Thin-film coating for Mu2e tungsten target emissivity enhancement - an SBIR Phase I summary

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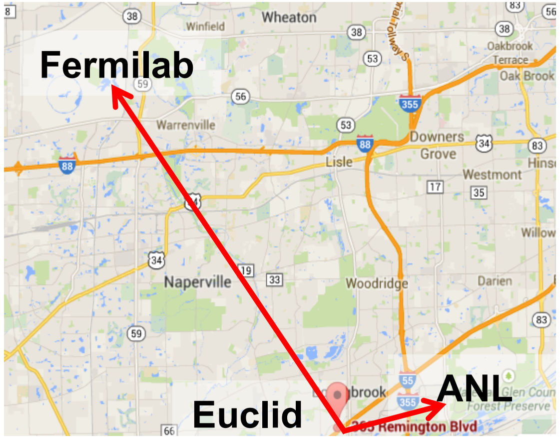
Project funded by DOE Office of science under contract SC-0019853, FNAL PI: Dave Pushka

About Euclid Techlabs

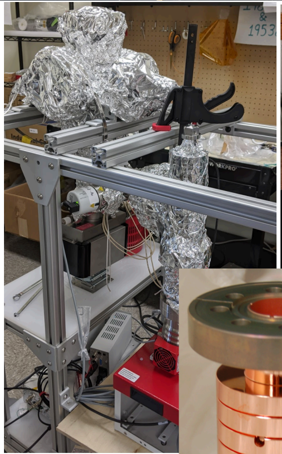
- Euclid TechLabs LLC, founded in 1999 is a company specializing in the development of advanced accelerator technologies, new designs for beam physics and high power/high frequency applications. Recently, Euclid has rapidly expanded our list of capabilities (Company Link):
  - Particle Accelerator Development
  - Sputtering Thin Film Deposition
  - Femtosecond Laser Machining of Exotic Materials including Diamond
  - Advanced Ceramic Sintering and Treatment
  - RF and Beam Simulations
  - Mechanical and Thermal Engineering
  - Fabrication and Prototyping
  - RF and Beam Lab Equipment
  - Low Level RF System Design
  - UHV suitcase for photocathode transfer

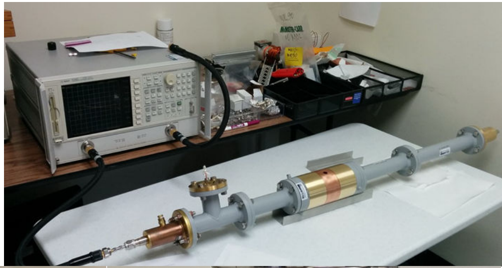
Visit us at our Lab Facility @ Bolingbrook, IL

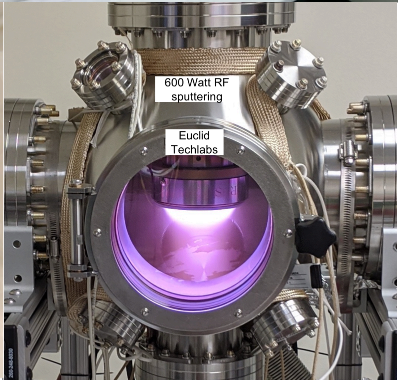


Our Lab Facility

- Available in our 11,000 sq ft facility:
  - Compact electron accelerator test area (MeV beam bunker)
  - Time resolved TEM stand (200 keV DC e- beam from a TEM gun)
  - “Cleanroom” and magnetron sputtering (Metals or dielectrics)
  - Field Emission cathode DC test stand (Quantum Efficiency test stand being built)
  - RF test lab (bead-pull, network analyzer, etc.)
  - UHV system design and engineering (fabrication outsourced to fast turnaround machining providers)





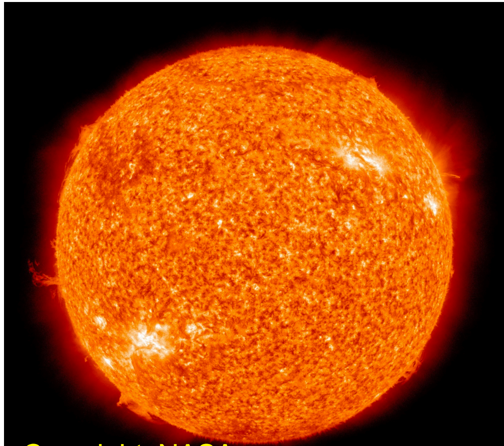


Euclid staff

- 15 Ph.D.s, 25 staff employees
  - Covering various aspects of expertise: accelerator physics, material science, chemistry, vacuum science, engineering (mechanical and electrical), programming.
  - 5+ PIs of SBIR projects (15+ ongoing)
- Supervise interns (undergrad and Ph.D.) and Ph.D. students (IIT, NIU, etc.)
- Close collaborations with virtually all U.S. National Labs (Fermilab, ANL, BNL, SLAC, Jlab, etc.)
- Me:
  - Indiana U + Fermilab Ph.D. program graduate (nuSTORM, A. Bross, D. Neuffer);
  - Fermilab postdoc (MAP + MICE)
  - Had interactions with TSD groups before this project (horn, target, MARS, etc.)
  - PI of 2 ongoing SBIR projects and Phase 1 of this project

Background

- **Passive cooling** for the *current* Mu2e production target design: heat transfer via thermal radiation.
  - No active coolant = risk of coolant leaks eliminated; remote target handling simplified;
  - Only radiation cooling = thermal fatigue and oxidation at high temperature.
- Target temperature depends on the deposited power *P*, target surface area *A*, and target material's total emissivity ε_T at all wavelengths.
  - $\varepsilon_T \sim 0.05$ to 0.3 depending on the polishing and aging for tungsten
- **A multidisciplinary and fun study!**



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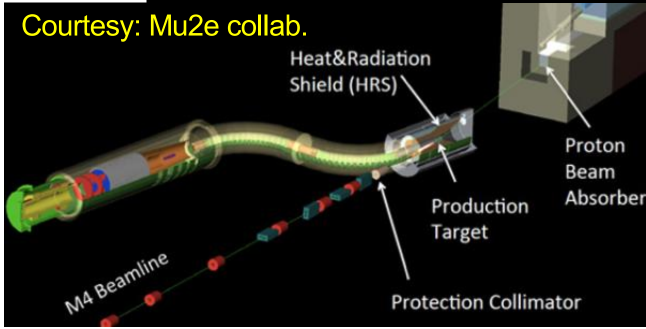
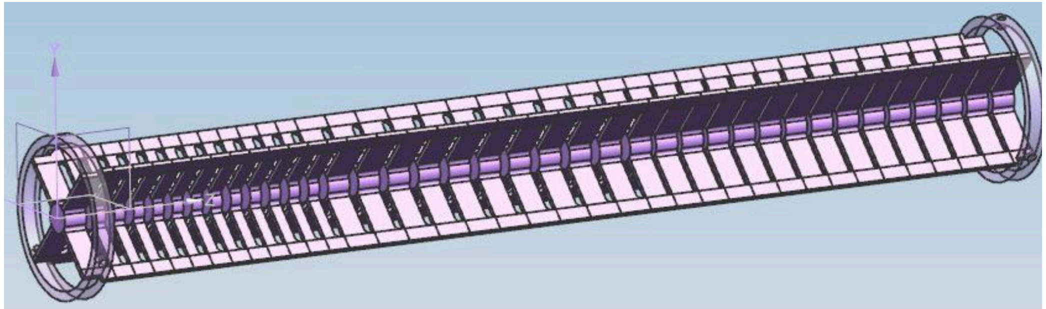


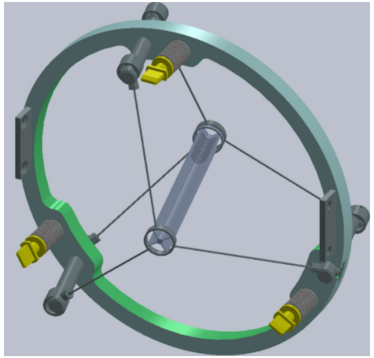
Courtesy: Mu2e collab.

Two ways to expedite the radiation cooling

- *Add surface area* to the target (current focus for the collaboration)
  - Added fins
  - Added segments
  - Compromising pion yield and cooling performance

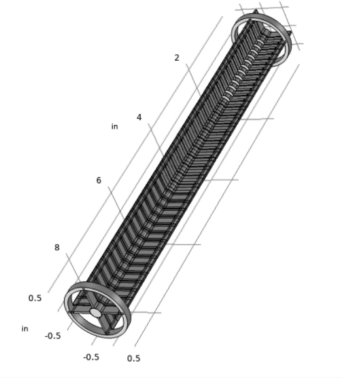


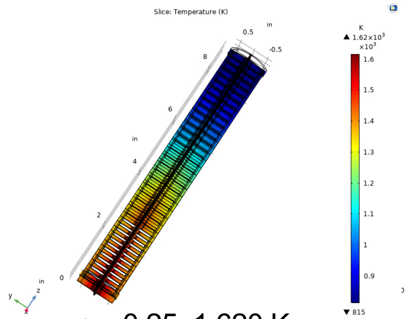
- *Thin-film coating* on the Tungsten surface to increase the thermal emissivity (focus for Euclid's SBIR)
  - Expedite radiation cooling from all coated surfaces
  - Shape-independent
  - Robust thin-film needed (> 1,500 °C) during operation



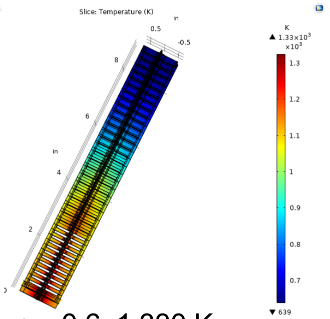
Guidance from simulation

- CAD model from Dave
- Thermal simulation done in COMSOL
  - Gaussian distribution in *x* and *y*, $\sigma_x = \sigma_y = 1$ mm;
  - Truncated Gaussian in *z*, $\sigma_z = 3$", peak at $z = 0$;
  - 4 kW *absorbed DC* beam power
- Define emissivity for *each* surface (not an easy task!)
- Two ε_T compared: 0.25 & 0.6
- Corresponding peak temperature: 1,620 Kelvin and 1,330 Kelvin, respectively
  - Room temperature assumed for the ambient environment;
  - No heat transfer from conduction or convection – only radiation.





ε_T=0.25: 1,620 K

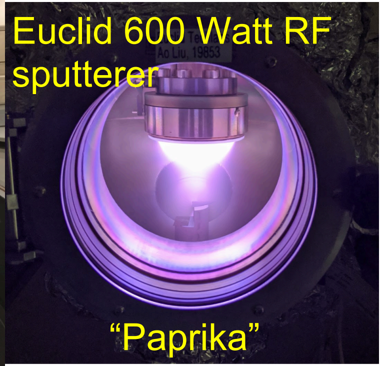


ε_T=0.6: 1,330 K

Getting the thin-film to grow on tungsten

- Two main categories:
  - Chemical Vapor Deposition (CVD): repeatable; less available; chemical safety hazard;
  - Physical Vapor Deposition (PVD): many setup parameters / less repeatable; less hazard; sputtering is one PVD method: available in-house at Euclid.

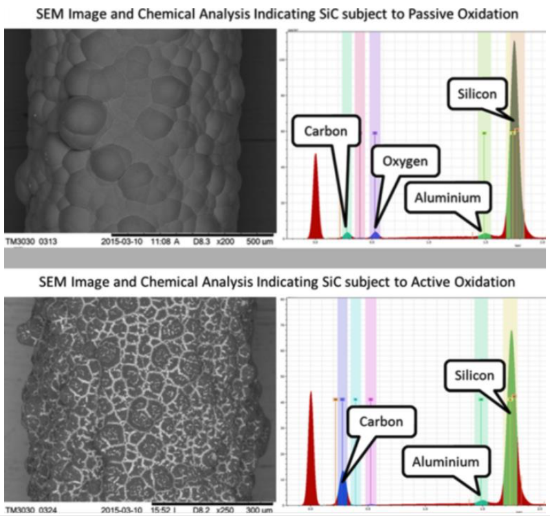
- Thanks to this project, Euclid has access to equipment at center of functional nanomaterials (CFN) of BNL (dedicated proposal).
  - PECVD, SEM, XPS, Filmetrics tools, etc.
- Euclid also has access to the center of nanoscale materials (CNM) of ANL
  - Similar tools available but mostly more restrictions



Paprika: 4" Magkeeper target; Conductive/non-conductive: TiN, Cu, Mo, SiC, etc.; UHP Ar gas; mid e-8 Torr

Candidate materials for thin-film coating

- Most materials with high ε_T and high melting point are non-conductive
- Most non-conductive materials not accepted/workable at CFN/CNM
  - Contamination concerns;
  - High sputtering power needed;
  - PECVD recipe limitation (Si-based compounds only)
- High power RF sputtering beneficial



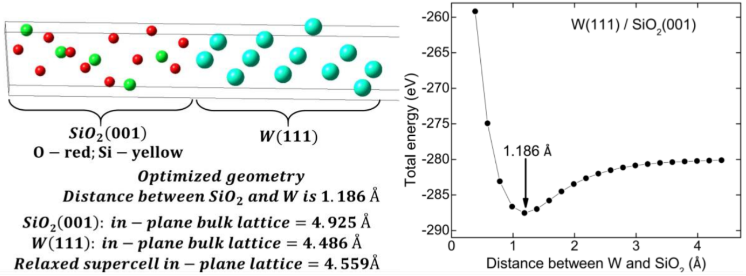
Densham, et al.

Candidate materials that pass these filters and are doable by Euclid:

- **SiN** (melting pt: 1900 °C, ε_T=0.85 to 0.93 (bulk), oxidizes in poor vacuum, PECVD)
- **SiO_2** (melting pt: ~1700 °C, ε_T~ 0.7 (bulk), robust against oxidization, PECVD/sputtering)
- **AlN** (melting point: 2200 °C, ε_T~ 0.6 (bulk), might oxidize in poor vacuum, Sputtering)
- **SiC** (melting point: 2730 °C, ε_T~ 0.8 (bulk), Rutherford of UK tested but fatigue (oxidation) was seen, only one sample tested. Sputtering)

Density function theory (DFT) simulations

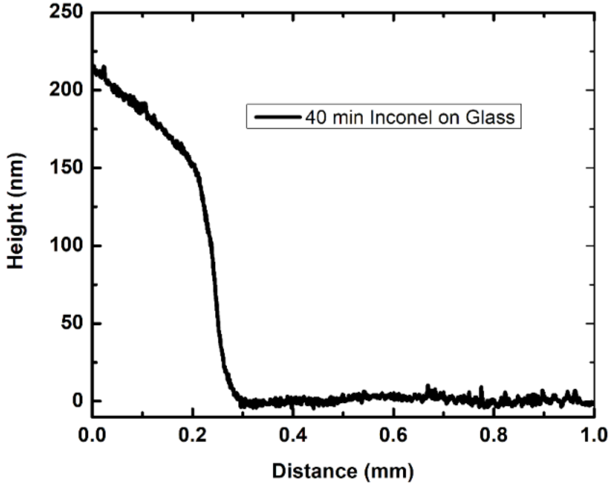
- Quantitative characterization of thin-film binding with substrate
- Calculates “work of separation” through “total energy”
  - Solves Schrodinger wave equations of molecular nuclei-electron system
  - Extremely computing resource hungry
  - Two layers (one epitaxially grown) of slabs: stable or not?
- SiO_2 (001) on W (111)
- Energy minimum at 1.186 Å: epitaxial growth of SiO_2 (001) on W (111) is energetically favorable.
  - Single crystal assumed, result might vary in reality

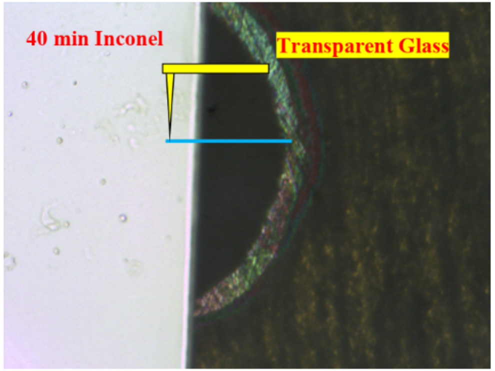


More materials can be Simulated in future studies

Early thin-film deposition practice: Inconel 600

- DC sputtering @ Euclid:
  - Inconel 600 coating, 40 mins:

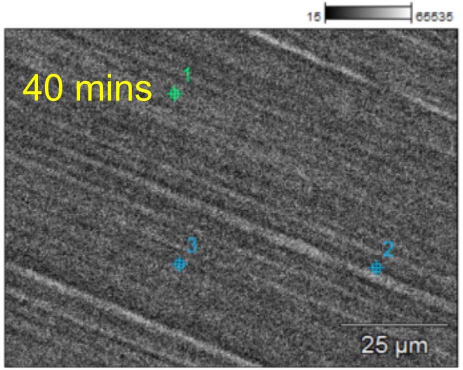


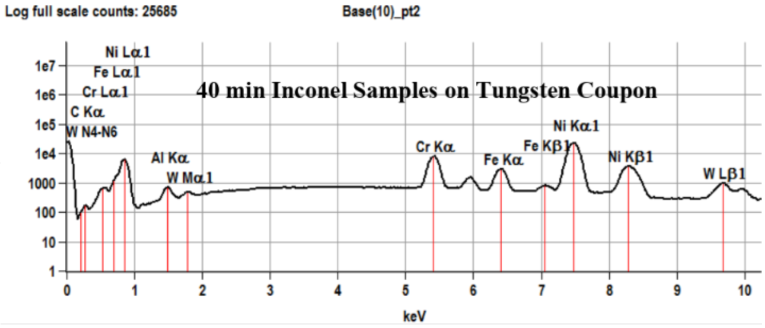


~ 200 nm thick from profilometer

Strong bonding of the thin-film with W substrate.

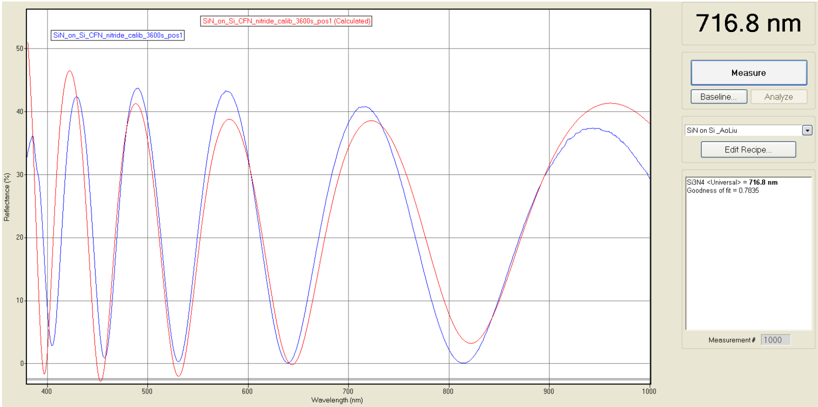
EDX and SEM scans done

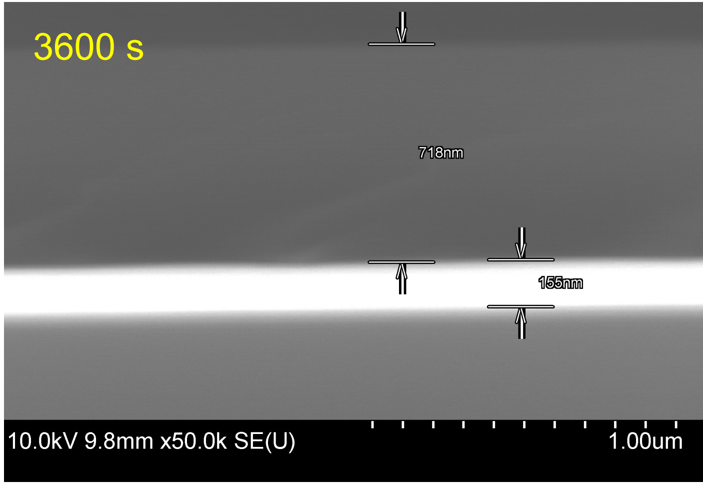




Thin-film coating practices: SiN_x

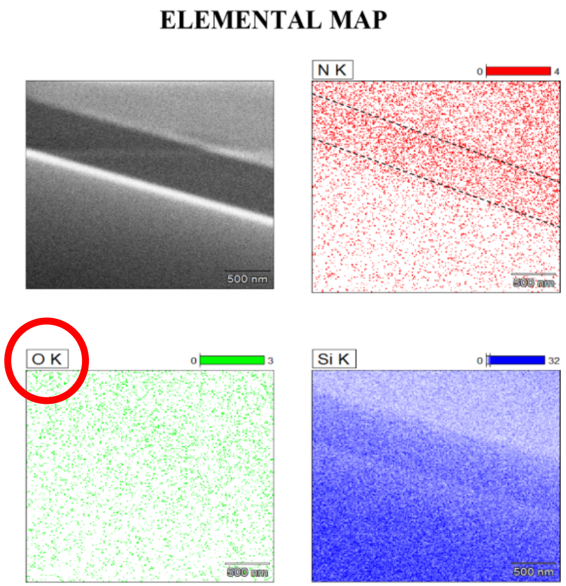
- PECVD @ BNL:
  - SiN_x on Si wafer and W
  - "*x*" needs to be determined stoichiometrically
  - Cross-section SEM (right) + Filmetrics thickness measurements - consistent: *~700 nm* for 1 hour and *~1400 nm* for 2 hours.
    - Thickness also confirmed by the Filmetrics F20 measurement



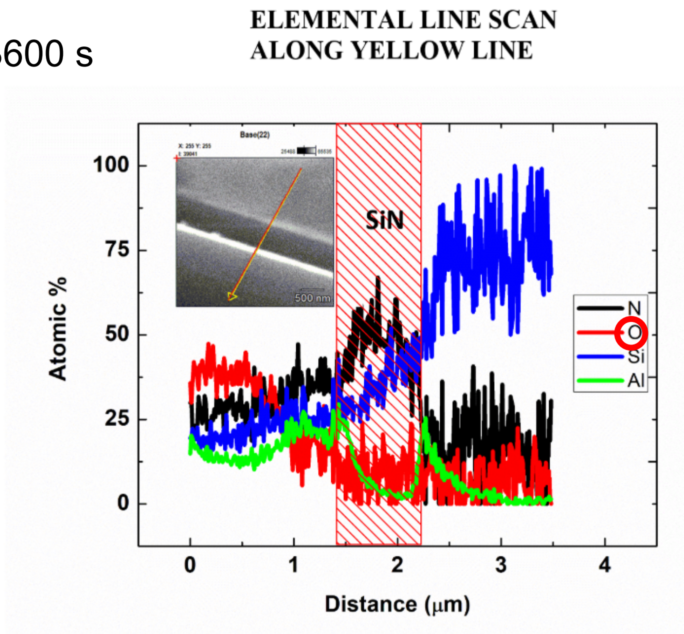


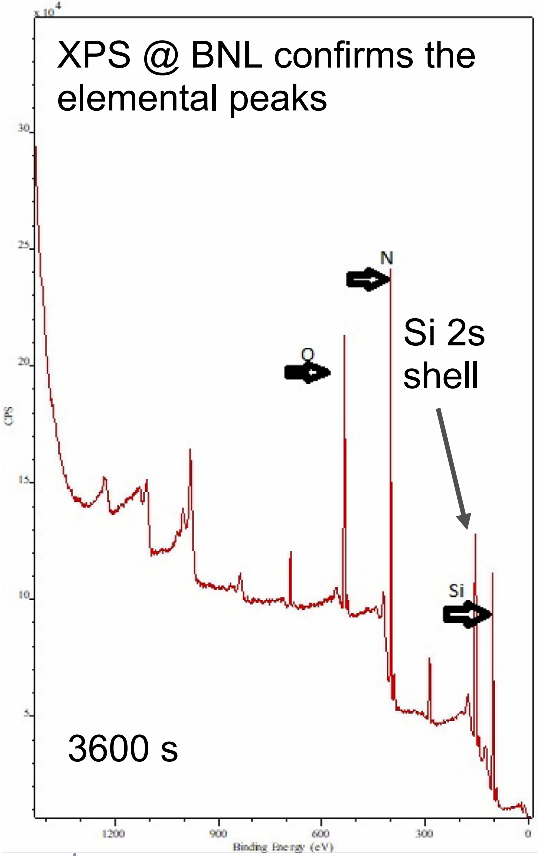
Thin-film coating practices: SiN_x (Cont'd)

- EDX elemental mapping done (on Si wafer):



3600 s

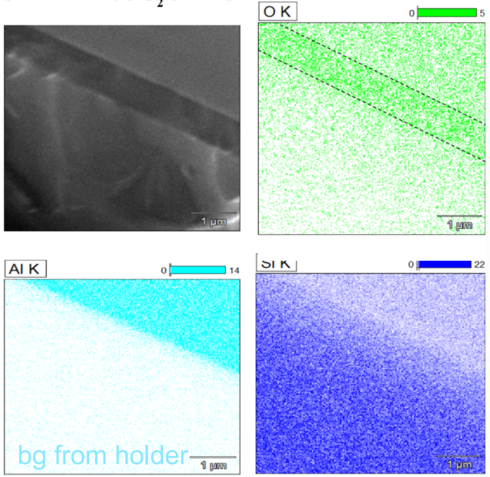




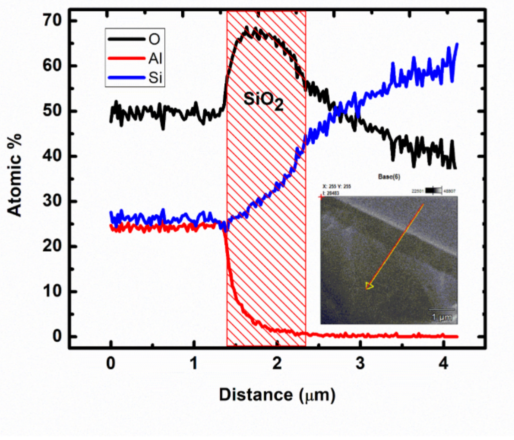
Some oxidization/impurities was expected and seen (in air and during deposition)

Thin-film coating practices: SiO_{2-x}

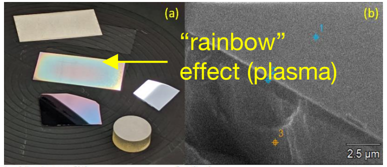
SAMPLE ID: SiO_2/Si 900 s

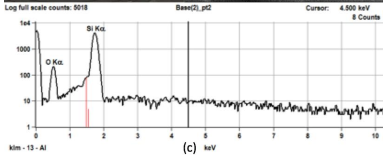


ELEMENTAL LINE SCAN
ALONG YELLOW LINE

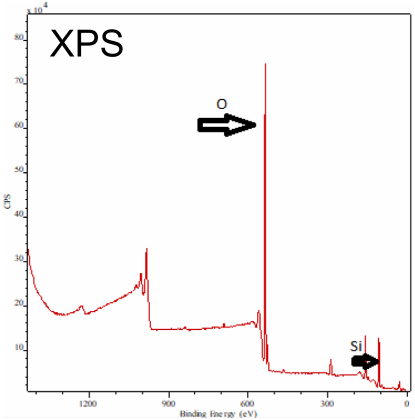


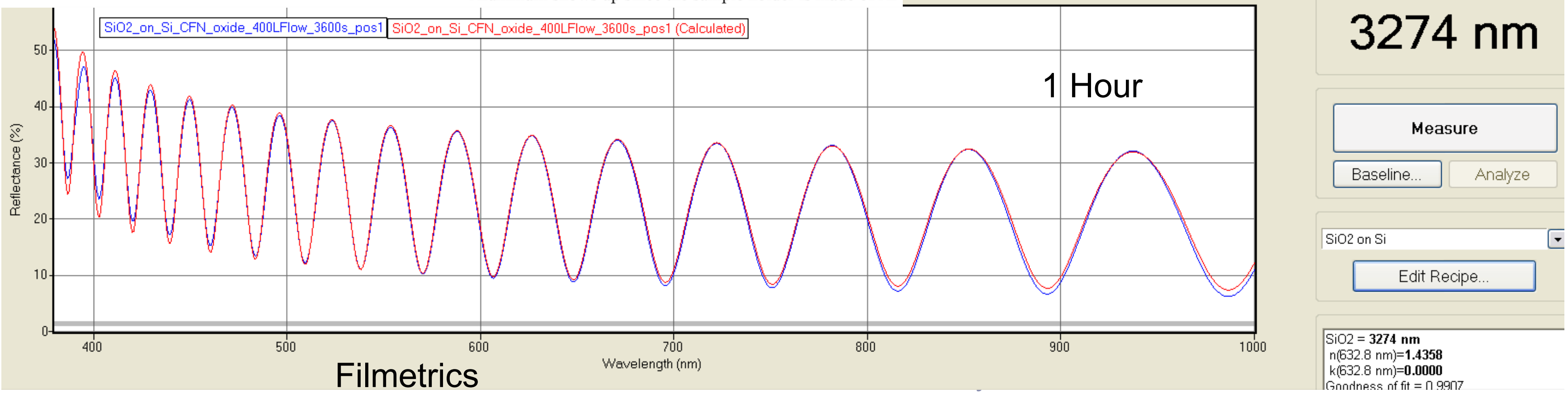
Aluminum shows up since the sample holder is made of Al





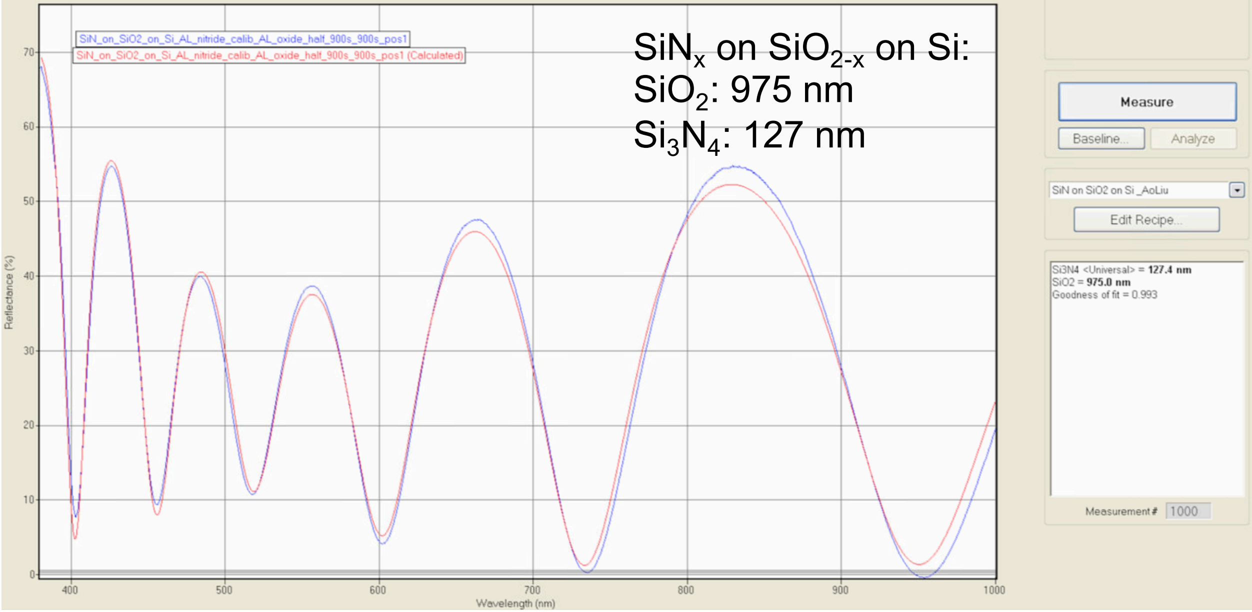
- PECVD @ BNL
- 3 µm of coated SiO_{2-x} in 1 hour





Thin-film coating practices: SiN_x and SiO_{2-x}

- An interesting test is to see the effect of combined thin-films
  - Goals/Possible benefits: Higher emissivity and better thermal expansion matching (and better bonding) than SiO_2, less oxidation than SiN_x (protection)

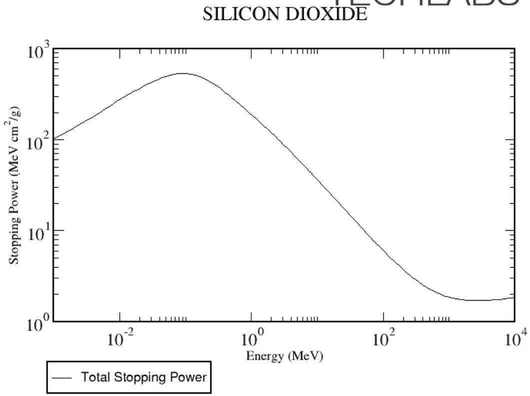


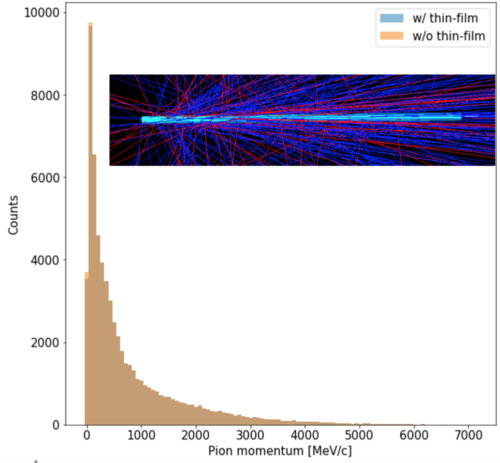
PECVD result;

SiN_x layer bonds with the SiO_{2-x} layer nicely.

Impact on production

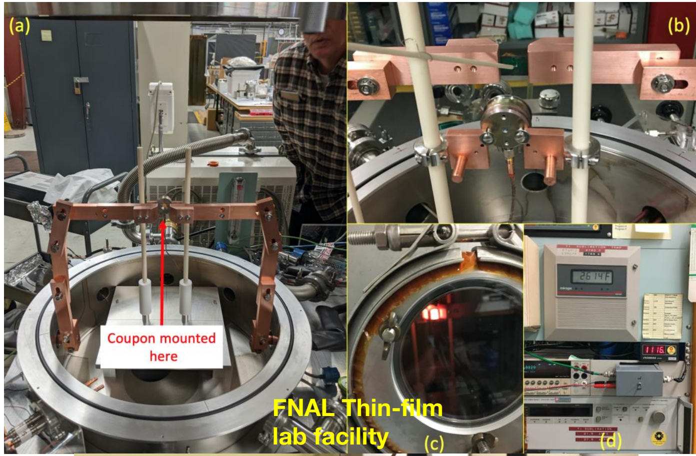
- Does the thin-film stop protons or pions?
- 1 µm SiO_2 ~ transparent to > 100 MeV-level hadrons
- Simulation in G4:
  - 8 GeV POT
  - 3 mm radius, 230 mm length W
  - Simulation divided into two steps: target then thin-film
    - Due to significant difference in tracking step size
  - Low-E bin heights vary due to thin-film
    - Will the physics group start yelling?
- May take iterations and get confirmed in MARS
- Critical study down the road!

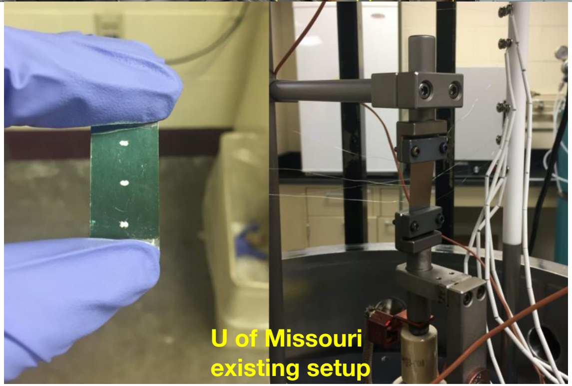




Preparing for emissivity tests

- "Conventional" way to measure ε_T
  - Joule heating
  - Pyrometer/thermocouple
  - Coupon dimensions selected considering:
    - Resistance
    - Deposition chamber/holder size
    - Availability
    - May be changed in future studies
- Two test sites:
  - Fermilab (CRADA contract); apparatus newly built; bumpy road to final setup;
  - U of Missouri (Mizzou) (intention to collaborate); existing setup; published emissivity measurement standard; started in 2020



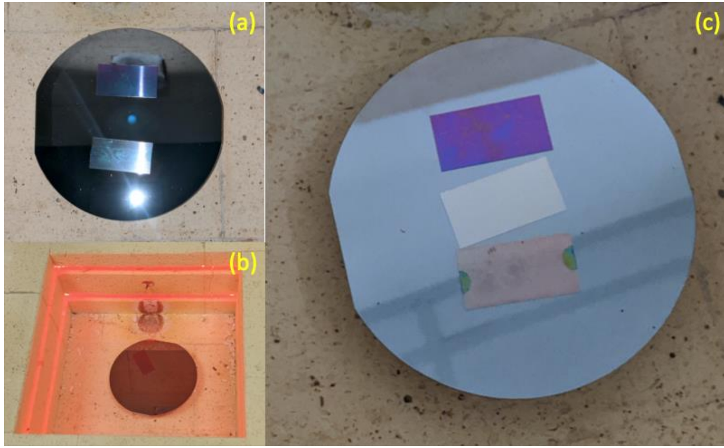


Emissivity measurement results

- Fermilab tests:
  - One coupon tested just before COVID hit – data not meaningful for emissivity calculation – feasibility study & prepare for continued tests in the future
  - Ramped to 200 Amperes, thermocouple reading 1130 ºC
  - Ready for more coupons (we have them)
- Mizzou tests:
  - Two coupons tested; high current source available but switch not with short notice; 100 Amps, 600 ºC reached
  - Adjusted ε_T for naked W: 0.40 (higher than literature by ~0.1)
  - Adjusted ε_T for SiO_{2-x} coated (1 µm) W: 0.47 (lower than bulk SiO_2 reported: 0.65)
  - Further systematic studies (more data) needed!
- Neither Fermilab nor Mizzou saw degradation or detachment of thin-film!

Poor man emissivity measurement in-house

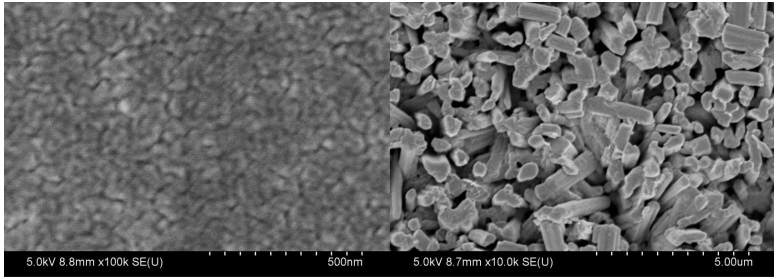
- Air furnace (high temperature) available at Euclid
  - Up to 1,100 °C
- Optical pyrometer (IR) + adjustable emissivity
- Coupons placed on Si wafer; heated together
- Assumptions (not perfect):
  - $T_{coupon} = T_{wafer} \sim= T_{sensor}$ at equilibrium
  - Oxidation of naked W is not too fast
- Adjust emissivity on pyrometer to match T
- Only ramped to 550 °C
  - Otherwise, naked W starts rapid oxidation
- **0.62±0.12** for coated; **0.35±0.18** for naked
  - Large errorbar due to measurement fluctuation

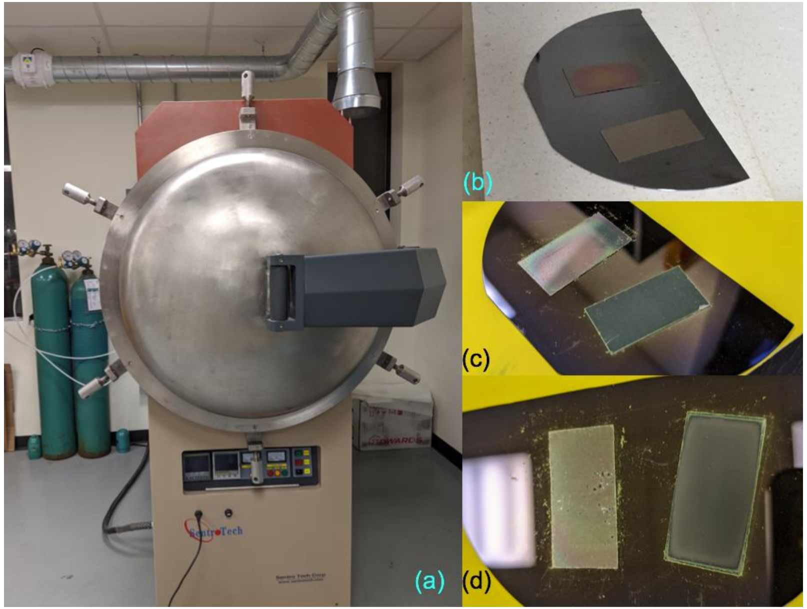


(a): initial status;
(b): in action (coated W taken out already);
(c): final status – top: naked, middle: new naked; bottom: coated (not on ears)

Thin-film protection for W at high temperature

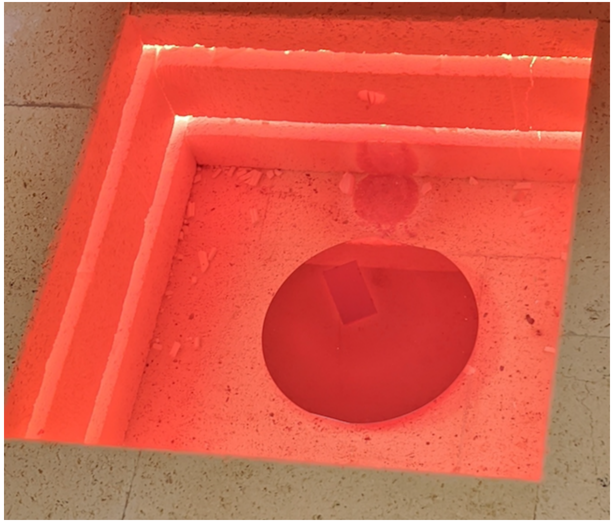
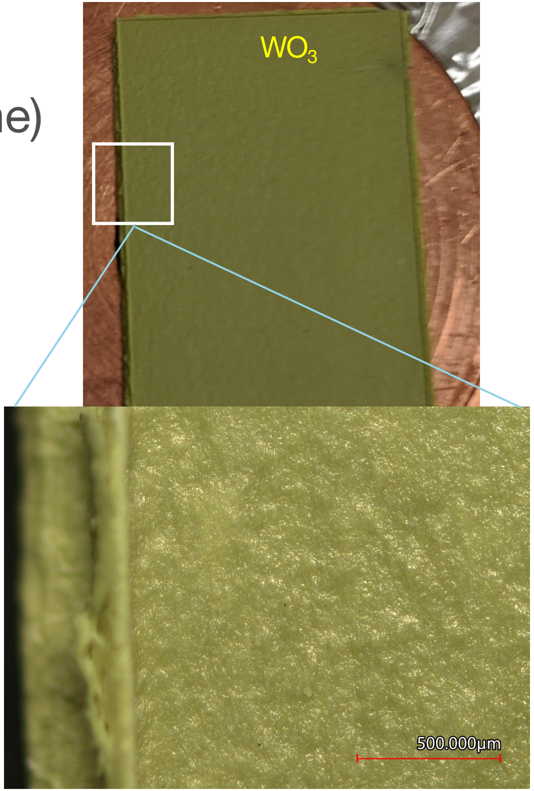
- Prolonged operation at high temperature leads to steady buildup of oxidation
- Thin-film protects
  - Tested at N_2 purged environment (O_2 level corresponding to 10^{-2} Torr)
  - Vacuum furnace in-house at Euclid
- ~ 1000 °C for 24 hours
- Naked W surface completely oxidized
- Coated surface almost unchanged
  - Edges (not well coated) still oxidized.
- SEM confirmed:





Not to scare anyone but just for fun

- Worst scenario: O_2 level as in air
- Still relatively low temperature
- 30 mins at 780 °C (alright I went too high and burned one)
- WO_3 looks shiny though

To continue…

- Lots of effort spent in Phase I
- Results *extremely* promising
- Well laid-out Phase II plan
- Demonstrated budget control
- Demonstrated project management
  - Gantt + Pert standard Euclid routine
- 8 support letters from the community
- However…
  - P2 application declined
  - One strongly opposing reviewer = dead end
  - Expected all results to be delivered in P1
  - Had good points & suggestions but did not realize that negative rating = no chance to try what's advised.

In Phase I of this project, two methods were used to produce high-emissivity coatings on tungsten coupons. The first was RF magnetron sputtering, which is the method proposer is developing for putting coatings on complex geometries. The second is plasma enhanced chemical vapor deposition, performed at the center for functional nanomaterials at BNL. The second method was used as it has been developed to the extent that well calibrated thicknesses of coatings can be applied. Models were completed to understand the nature of the coatings and their performance and the coated materials were examined with various methods. The results reveal that the proposed coating technique is promising. The proposed phase II activities build directly upon the scope of work proposed and executed in phase I. In phase II, emissivity testing systems will be improved, a new RF sputtering system will be fabricated that will allow for coating of more complicated target geometries, and further coated sample characterizations will be performed. The proposed phase II activities follow upon the executed phase I work and are the logical steps toward moving the technology forward.

Reviewer 1

This Phase II proposal clearly follows and extends the scope of work originally proposed in Phase I.

Reviewer 2

In Phase I, the authors have accomplished their goals in the areas of simulations, thin-film deposition practices and preliminary emissivity measurements even though the emissivity measurements were more preliminary than expected due to the COVID19 interruption.

The proposed work addresses a legitimate need; namely, the application of a high-emissivity coating to metallic targets to improve radiative heat transfer characteristics and also to minimize oxidation and associated target degradation. The specific example is the mu2e tungsten target, which is a very challenging design producing very high temperatures and consequent oxidation in the poor vacuum of the target chamber. However, in my opinion, the overall market need (Section 1.2 of the Phase II commercialization plan) overstates the demand for such coatings on HEP-relevant targets. There are a number of potential applications beyond mu2e, and beam dumps may benefit from such coatings, but most HEP-relevant targets do not reach temperatures at which radiative heat transfer is significant, and many targets are not subject to oxidation concerns like the tungsten target of mu2e.

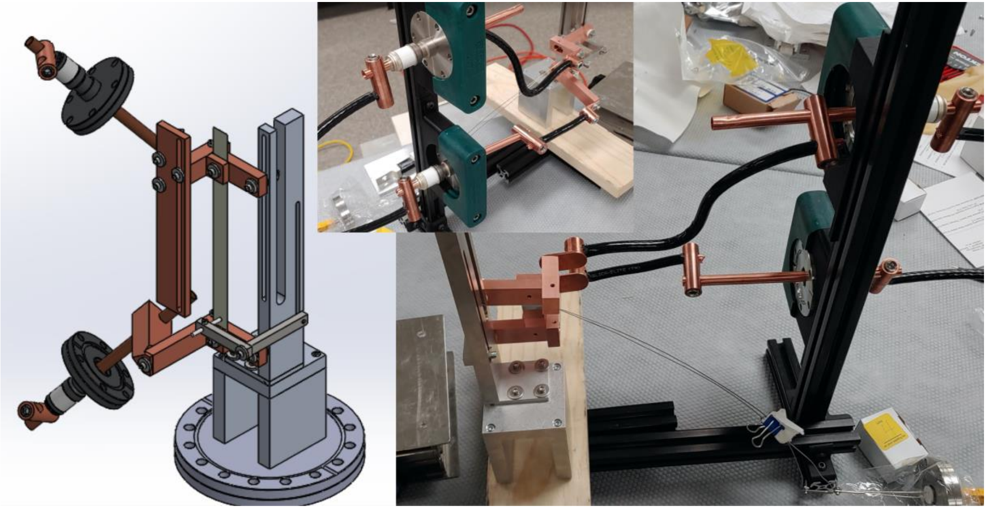
Reviewer 3

Given the lack of characterization in Phase I, the plan for Phase II is very vague about narrowing down the coating options to the optimum materials, coating process, and coating characteristics. All of this is left for Phase II, which increases the technical risk that a suitable coating will not be found. Coupled with the lack of emissivity and oxidation data from Phase I, and it almost seems like Phase II is starting from scratch.

Really, to CONTINUE…

- Program manager suggested next Phase I (submitting to “Other”)
- Odds much lower if no appropriate topic is included
- Looking for TSD support:
  - Contract for the dedicated study?
  - Other funding sources (white paper, LDRD, etc.)?
  - Other related studies?
  - Open to suggestions.

- Euclid has built a high vacuum apparatus to measure emissivity of coupons with different lengths (see photo/CAD demo)
  - Works with the spherical Paprika chamber
  - Designed to tolerate 250 A, up to 1,000 °C (need o-rings removed on flanges)



Summary

- Euclid has successfully grown thin-films of high emissivity materials on W coupons
  - Nanomaterial facilities (PECVD) and in-house (high power RF sputtering).
- Simulations were set up:
  - Thermal sim shows significant temperature improvement with higher emissivity;
  - DFT sim shows stable binding of thin-film and W substrate;
  - G4 sim shows ignorable impact on pion productivity;
  - All above can be optimized and systematically studied.
- Initial experiments are extremely promising:
  - Preliminary data shows improved emissivity from coated W;
  - Demonstrated very effective oxidation protection;
  - Three sites (Fermilab, Mizzou and Euclid) for further measurements.
- Very close to getting P2 support
  - We should **not stop** and we need your **help**.

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