



**Savannah River
National Laboratory™**

OPERATED BY SAVANNAH RIVER NUCLEAR SOLUTIONS

We put science to work.™

Effect of Various Impurities on the Hydrogen Absorption on SAES ST198

Gregg A. Morgan

David W. James

36th Tritium Focus Group – Fall 2015

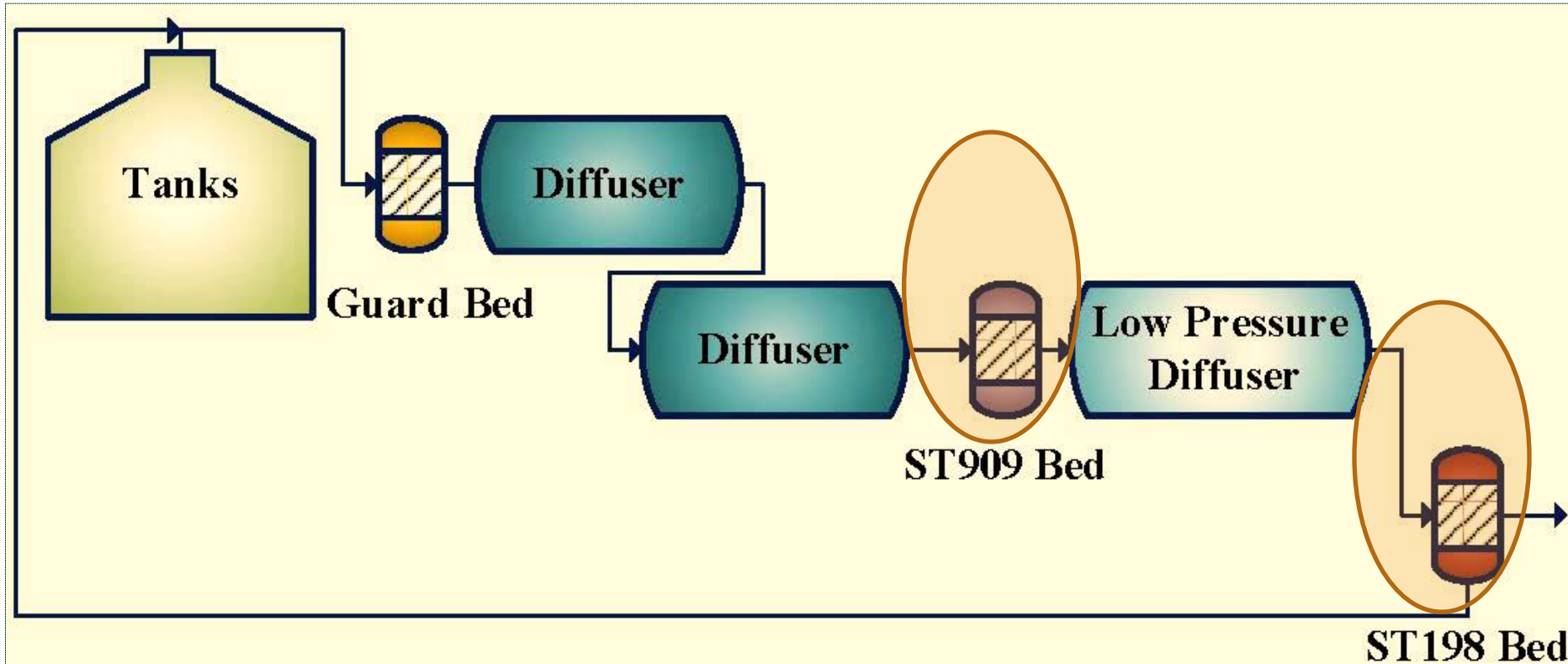
Los Alamos, NM

November, 3-5, 2015

SRNL-STI-2015-00592

- **Tritium Purification System**
- **ST909**
- **ST198**
- **Results of ST198 Impurity Testing**
- **Conclusions/Summary**

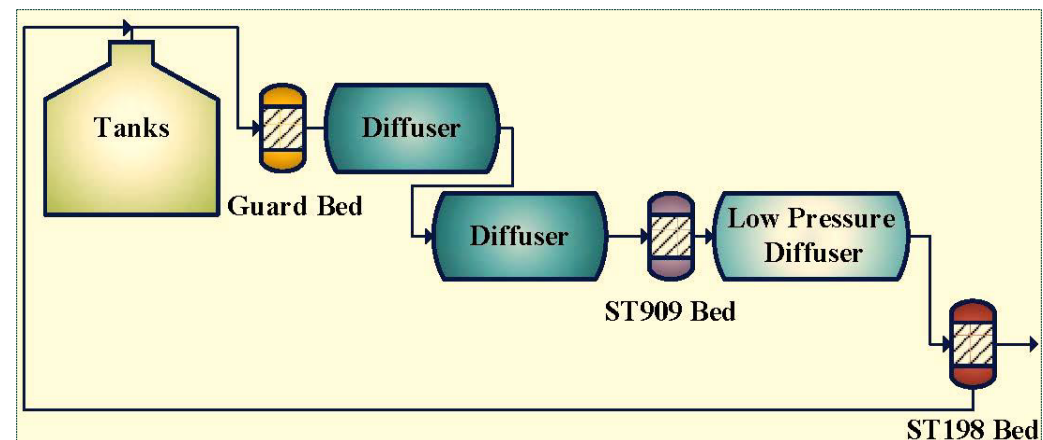
TPS Overview



- **Zr(Mn_{0.5}Fe_{0.5})₂ or Zr-Mn-Fe (40.5% Zr, 24.5% Mn, 25.0% Fe, 10% Al)**
- **Manganese and iron – catalytic active sites for decomposition**
 - CH₄
 - NH₃
 - CO
 - CO₂
 - Q₂O
- **Zirconium - active sites for the gettering of elements**
 - O
 - N
 - C
- **Literature tells us**
 - Other getter materials, may have better methane cracking performance than ST909,
 - No other single material was best for use in helium, hydrogen, or nitrogen carrier gas streams

Challenges with ST909

- **SAES ST909 is expensive**
 - (~\$4200-5300/kg, depending on the quantity of the material requested)
- **Consumable material**
- **Foreign sole source supplier**
- **Difficult to Obtain – Special Order**



What does ST909 have to do with ST198

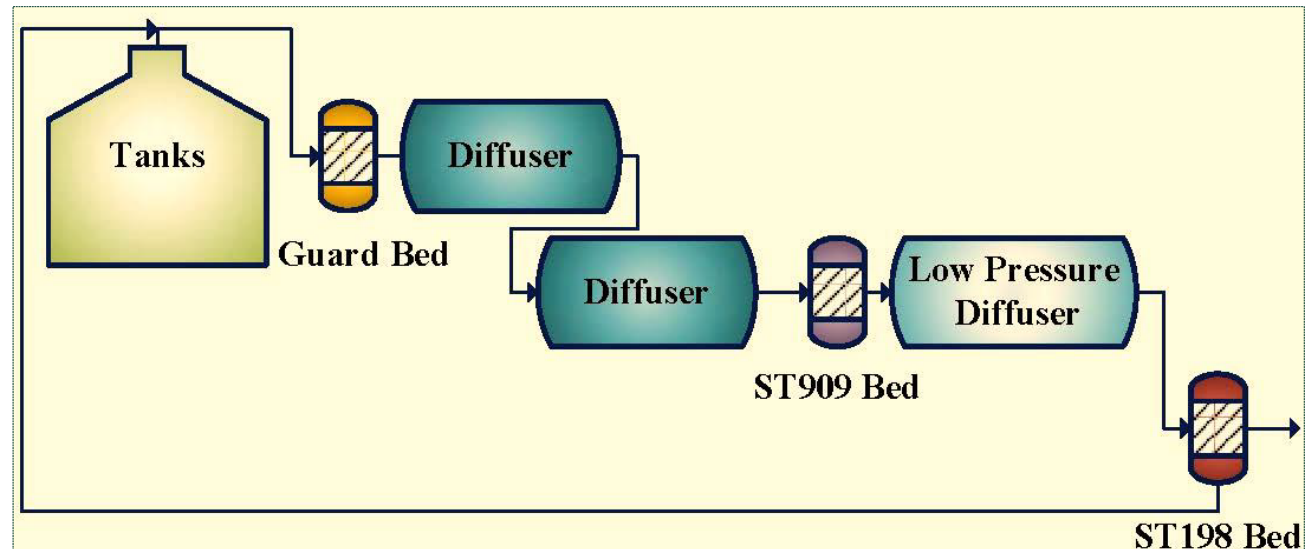
- Downstream effects if ST909 did not perform

- ST198 Impurity Testing

- Baseline Testing

- Impurity Effects

- CH₄
- CO
- NH₃



- Effect of Temperature in relationship to Impurities

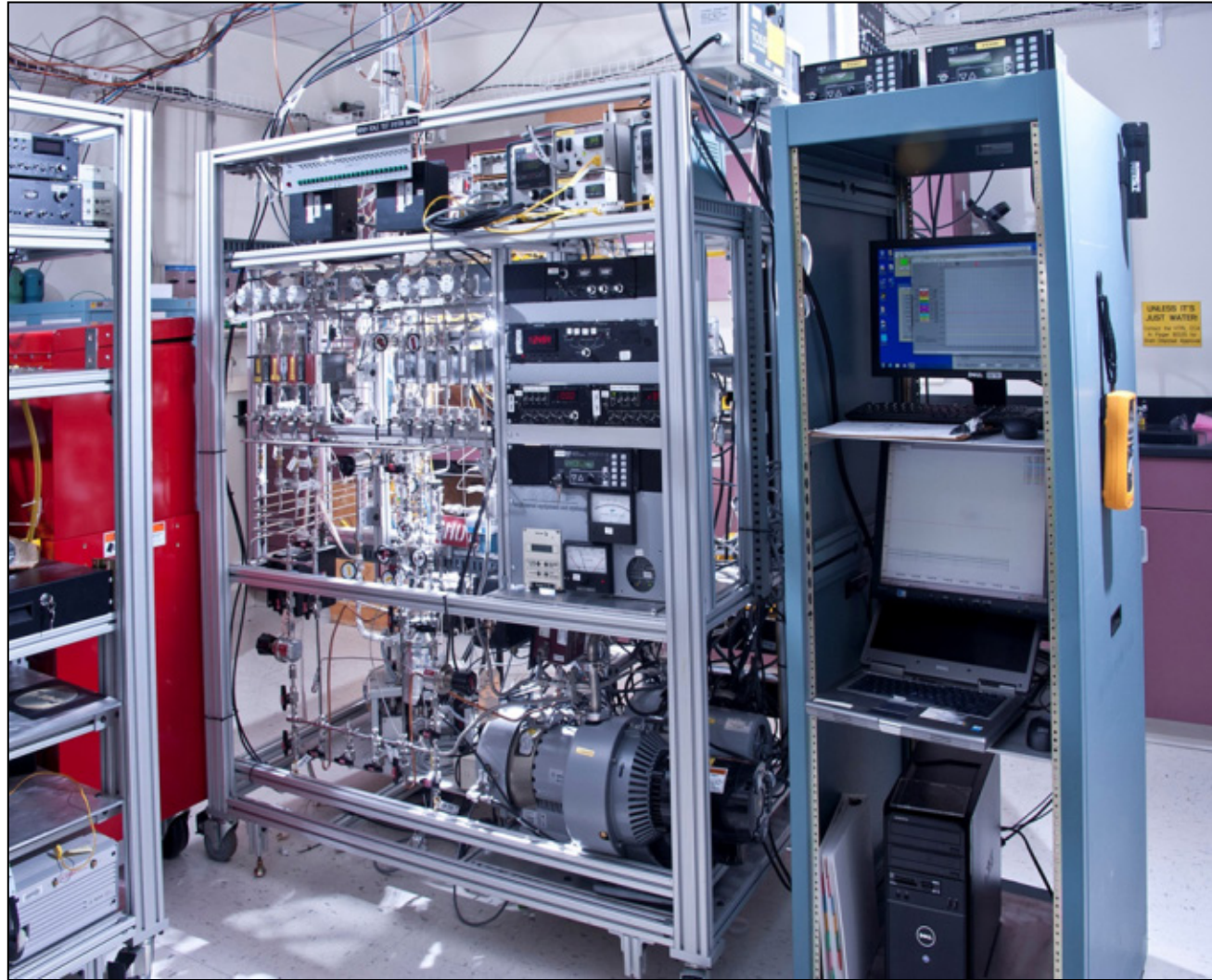
Properties and Characteristics of ST198

- **ST198 is a metal alloy getter manufactured by SAES**
 - Nominal composition of 76.5% Zr and 23.5% Fe
 - Primary phase is Zr_2Fe
- **ST198 reacts with:**
 - hydrogen isotopes, oxygen, water, and other gases
 - unreactive towards nitrogen.
- **Each pellet is roughly 0.6 grams with a density of 5.3 g/cm₃**
- **Useful for tritium removal from process gas streams with low tritium concentrations.**
 - Removes tritium without the formation of tritiated water
 - Tritium is unrecoverable

Experimental Apparatus

Experimental Test System

- Pressure transducers
- MKS Flow Controllers
- Gas supplies of Ar, N₂, and H₂
- 2 Residual Gas Analyzers
- 1 Inficon micro GC
- Flow through testing

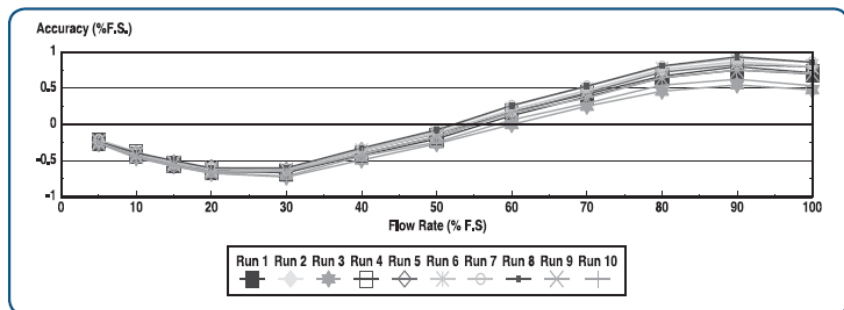


ST198 Test Matrix

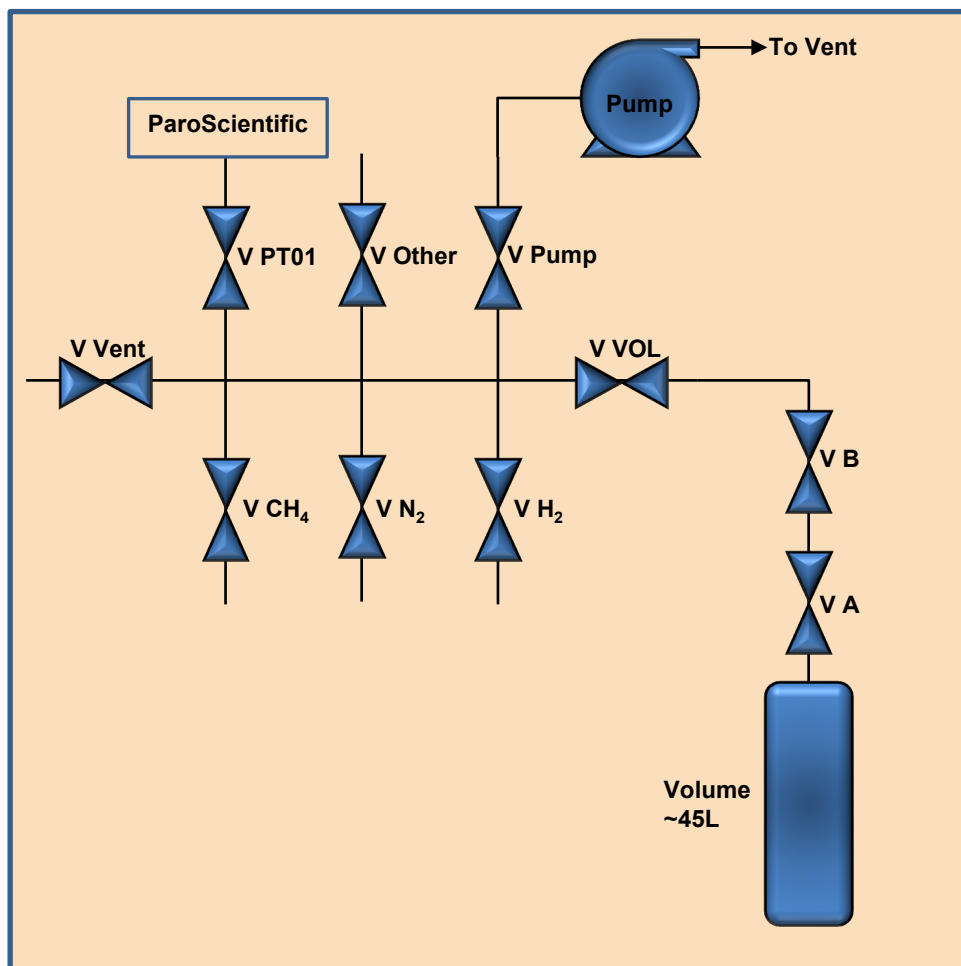
ST198 Test	Gas Constituents (percent by volume)	Test Cell Temperature (°C)
1	1% H ₂ , 99% N ₂	350
2	1% H ₂ , 99% N ₂	24
3	0.1% H ₂ , 99.9% N ₂	350
4	0.1% H ₂ , 99.9% N ₂	24
5	0.1% H ₂ , 0.1% CH ₄ , 99.8% N ₂	350
6	0.13% H ₂ , 0.1% CH ₄ , 0.1% NH ₃ , 99.67% N ₂	350
7	0.13% H ₂ , 0.1% CH ₄ , 0.1% NH ₃ , 99.67% N ₂	350
8	0.13% H ₂ , 0.1% CH ₄ , 0.1% NH ₃ , 99.67% N ₂	24
9	0.1% H ₂ , 0.1% CH ₄ , 0.05% NH ₃ , 99.75% N ₂	350
10	0.1% H ₂ , 0.1% CH ₄ , 0.05% NH ₃ , 99.75% N ₂	24
11	0.1% H ₂ , 0.1% CO, 99.8% N ₂	350
12	0.1% H ₂ , 0.1% CO, 99.8% N ₂	24
13	0.1% H ₂ , 0.1% CO, 99.8% N ₂	24
14	0.1% H ₂ , 0.1% CO, 99.8% N ₂	350
15	0.1% H ₂ , 0.1% CH ₄ , 99.8% N ₂	24

Preparation of Gas Mixtures

- Low concentration of impurities could not be reliably controlled with flow controllers at low flow rates

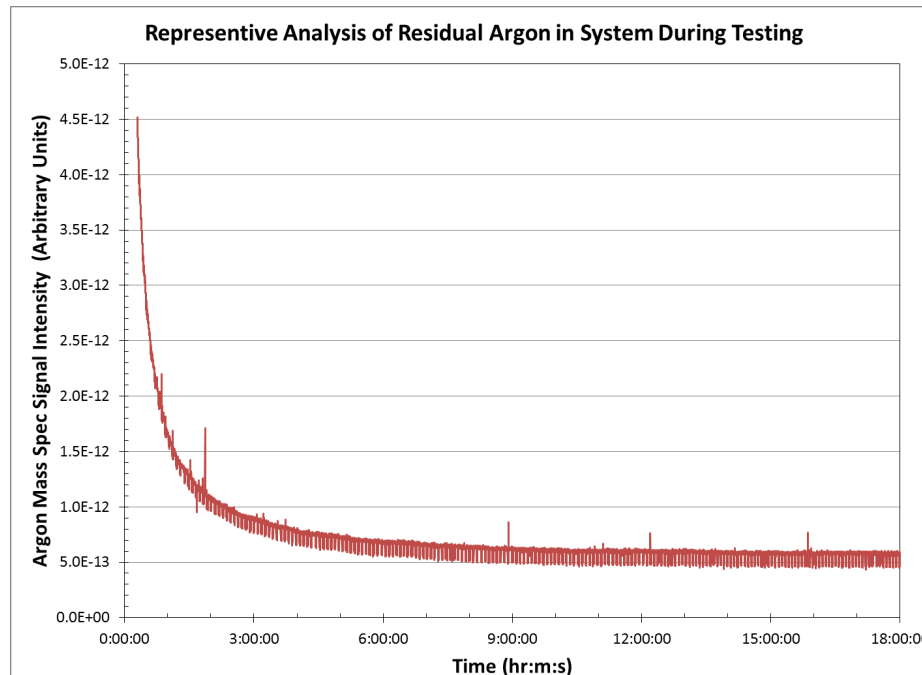


- Assembled a gas mixing manifold to prepare gas mixtures with low levels of H₂ and various impurities.



Activation Protocol

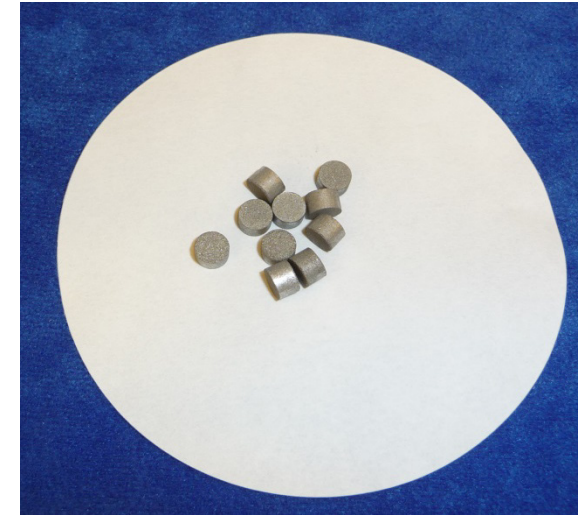
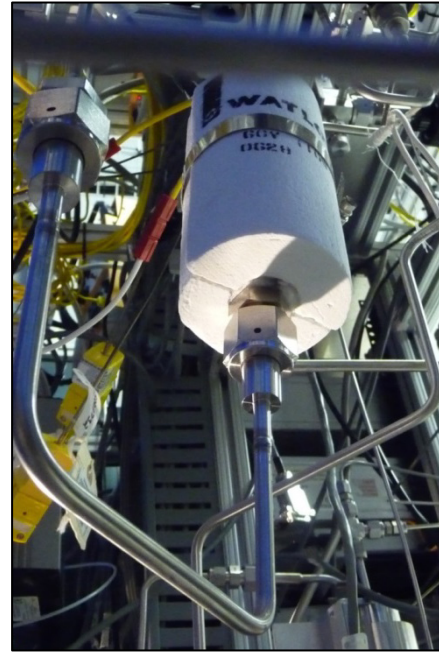
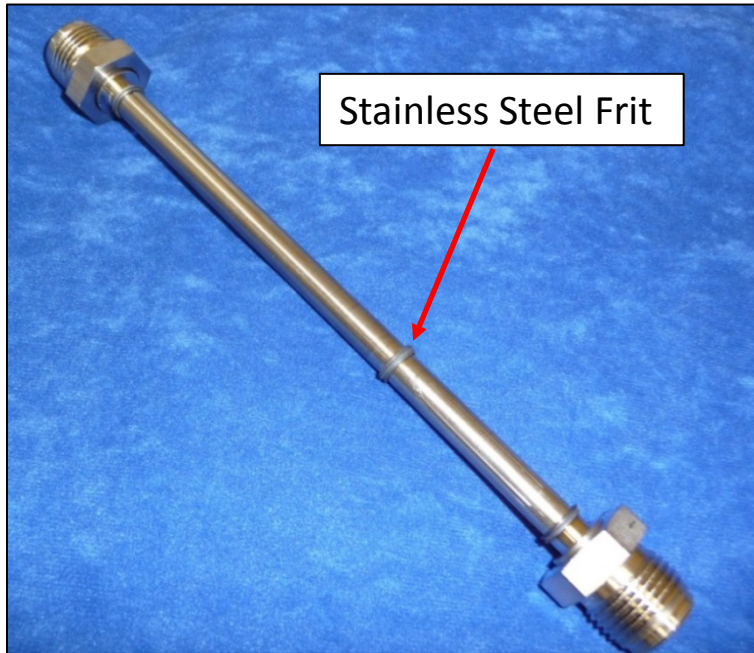
- Argon purge at ambient temperature to displace the residual air after activation
 - 30 sccm and ~1875 torr
- Nominal temperature ramp to 450°C for 4 hours
- Evacuation of the system for 1-2 hours to remove the residual argon



- Upon completion of the activation the sample temperature was lowered to 350°C or 24°C depending on the test protocol.



Test Cell Configuration

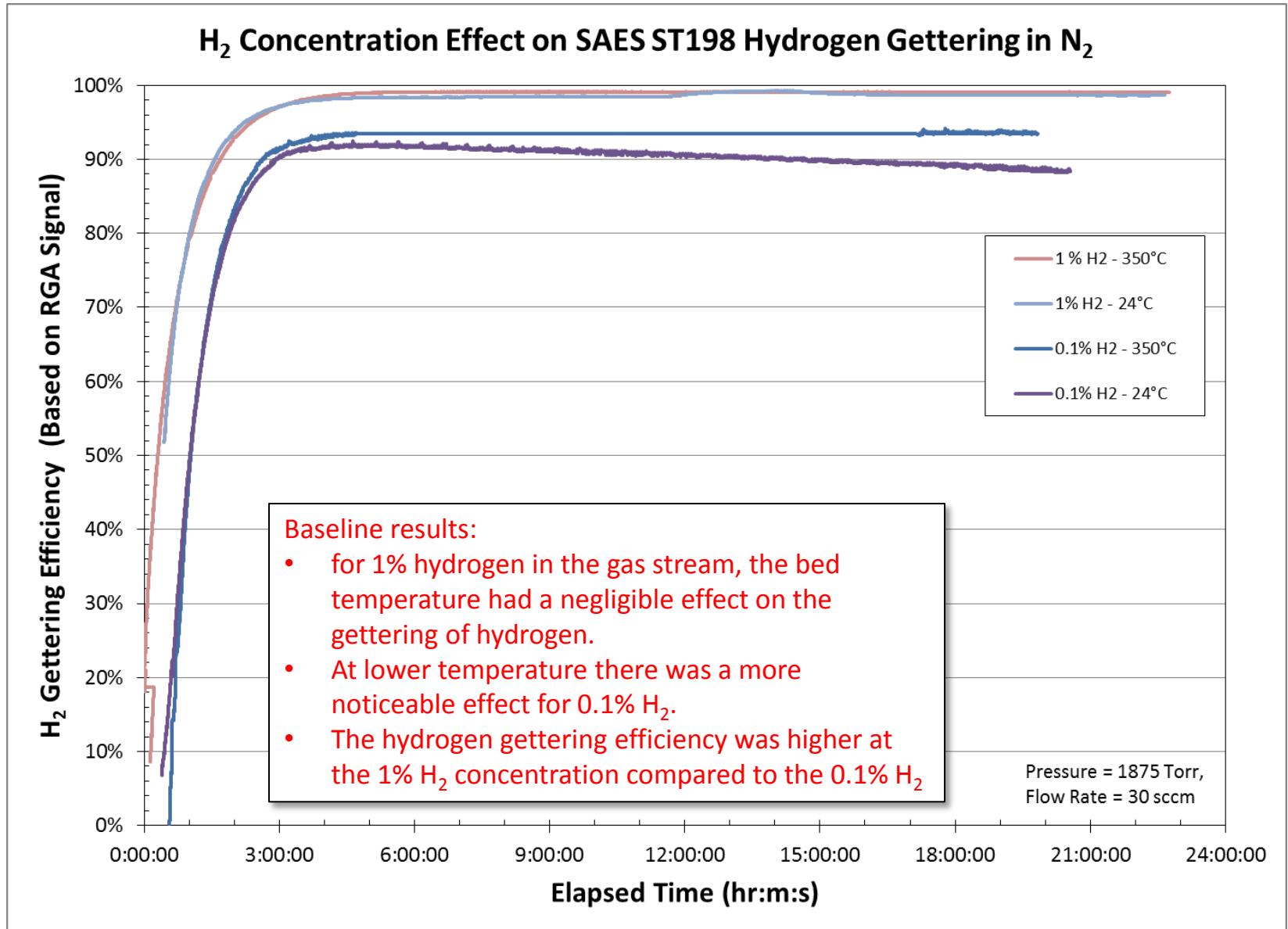


- $\frac{1}{4}$ " diameter tubing 9" long
- VCR #8 fittings
- Welded Fritted Disk up about $\frac{1}{3}$ of the way
- Vertical Configuration

- Approximately 6 grams total
- 10 Pellets
 - Right cylinder pellets
 - 6 mm diameter x 4 mm tall
- Stacked configuration



Effect of H₂ Concentration on Gettering



ST198 Decreptitation



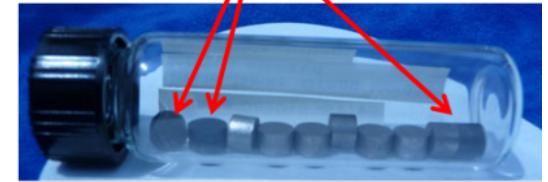
Virgin ST198 Pellets

Decreptitation



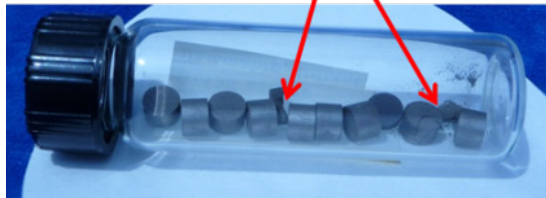
1% H₂
350°C

Decreptitation



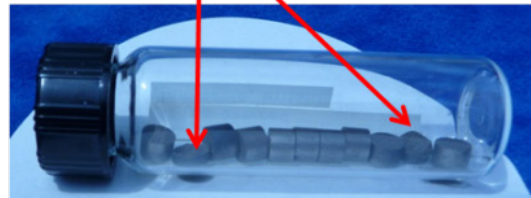
0.1% H₂
350°C

Decreptitation



0.1% H₂
24°C

Decreptitation



0.1% H₂, 0.1% CH₄
350°C

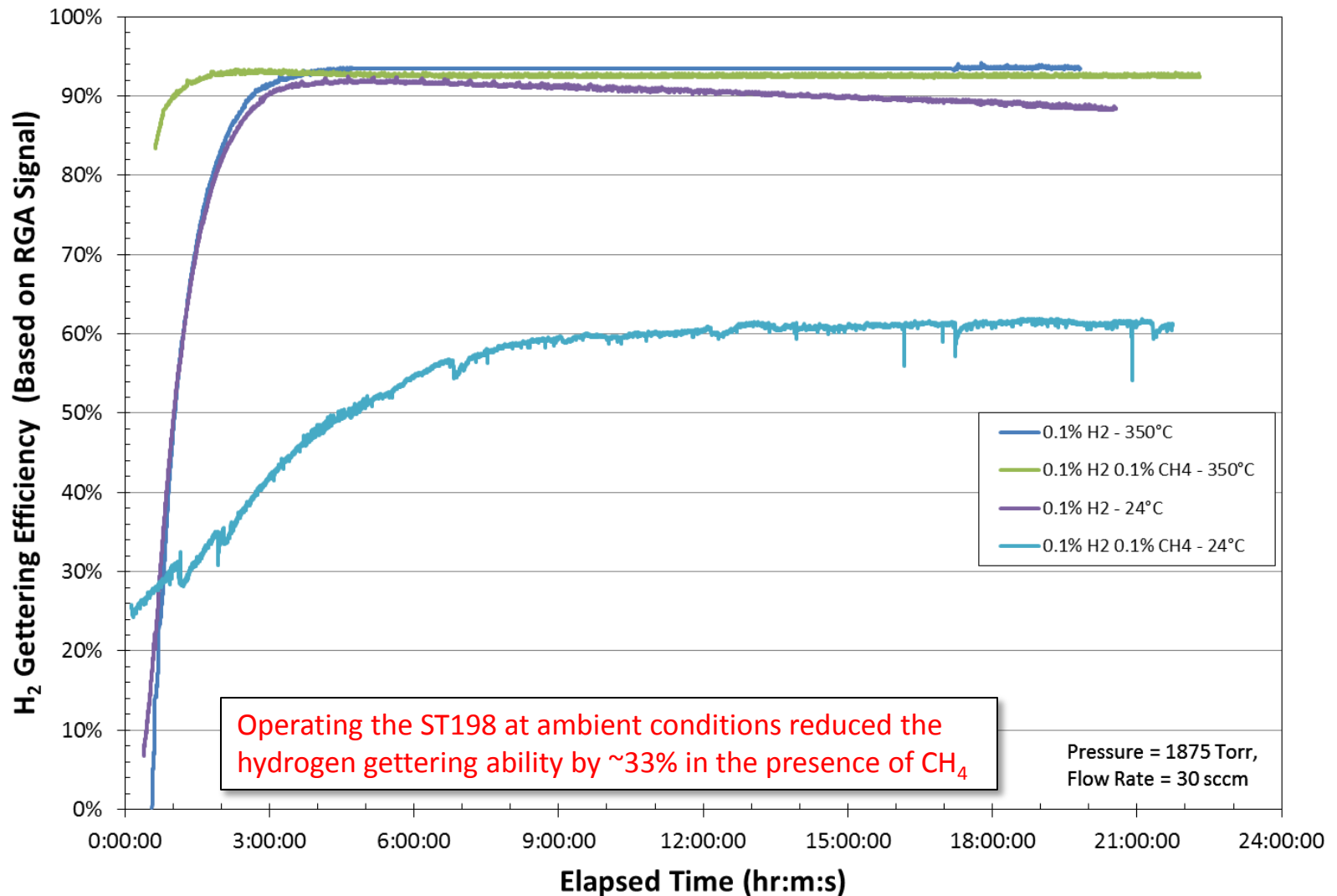


0.1% H₂, 0.1% CH₄ 0.1% NH₃
350°C

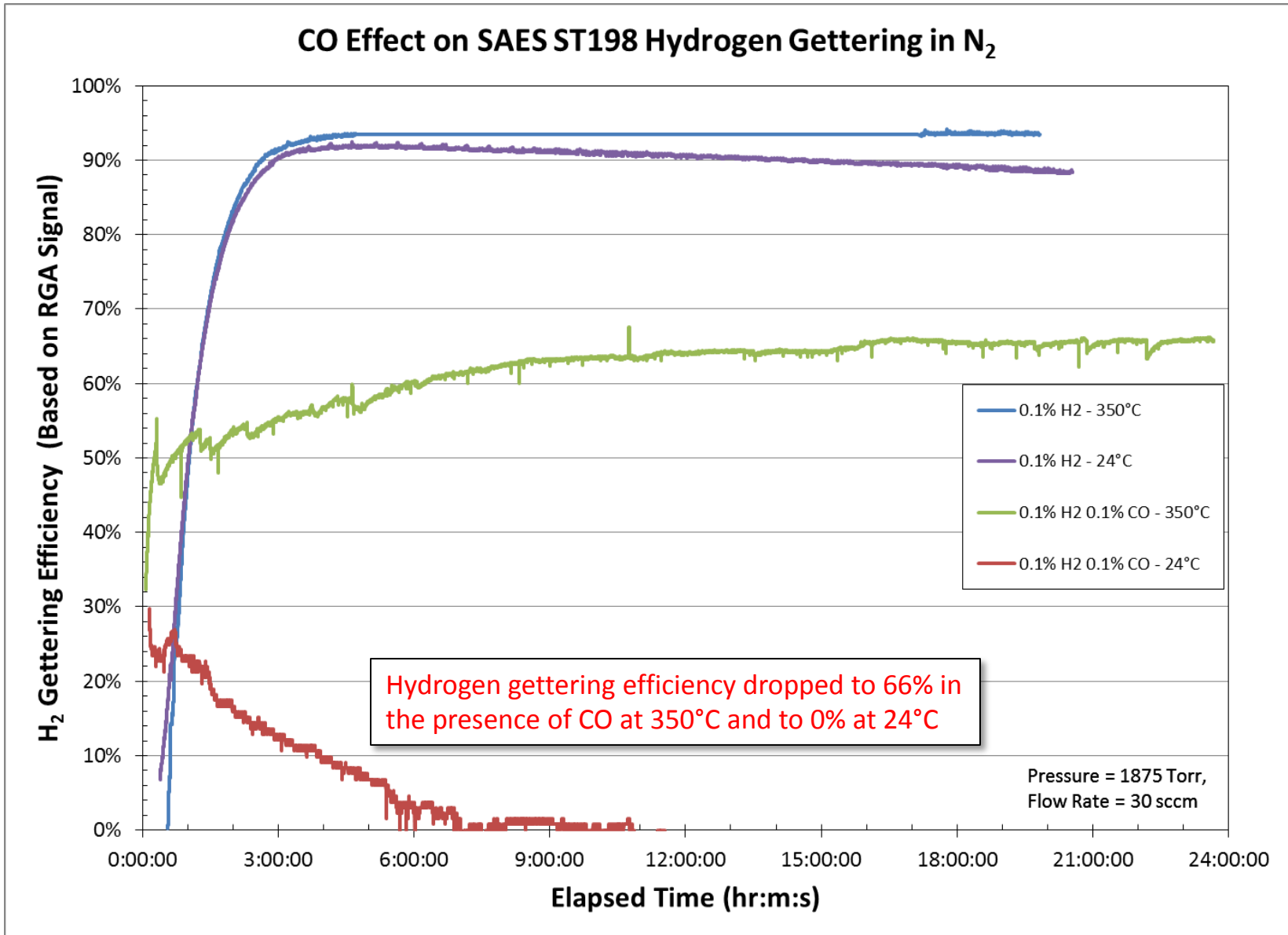


Effect of CH₄ on H₂ Gettering Ability of ST198

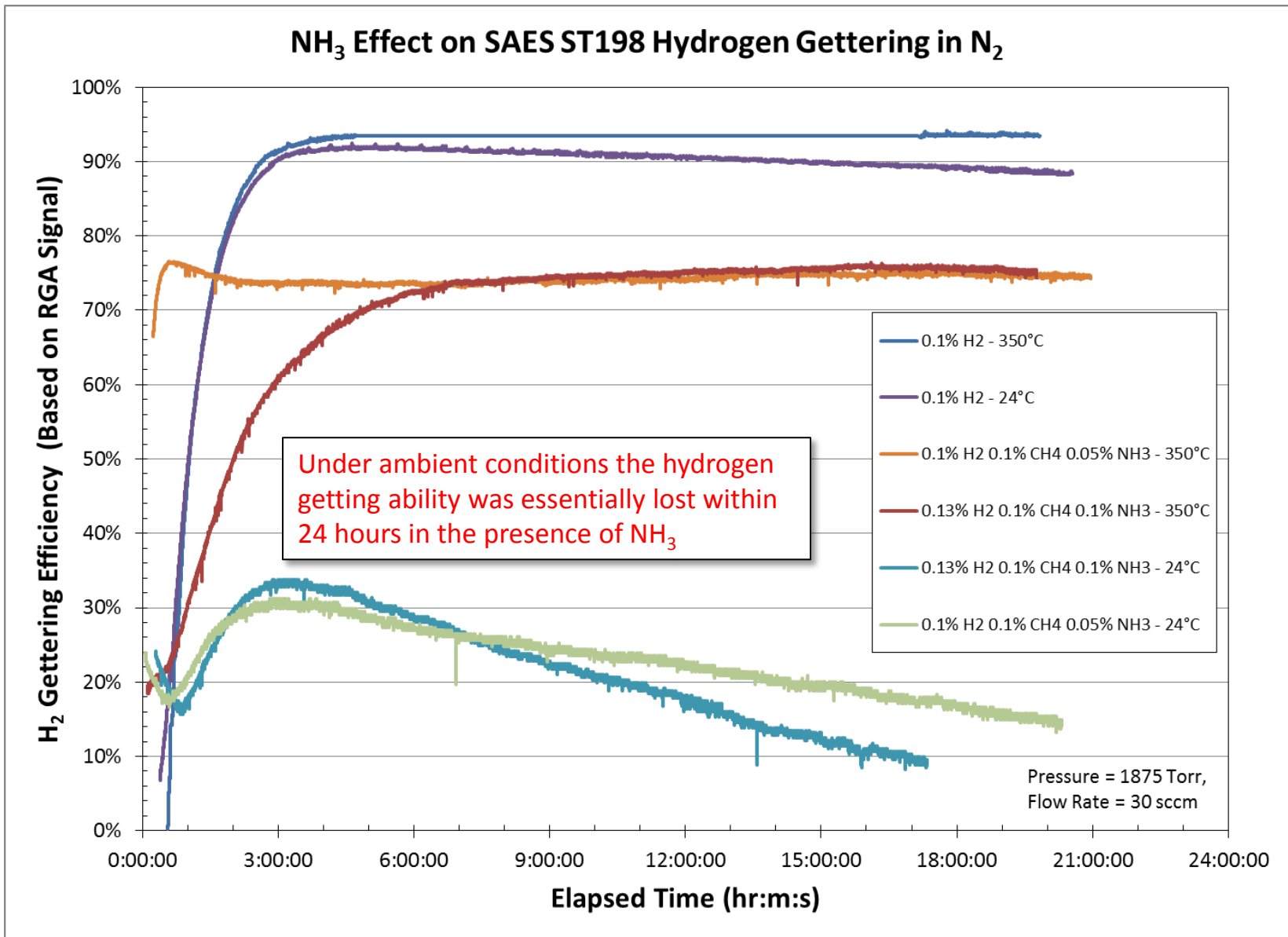
CH₄ Effect on SAES ST198 Hydrogen Gettering in N₂



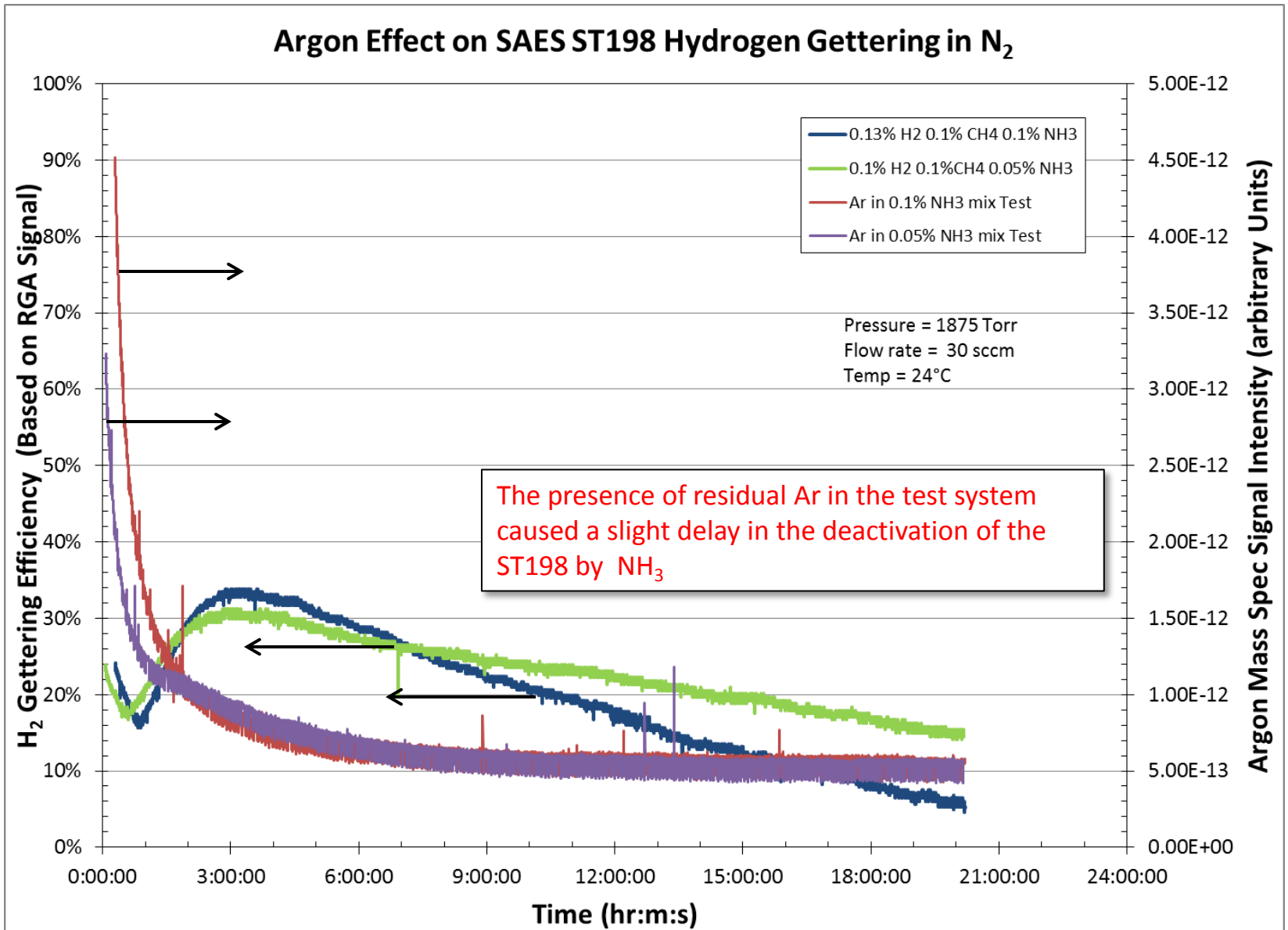
Effect of CO on H₂ Gettering Ability of ST198



Effect of NH₃ on H₂ Gettering Ability of ST198

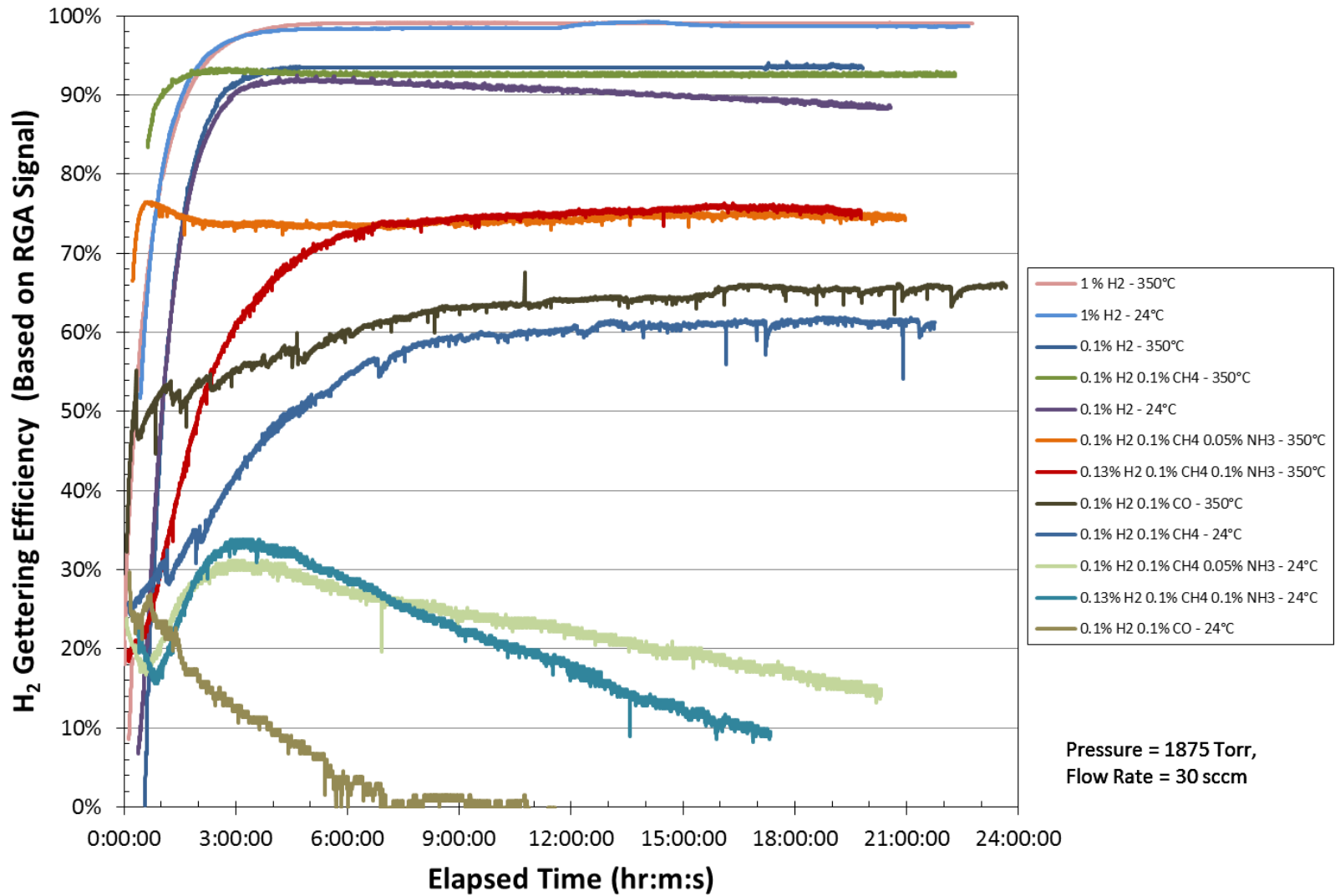


Effect of NH₃ on H₂ Gettering Ability of ST198



Alternative Getter Materials - Summary

SAES ST198 Hydrogen Gettering in N₂



Conclusions

- **1% hydrogen in the gas stream bed temperature negligible effect**
- At ambient conditions impurities compromised ST198 hydrogen gettering ability to a greater extent than at higher temperatures.
- **Experiments showed only a slight reduction of gettering ability with the addition of methane (CH₄) at 350°C.**
- At ambient temperature conditions, the hydrogen gettering efficiency was reduced by 33% in the presence of CH₄.
- **Reduction of hydrogen gettering with carbon monoxide at both temperatures.**
 - Reduction of hydrogen gettering capability dropped ~29% for the tests at 350°C (compared to baseline capabilities as related to the reduction of the RGA signal from the mass spectrometer).
 - At ambient conditions, the hydrogen gettering effectiveness was reduced to zero within 10 hours upon contact with CO.
- At ambient temperature, ammonia also reduced the hydrogen gettering efficiency but to a lesser extent than the CO.

Acknowledgements

- **Savannah River Tritium Enterprises (SRTE) Engineering and Operations, Savannah River Field Office, and National Nuclear Security Administration (NNSA) Technology Maturation Division (NA-123.2) for the Plant Directed Research and Development (PDRD) program funding of this project.**
- **The authors acknowledges the contributions of:**
 - Henry Sessions
 - Benton Randall
 - Jim Klein
 - Anita Poore

Thanks for your attention!!