath
Outside discrete and non-uniform (Mn,Cr) oxide, inside dense and continuous Cr_2O_3-rich oxide
T<0.5μm (800°C/2h)





Effect of pre-oxidation on tensile strength



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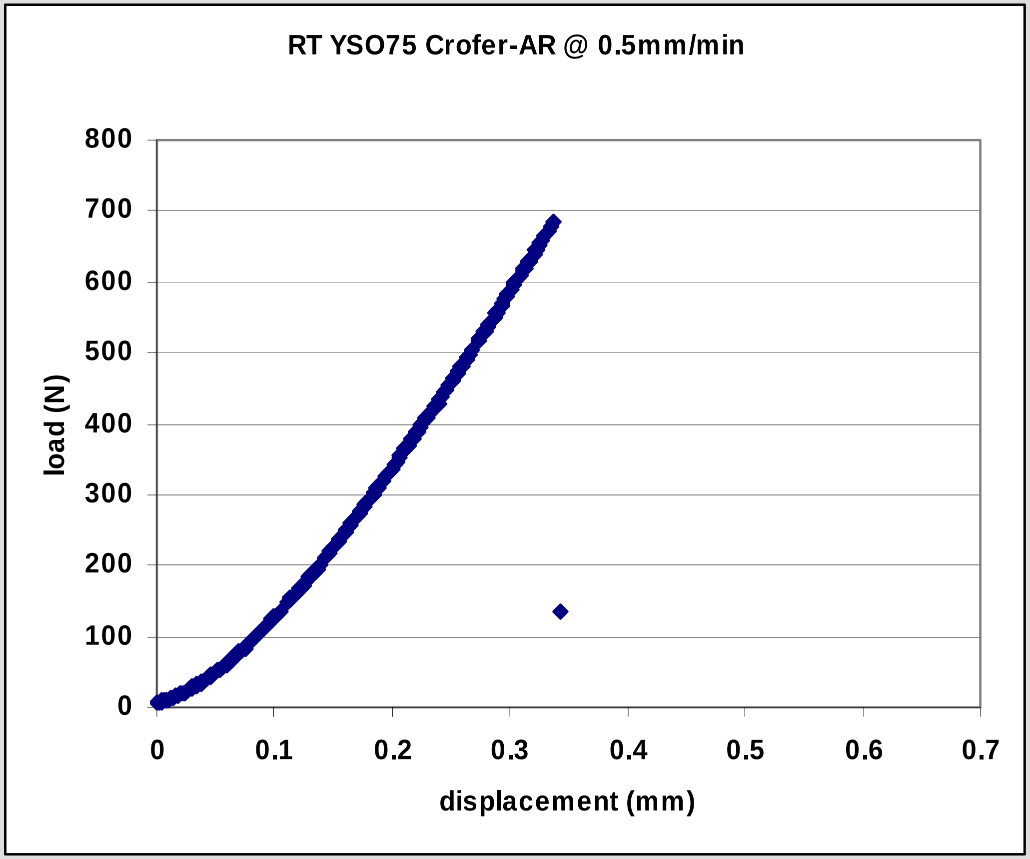
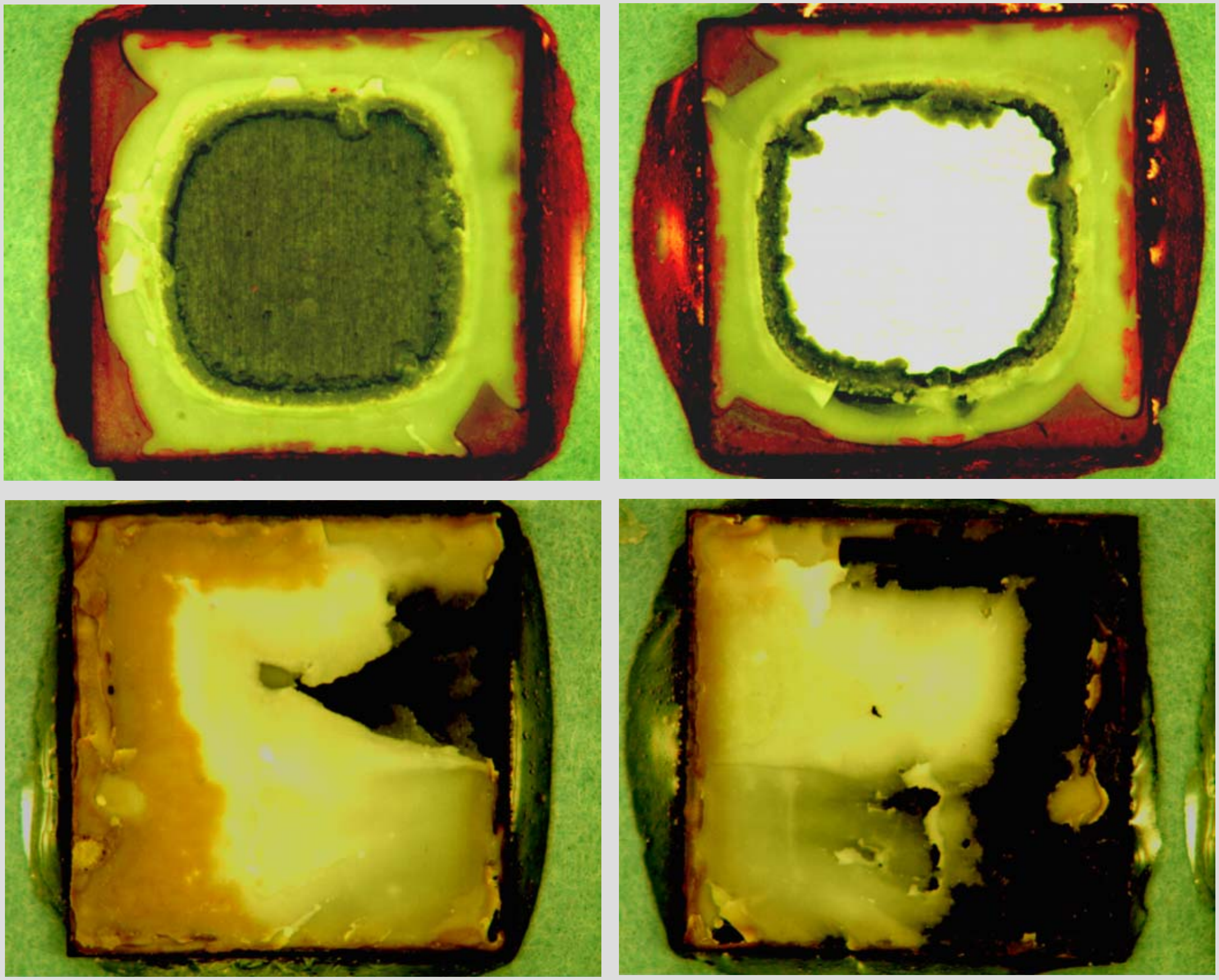
Load-displacement curve showed typical brittle failure

All showed brittle failure: coating, aging, oxide layer thickness, environment



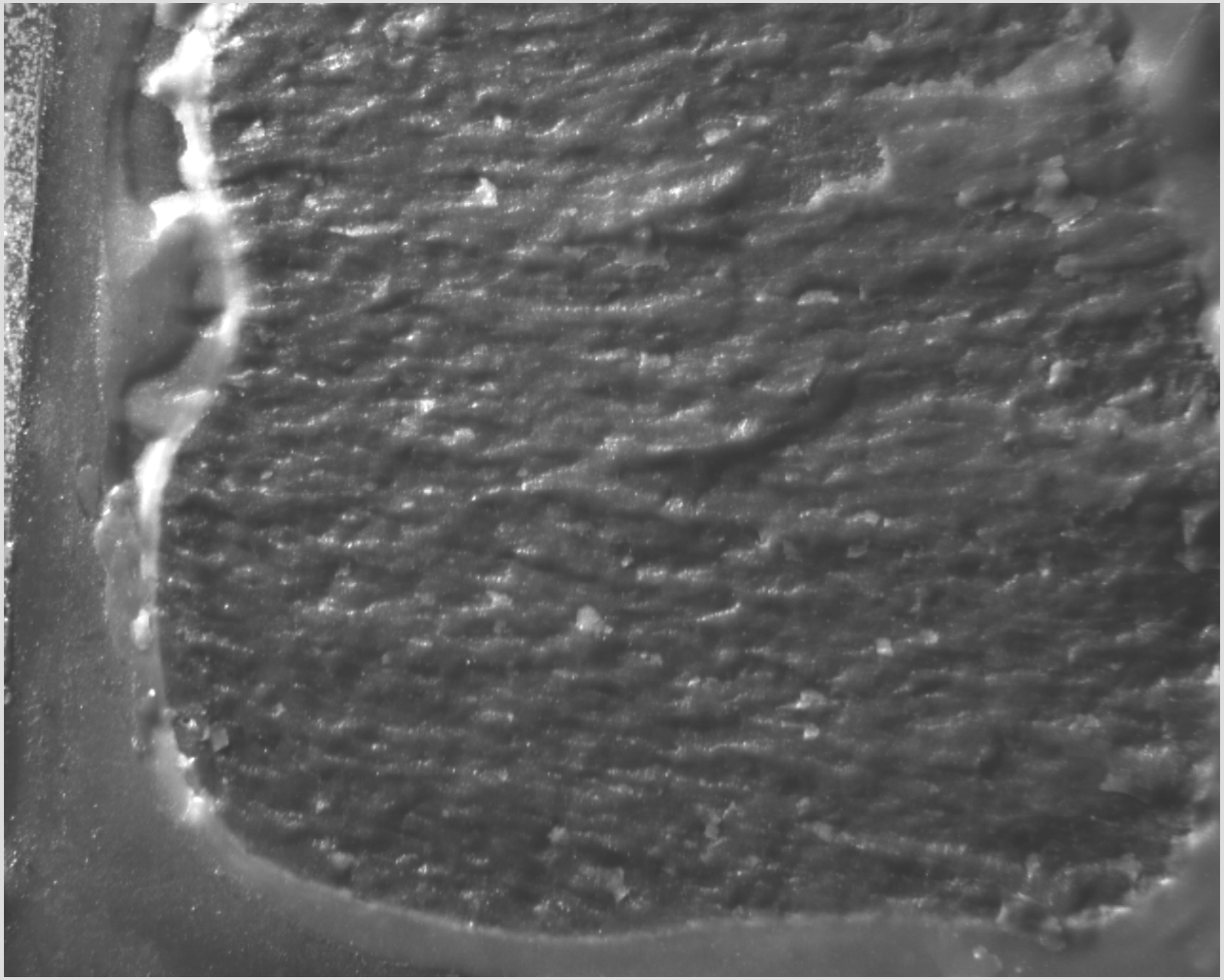
Mixed fracture



As-received

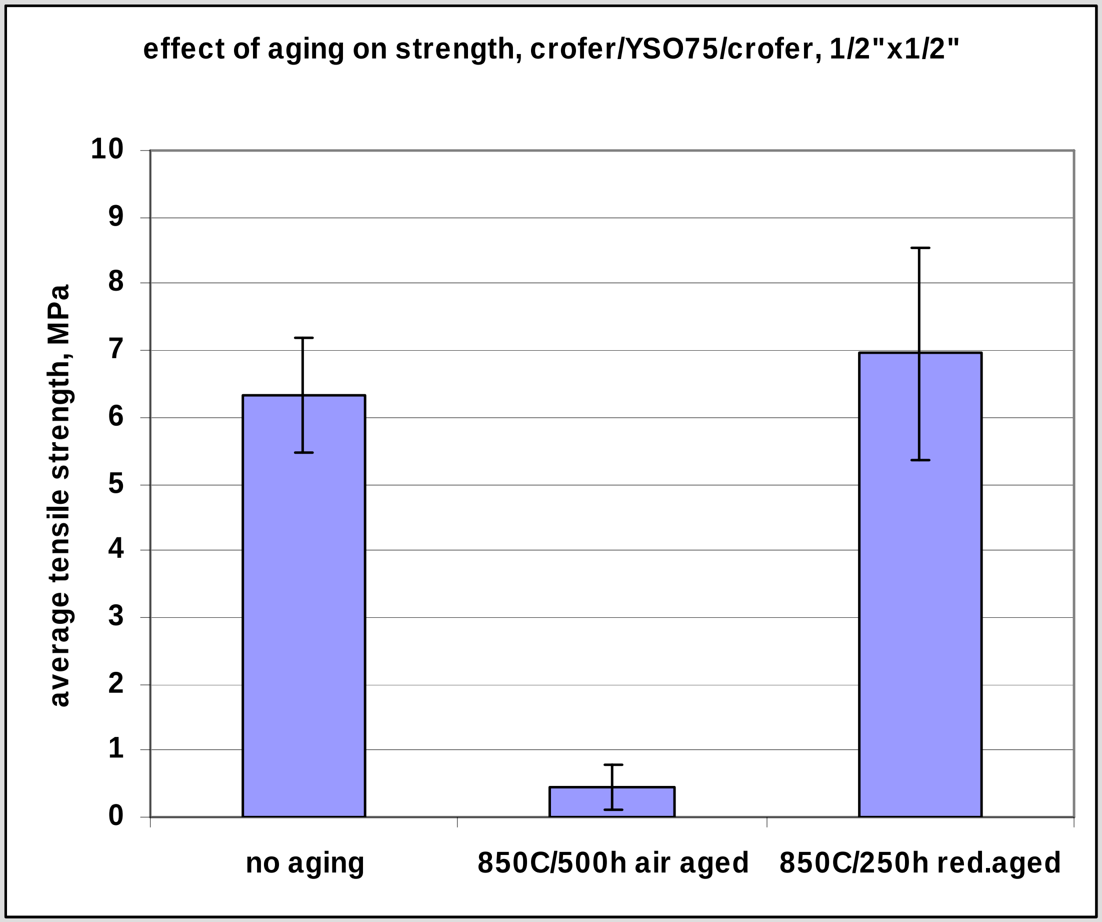
1000°C/2h
Pre-oxidized

Fracture initiated mostly from edge flaws



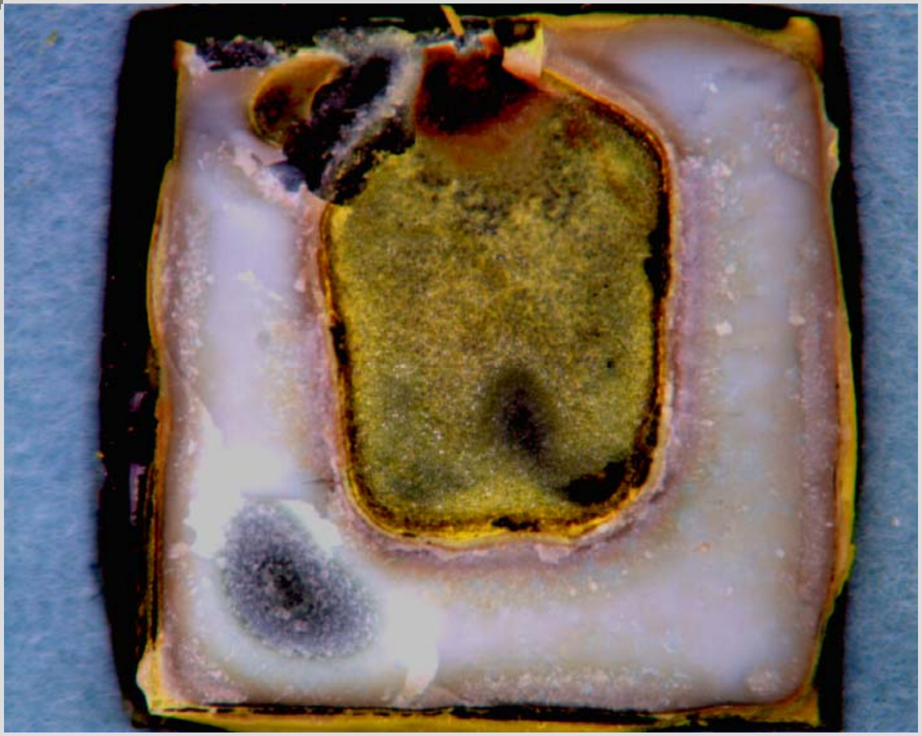
Effect of aging and environment

Air: 850°C/500h
Wet and reducing: 30%H_2O, 70%(2.7H_2/Ar) 850°C/250h

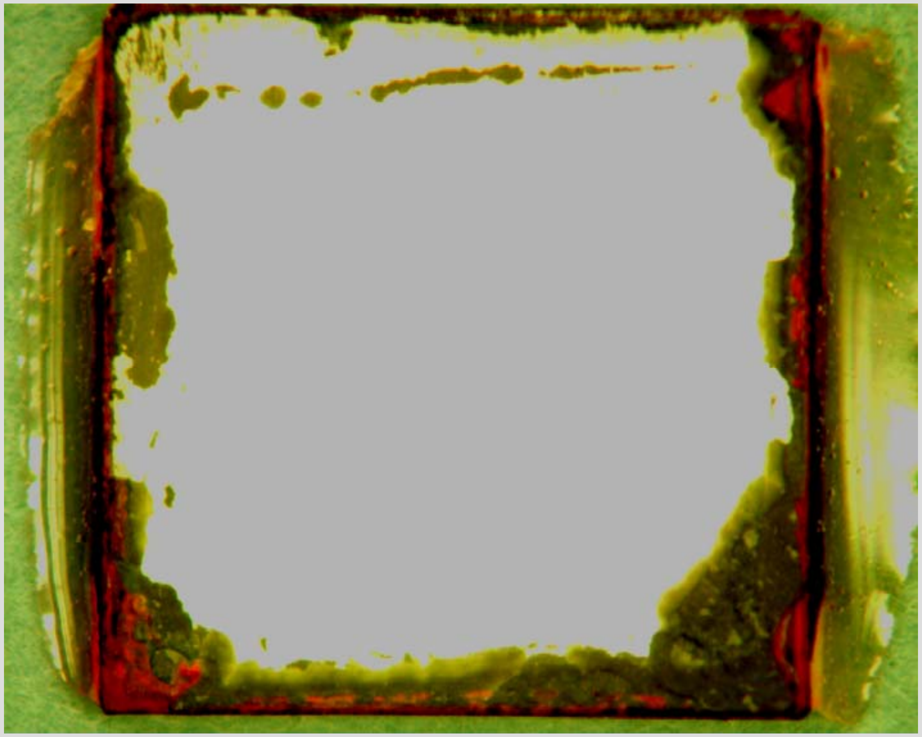


Fracture surface of aged sample

850°C/500h air



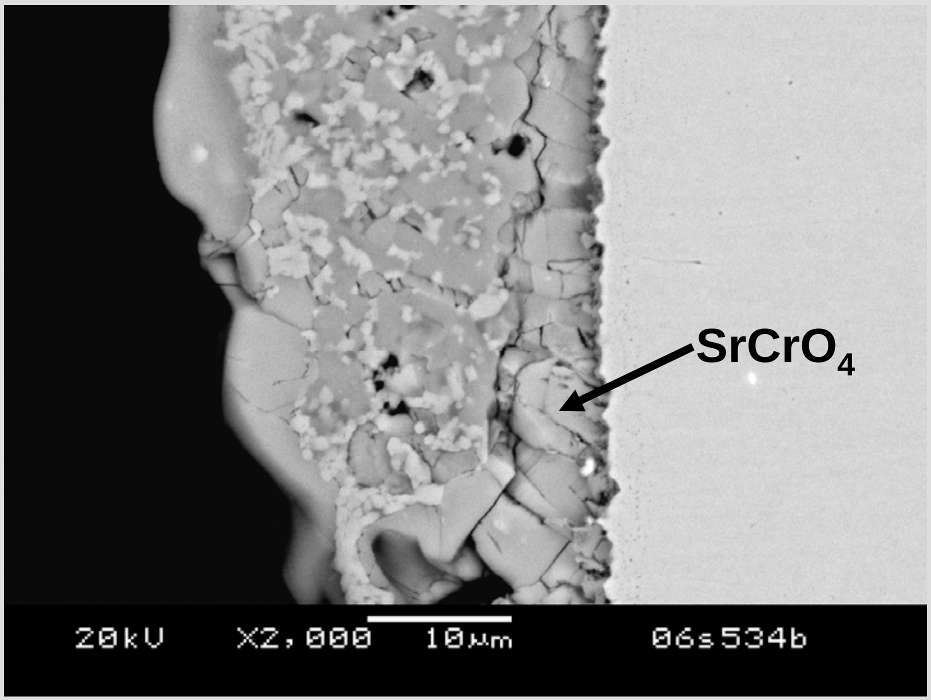
850°C/250h reducing gas



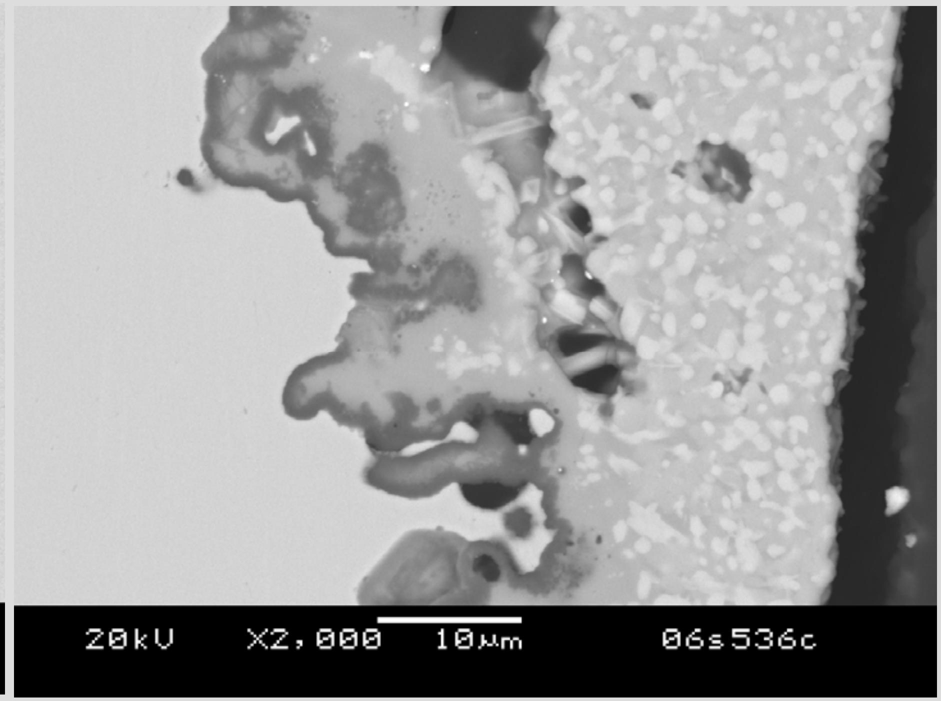
Presence of substantial amount of $SrCrO_4$

Aluminized crofer showed no chromate formation

Plain crofer

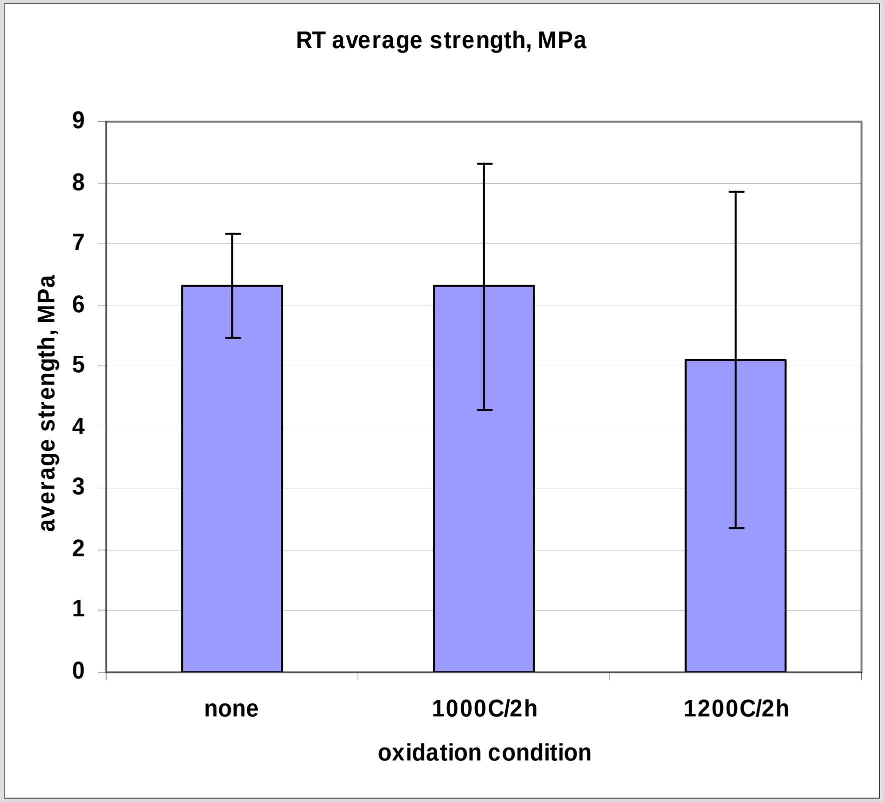


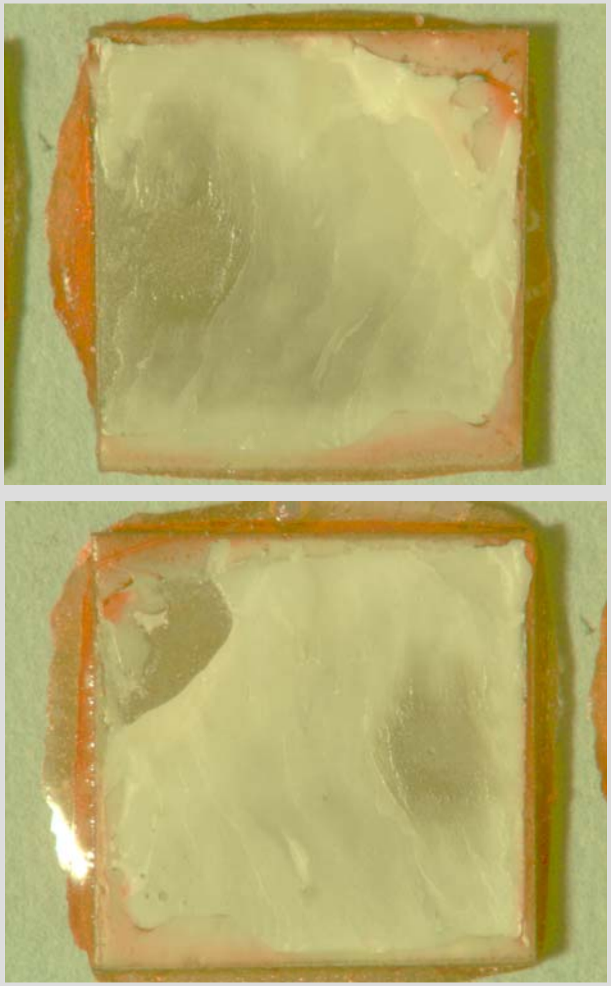
Aluminized crofer



Tensile strength of aluminized crofer from pack cementation

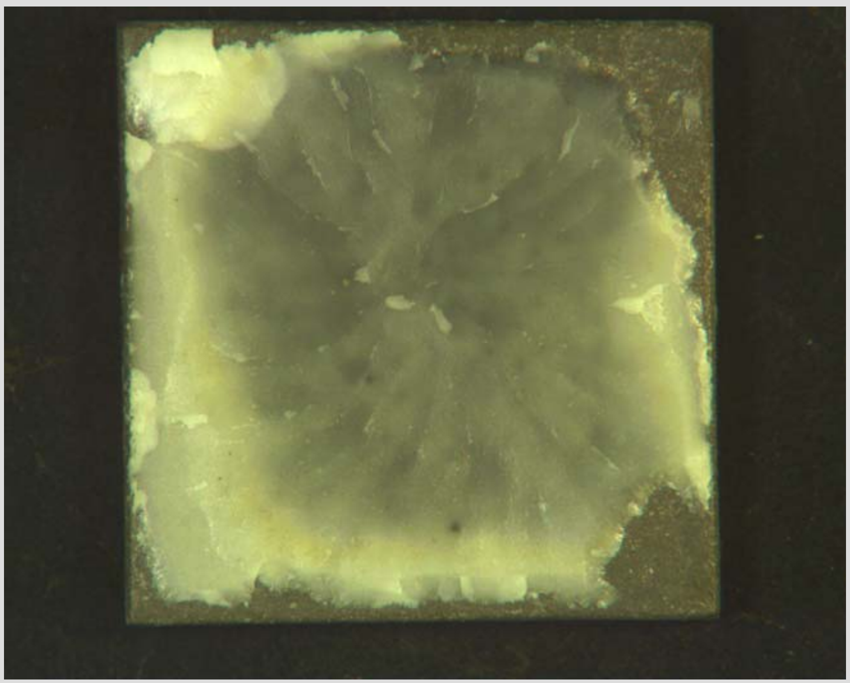
Pack cementation, followed by heat-treatment in air

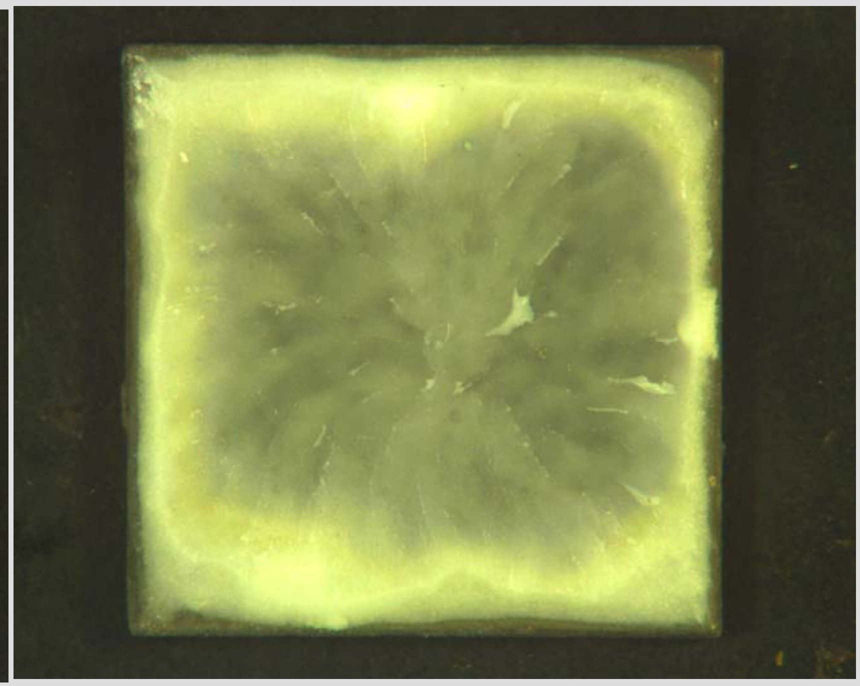




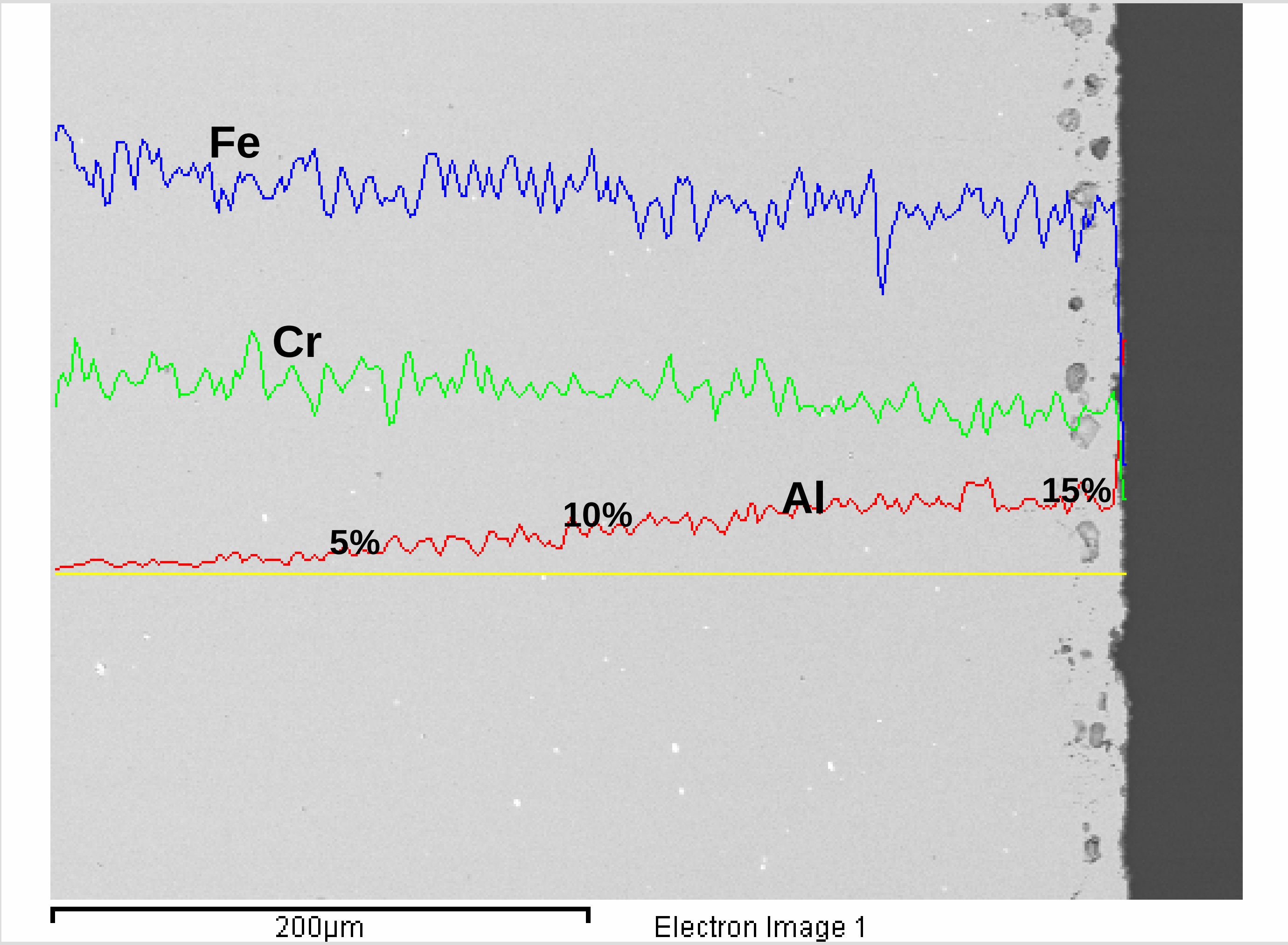
Sealing with aluminized crofer from vapor phase deposition

Vapor phase deposition followed by heat-treatment at 1000°C/2h air
As-sealed coupons all fractured through glass.
Glass bonded well to aluminized crofer coupons.



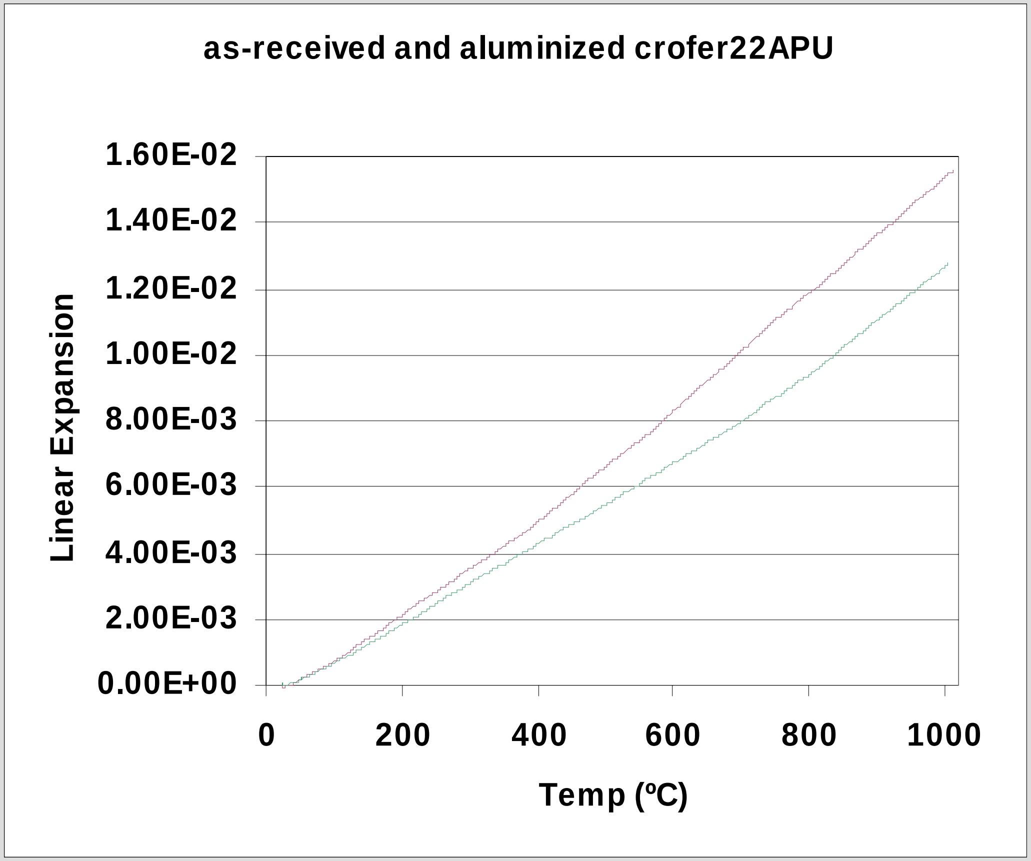


Appreciable amount of Al diffusion



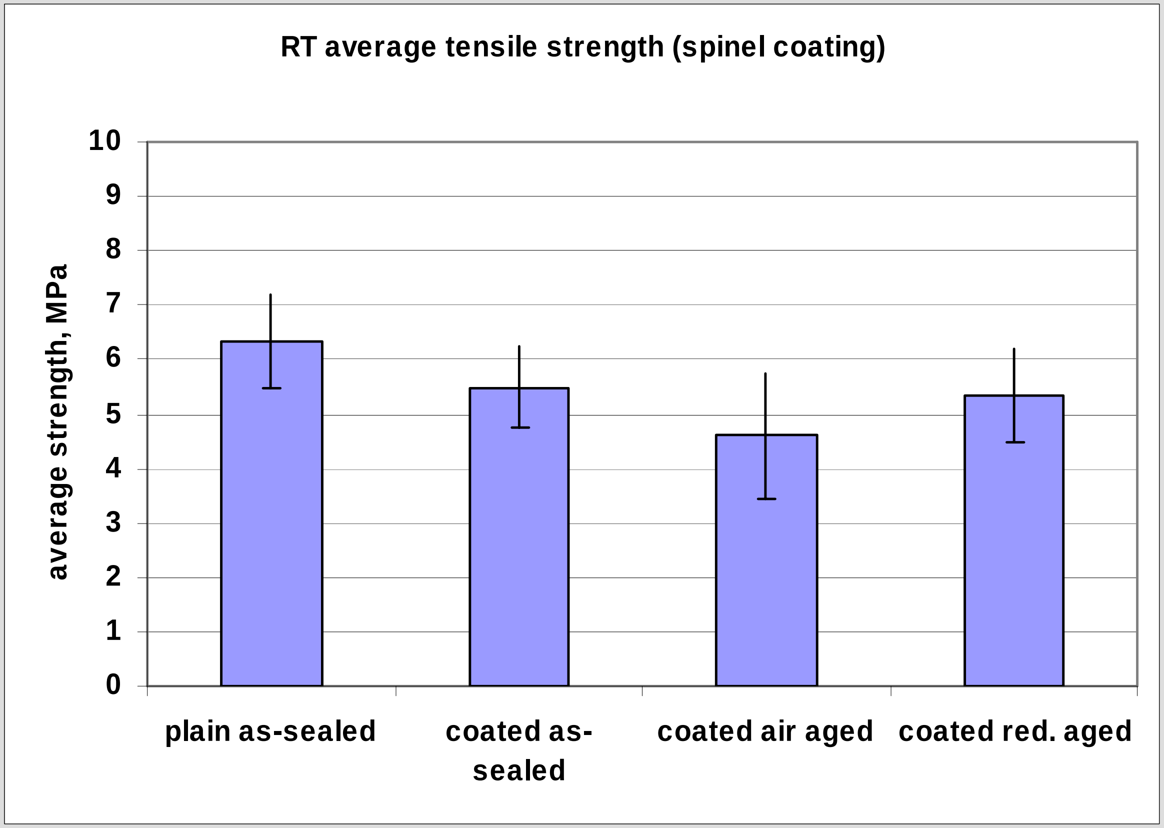
Issue of overdosing Al

Aluminizing by vapor phase deposition followed by heat-treatment at 1000-1100^oC/2h air



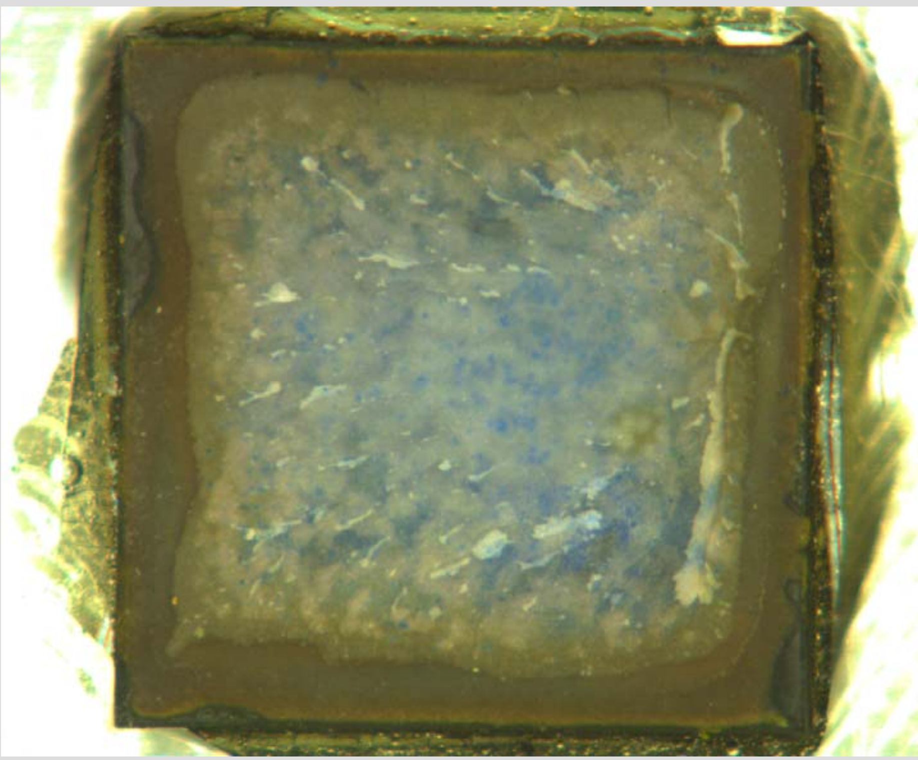
RT seal strength of spinel-coated crofer/YSO75 glass

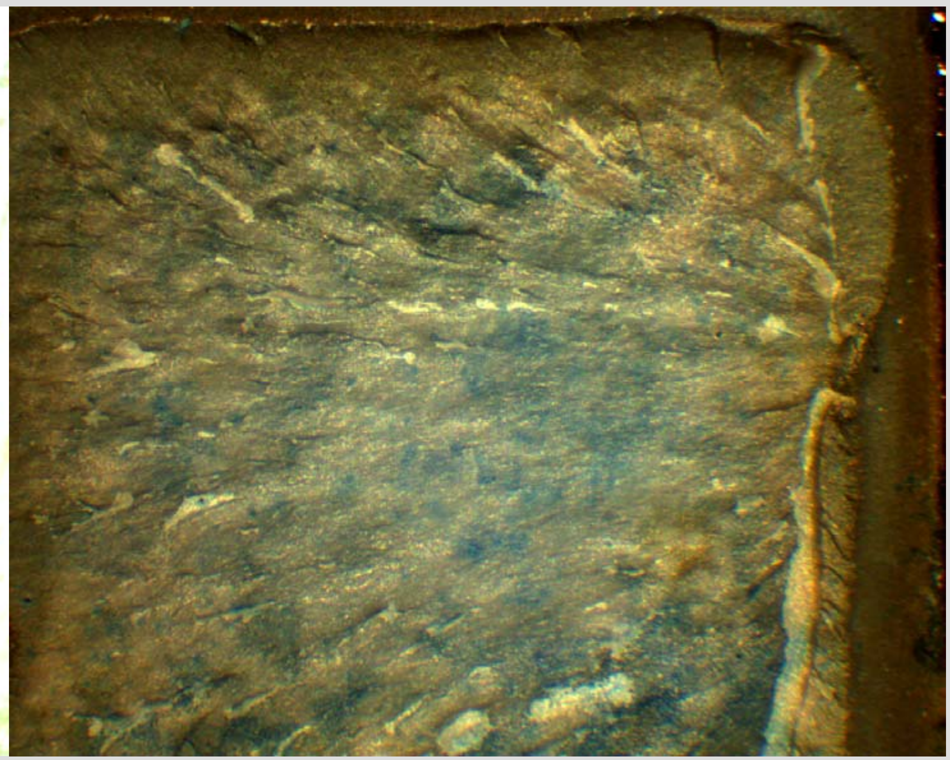
$(Mn,Co)_3O_4$ coated crofer22APU



Fracture surface of as-sealed $Mn_{1.5}Co_{1.5}O_4$-coated crofer22APU

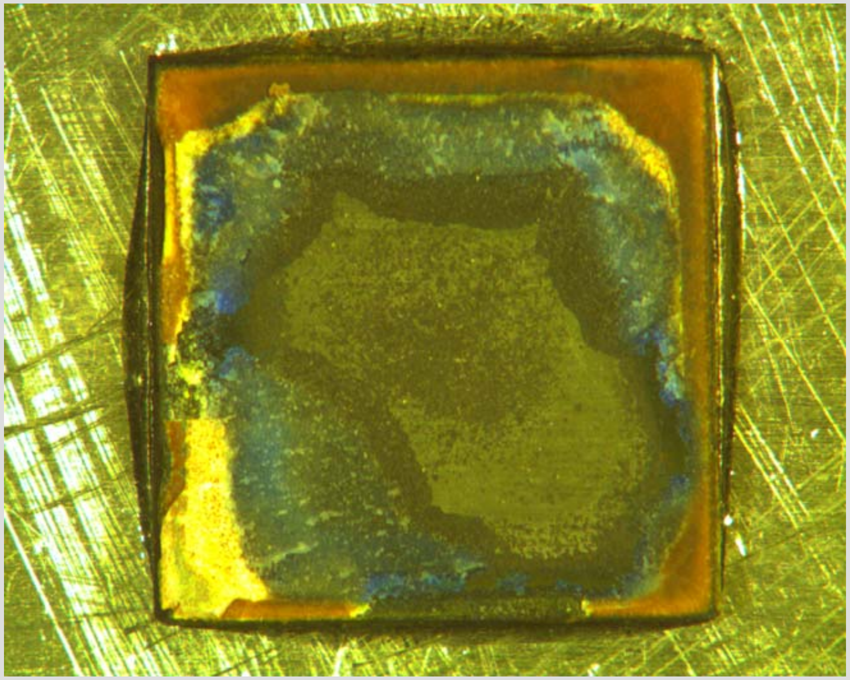
Fracture through glass, not at the interface. YSO75 not fully coverage
All 8 samples showed edge flaw

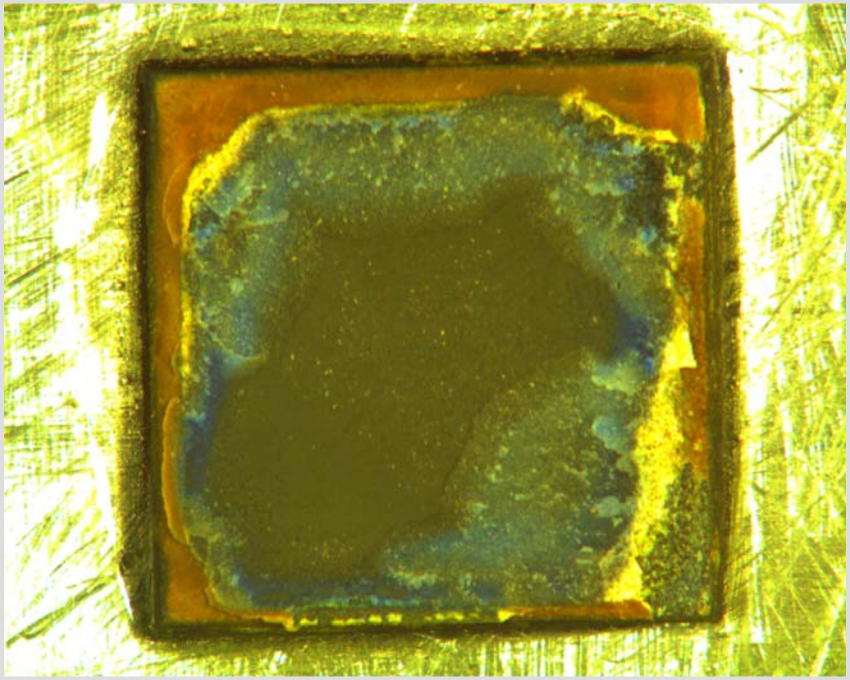




Fracture surface of air aged $Mn_{1.5}Co_{1.5}O_4$-coated crofer22APU

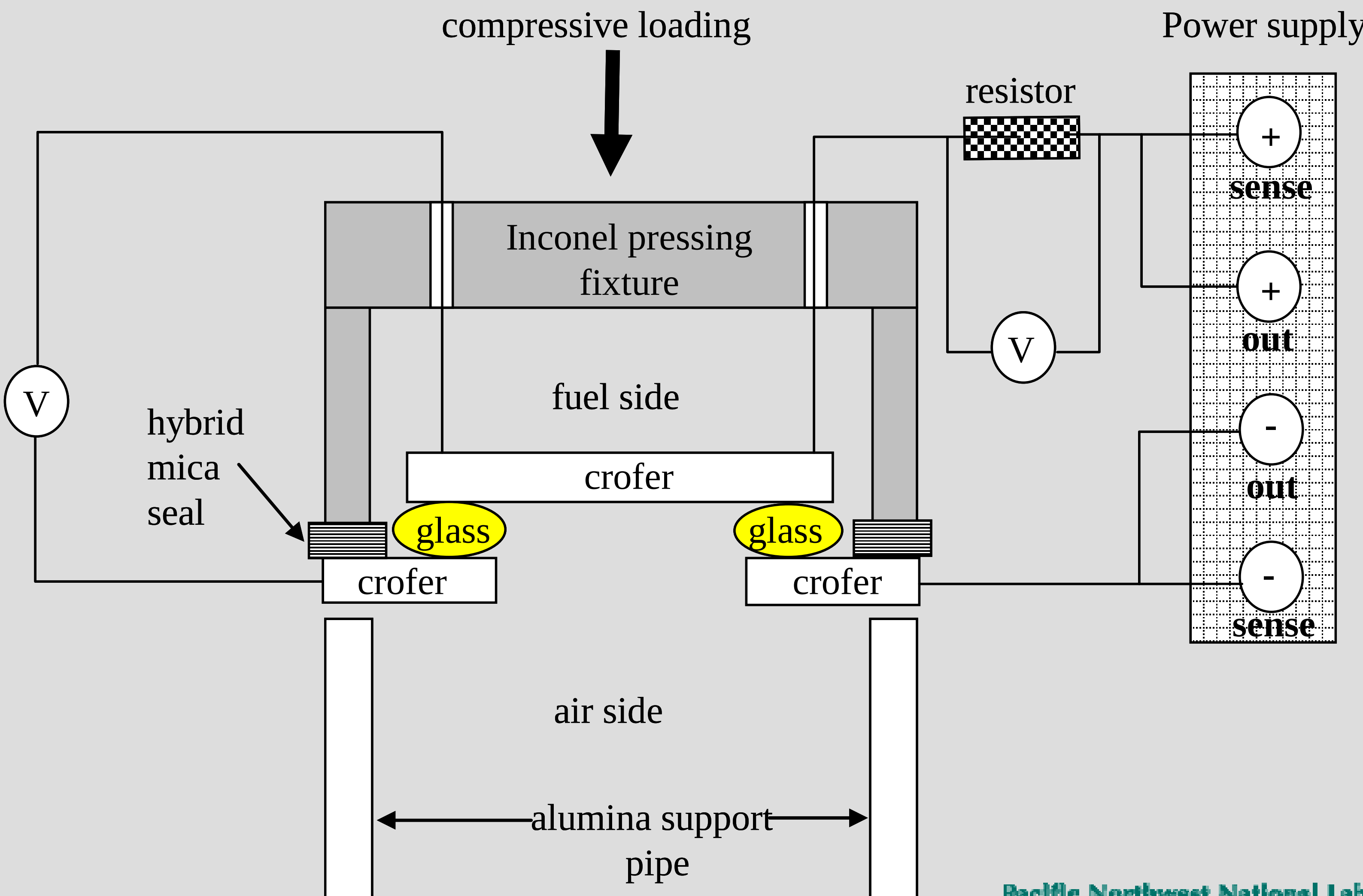
850^{o}C/500 h air





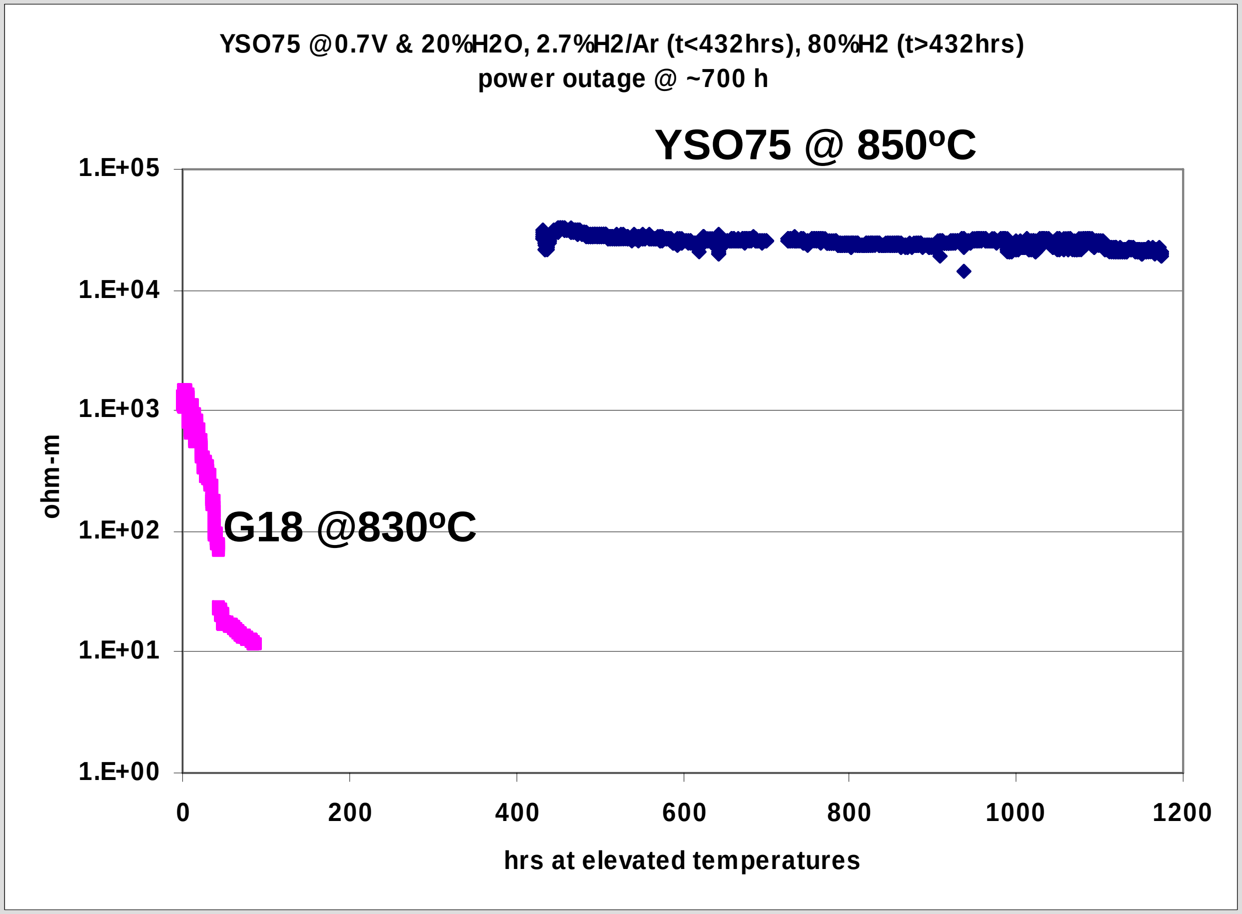
Suggest 5 μm coating was not enough

Setup for resistivity measurement

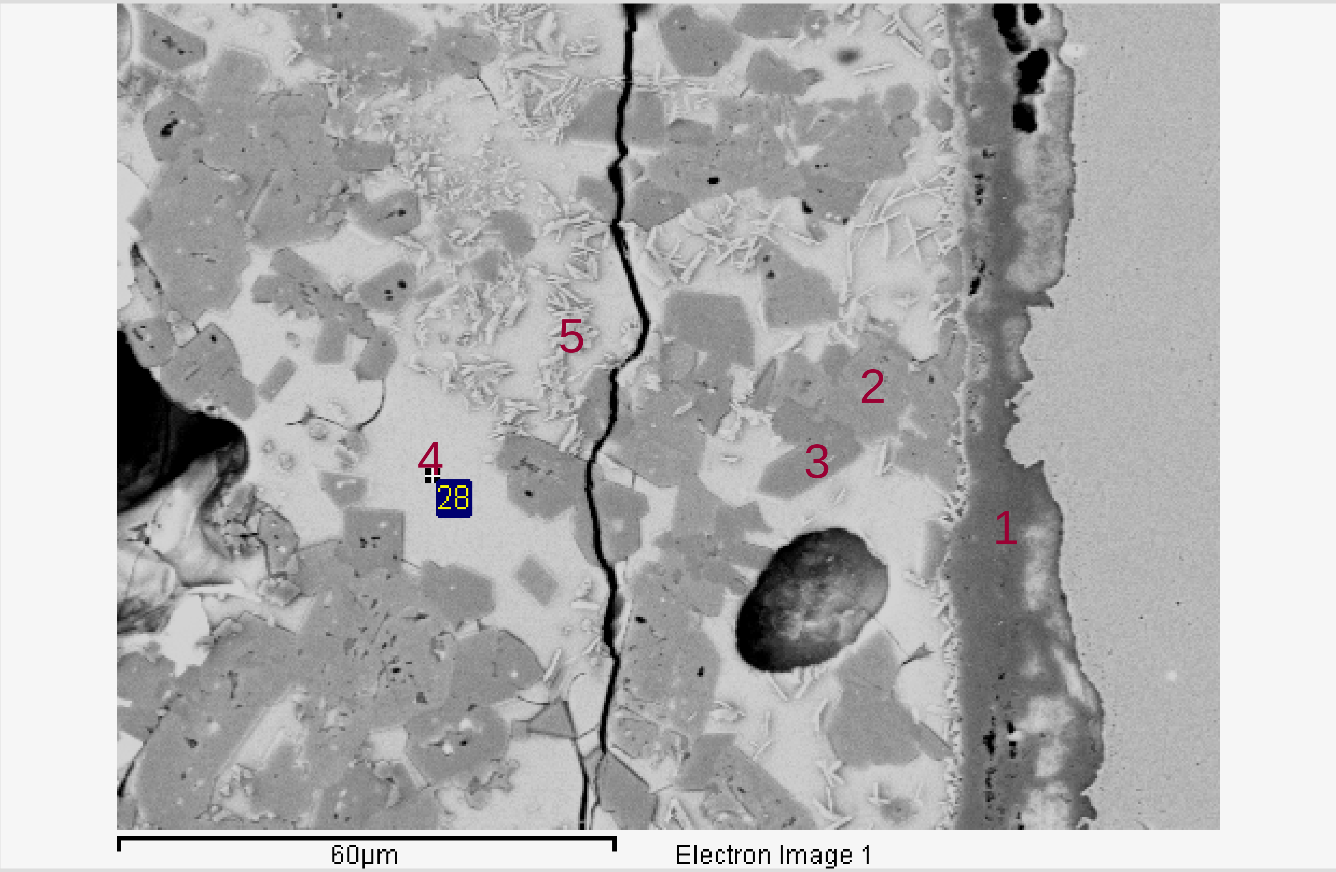


Good electrical stability for HT glass on plain crofer22APU

Crofer(as-received)/glass/crofer(as-received) @ 0.7V



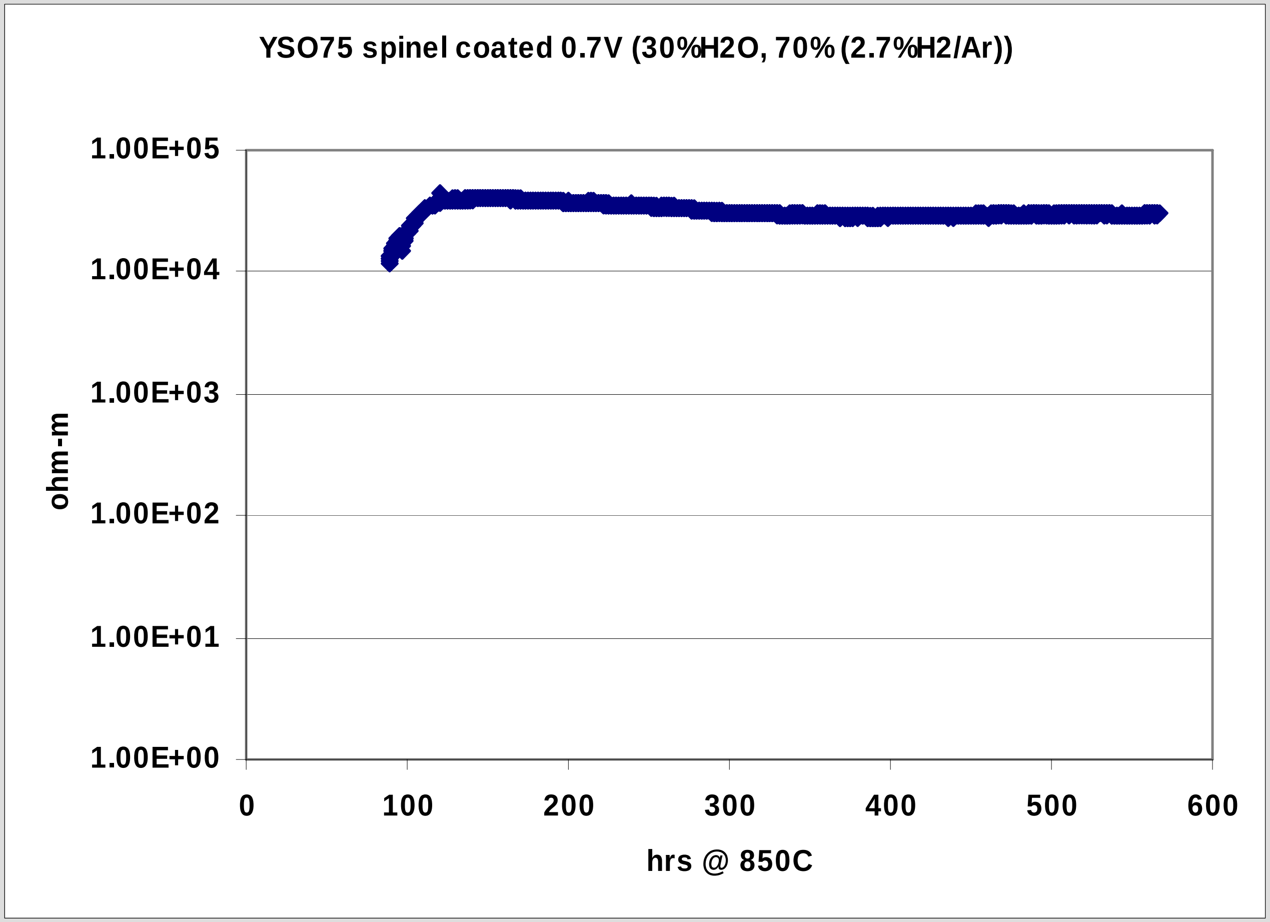
EDS showed substantial Fe diffusion



point	O%	Al%	Si%	Ca%	Cr%	Fe%	Ba%
1	43		1	1	50	4	1
2	44		27	16		1	12
3	43	20	24			1	12
4	47	3	16	6		7	21
5	44	1	5	2	11	28	9

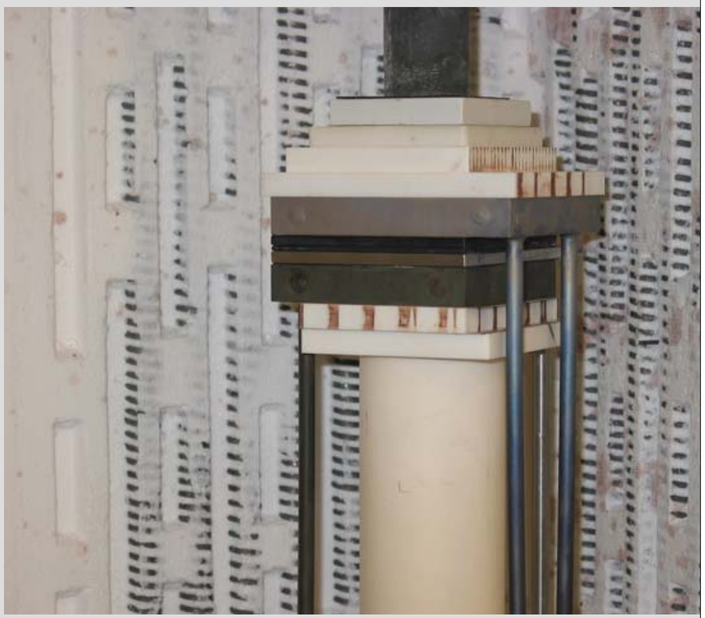
G18 near air side after electrical stability test

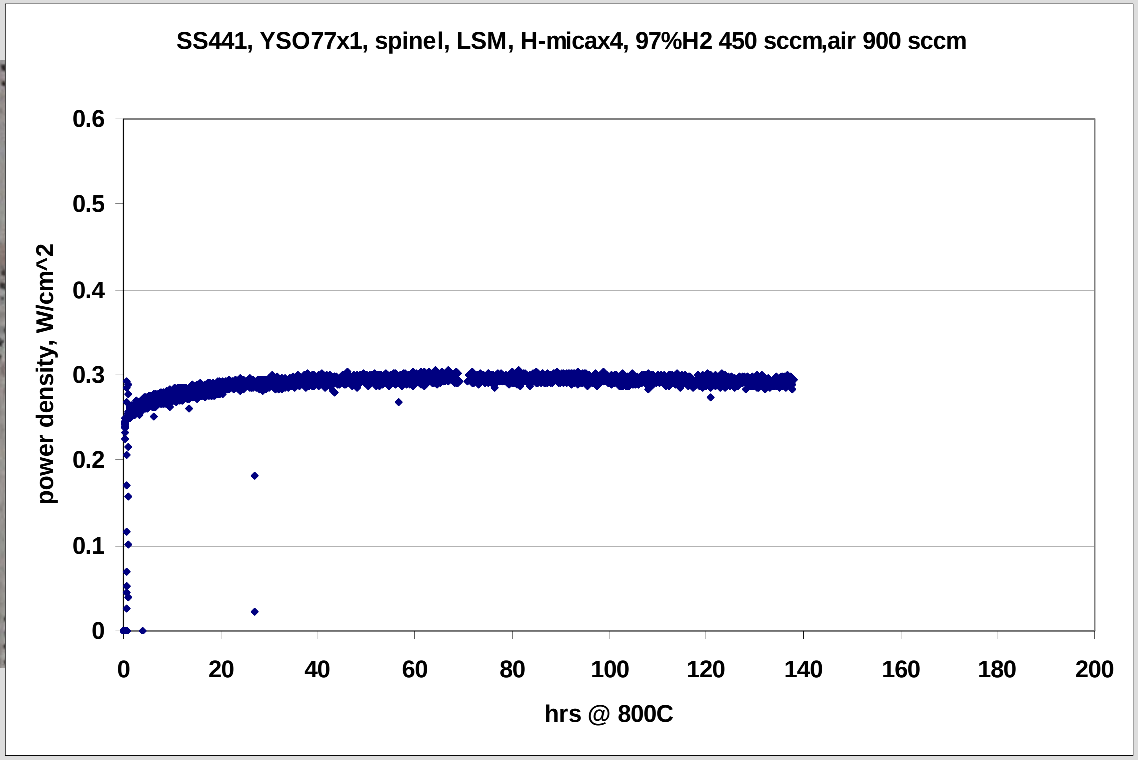
Good electrical stability for HT glass on $(Mn,Co)_3O_4$-coated crofer22APU



Materials set evaluation in 2"x2" singel cell test @ 800ºC

INDEC cell, SS441, spinel coating, LSM contact paste, aluminizing





Summary FY07

- Mechanical strength tests of M/G/M coupons showed strength degradation for uncoated crofer22APU only if aged in air or started with thick Cr_2O_3 surface layer.
- Aluminizing is effective in blocking Cr; however, overdosing would increase CTE for thin samples and leads to seal failure.
- $(Mn,Co)_3O_4$-coated crofer showed similar initial seal strength as plain crofer or aluminized ones. Minimal strength degradation when aged in air.
- HT glass showed very good electrical stability in SOFC and DC loading at 850ºC up to ~1200h.
- Successful demonstration of 2”x2” single cell test at 800ºC with candidate materials set: HT glass, spinel coating, SS441, and LSM contact paste.

Future work

- Materials set validation with single cell (2”x2”) stack testing and standardize the design:

1. Sealing glass: high-temperature, self-healing and composite.
2. Metallic interconnect: SS441 (standard or low Si), crofer22APU
3. Protective coating: $(Mn,Co)_3O_4$, alumina

- Short-term (200-500h) performance test at 800-850^{o}C
- Short-term thermal cycling test (800^{o}C/24h, cool to RT)x10
- Collaboration with modeling work
- Strengthening of candidate high-temperature glass with reinforcement