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# The Effects of a Hydrogen Environment on the Lifetime of Small-Diameter Drift Chamber Anode Wires

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## *Abstract*

*Possible deterioration of anode sense wires used in a hydrogen-filled neutron detector is investigated. Wires were loaded with free weights and put into a wire detector environment. Stainless Steel, Tungsten, and Platinum wires did not break after exposure to charge equivalent to many wire lifetimes. Furthermore, exposure to hydrogen gas caused no noticeable surface degradation or change in wire yield strength.*

## **Introduction**

A momentum-transfer neutron detector is implemented using a hydrogen-filled Time Projection Chamber (TPC), a type of Multi-Wire Proportional Detector with three-dimensional particle tracking. The measured spectral and directional properties of recoiled protons provides information about incident MeV-scale neutrons. Such events indicate a fissile source's presence, location, and identity. This neutron-TPC may be an

effective and mobile way to detect, identify, and locate Special Nuclear Material for nonproliferation and inspection applications. In the process of assessing the practicality of fielding such a device the limits of the wires in the TPC are investigated over their expected operational lifetime. Because the structural integrity and surface characteristics of gain wires in a TPC are crucial to its successful operation, changes in them pose a threat to fielding a neutron-TPC. The work reported here was undertaken to determine whether the environment in a hydrogen-filled TPC adversely affects ultra-small diameter wires.

Previous TPC studies are mostly concerned with the design and operation of existing TPCs [1-3], and not with the operational lifetime of sense wires. Experiments have been conducted on the lifetime of such wires [4]; however, this study did not account for oxygen in the hydrogen gas, which can inhibit hydrogen embrittlement.

Hydrogen attack of metals is a well-documented phenomenon and the number of relevant articles is too numerous to cite. The interested reader is referred to the following seminal documents for a review of the literature [5-7], Hydrogen can be introduced into the metal during solidification, electrolytically or upon exposure to a gaseous environment. For high strength steels and other metals with body-centered crystal structures, hydrogen attack can take the form of sustained crack growth. A crack-like defect and applied stress are required for crack growth [8], [9]. For austenitic alloys, hydrogen attack is manifested by somewhat reduced yield and ultimate tensile strengths and reduced ductility due to exposure to hydrogen during a tensile test. Materials that form stable

hydrides like Titanium and Zirconium alloys usually display a sustained crack growth due to the formation of brittle hydrides in the crack path. In this case of stable hydride formers, the cause of the embrittlement appears clear. In all other systems, the exact mechanism of hydrogen attack has yet to be determined.

Nearly all previous hydrogen embrittlement studies are concerned with samples whose diameters are orders of magnitude larger than the diameter of wires used in a TPC.

Notable exceptions are the two studies previously published on hydrogen-filled TPC's [1], [4]; however, these studies are more focused on the overall operation of the TPC rather than the survivability of the TPC gain wires. Some work has been reported on thin Pd [10], and thin Fe [11] foils where enhanced plasticity was observed. The most relevant work to this study was reported by Devine [12] on small diameter tungsten wire wherein he reported no effect on the wire unless a crack-like defect was introduced into the wire prior to testing under mechanical stress and exposure to hydrogen. However, even Devine's wire is an order of magnitude larger in diameter than the wires required for inducing gas gain in TPC's.

This research seeks to determine if a hydrogen rich environment caused deterioration of the sense wires. For the work reported here, three experiments were run: two in clean hydrogen and a control in helium both at a pressure of one atmosphere. In each run, seven types of wire shown in Table 1 **Error! Reference source not found.** were tested for the duration of one week.

## Experiment

The experimental setup, demonstrated in Figure 1, exposed seven wires to a radial electric field in hydrogen gas, simulating the environment found in a TPC. Oxygen levels and individual wire currents were monitored, and the voltage was adjusted to deposit at least 0.2 Coulombs of charge per centimeter on each wire over the duration of the run. In simulating the TPC environment, no quench gas was used because this experiment dealt only with wire deterioration due to hydrogen embrittlement. The effects of quench gasses on wire lifetime were assumed minimal and not investigated.

A Honeycomb Metal Cathode (HMC) consists of seven, 15.2cm stainless steel tubes with a 1.6cm outer diameter clustered into the shape of a hexagon. A wire was hung vertically down each of these tubes, individually glued with Torr-Seal<sup>®</sup> [13] to a Teflon cap, and loaded with a tungsten-alloy weight. The tungsten weights loaded the wires to 70% of their yield strength. Uncoated stainless steel 316L and platinum wires were only loaded to 30% of their yield strength because they were incapable of supporting more weight.

The absence of oxygen is essential to TPC functionality [2]. Additionally, literature indicates that the presence of oxygen can inhibit hydrogen embrittlement [12]. To ensure hydrogen purity throughout the duration of the experiment, a closed loop oxygen sensing and filtration system was implemented [2]. With the wires loaded, the HMC was sealed into the test chamber. The test chamber was constructed of stainless steel vacuum chamber ConFlat<sup>®</sup> flange components. From the main chamber, stainless steel tubing connected an Illinois Instruments model 911 oxygen sensor [14], which continually

sampled the gas at 150 cc/min and measured its oxygen concentration. Prior to feeding the sampled gas back into the main chamber, the gas was fed through an Agilent® oxygen trap [15] to remove any remaining oxygen such that throughout the duration of the experiment the test wires were exposed to hydrogen with no more than 10 ppm of O<sub>2</sub> present (see Figure 2).

To create a radial electric field around the sense wires, the HMC was negatively biased while the test wires were held at ground. The test wires were individually held at ground through a Keithley current-multiplexing picoammeter [16]. Wire currents, oxygen levels, temperature, and bias voltages were regularly monitored and recorded by a custom LabVIEW™ Virtual Instrument [17].

Measurements of the wire's yield strengths and Scanning Electron Microscope (SEM) images were used to study the wire characteristics before and after exposure to hydrogen and helium.

It was calculated that hydrogen completely diffuses through pure tungsten and stainless steel wires in 13 minutes and 3 minutes respectively. This calculation was performed using the approximate diffusion constants  $D = 7.3 \times 10^{-10} \text{ cm}^2/\text{sec}$  for pure tungsten [18], and  $D = 4 \times 10^{-10} \text{ cm}^2/\text{sec}$  for stainless steel [6], at room temperature. Because hydrogen diffuses entirely in minutes, experiments lasted seven days to allow substantial amounts of charge to deposit on the wires.

## Data

To deposit at least 0.2 – 1.0 coulombs of charge per centimeter on each test wire, the HMC bias voltage was adjusted for each run. The voltage was held constant for the first and second hydrogen runs at -1850V and -1900V respectively. In the helium run, however, the voltage had to be readjusted from -800V to -2000V to ensure proper current deposit due to falloff of current on the wires over time. Figure 2 shows the magnitude of current measurements from the second hydrogen experiments. We believe the peak in current during the beginning of the experimental run was caused by decreasing oxygen levels due to the continual filtration system. Once the oxygen concentration was very low, the current measured on the wires decreased exponentially throughout the course of each experimental run. The time required for the current to decay to zero in a helium environment was approximately one day, while in hydrogen it took approximately one week.

The integrated current, or charge, per length of wire for all three experiment runs is given in Table 2. The charge per length varied from wire to wire by 0.2 to 1.8 coulombs per centimeter. Platinum and gold-coated stainless steel 316L were not tested during the initial hydrogen run. The smallest-diameter wires, uncoated stainless steel 316L and pure tungsten, had the greatest charge deposited in both hydrogen runs. However, in a helium environment, the current on these wires behaved erratically throughout the run.



## **Analysis**

Preparation of the wires involved cutting each 15 cm sample into three large pieces (approx. 4.5 cm) and one shorter piece (approx. 1.5 cm). Each end of the large pieces was glued using Torr-Seal® to two pieces of sheet metal for tensile testing. The shorter piece was analyzed in the SEM.

### *Yield Strength Characterization*

The wire's mechanical properties were evaluated by measuring the yield strength before and after exposure. The samples ranged in length from approximately twenty to forty millimeters and were pulled axially at a constant rate of 0.05 mm/sec. Yield tensile strength (YTS) was determined using the 0.2% offset method. A summary of the findings can be seen in Table 3 which lists the average YTS for each wire material pre-exposure, after exposure to hydrogen, and after exposure to helium. Small sample sizes limit statistical conclusions, but there is no evidence either gas changes wire strength.

### *Surface Characterization*

SEM pictures were taken of unexposed, hydrogen-exposed, and helium-exposed wires after each test (Figure 3). The surfaces of the unexposed wires, particularly the copper-coated and gold-coated wires, were smooth with small markings possibly induced during fabrication or by handling the wires in preparation for the SEM. The surfaces of the wires exposed to hydrogen did not significantly vary from the surfaces of unexposed wires. More serious variations exist in the surfaces of the control wires exposed to helium.

## **Conclusions**

It has been demonstrated that a one-atmosphere clean hydrogen environment does not significantly affect the lifetime or mechanical properties of anode sense wires in a TPC. The charge deposited on all the wires was several times larger than would be deposited on any wire in a TPC. Although hydrogen diffuses through small-diameter metallic wire within a matter of minutes, each wire survived the one-week tests without breaking. Additionally, the yield strengths of the wires were not reduced by exposure to hydrogen or helium. When examined with an SEM, the wires exposed to hydrogen displayed the same changes in surface characteristics as the wires exposed to helium. Hydrogen gas alone will not significantly deteriorate anode sense wires in a field-deployed TPC. In the future, the operational gain regime should be established with a radioactive source, and the results should be confirmed over a longer experimental run.

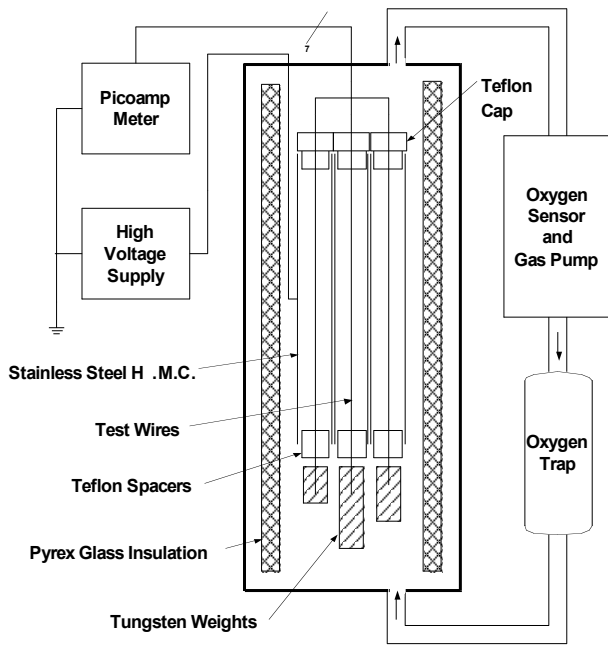
## **References**

- [1] E.M. Maev, et al., Nucl. Instr. and Meth. A 478 (2002) 158-162.
- [2] T.J. Chapin, et al., Nucl. Instr. and Meth. A 197 (1982) 305-315.
- [3] J. Huth and D. Nygren, Nucl. Instr. and Meth. A 241 (1985) 375-386.
- [4] E.M. Maev, et al., Nucl. Instr. and Meth. A 515 (2003) 288-291.
- [5] Stress Corrosion Cracking and Hydrogen Embrittlement of Iron Base Alloys, NACE, Houston, Texas, 1977.

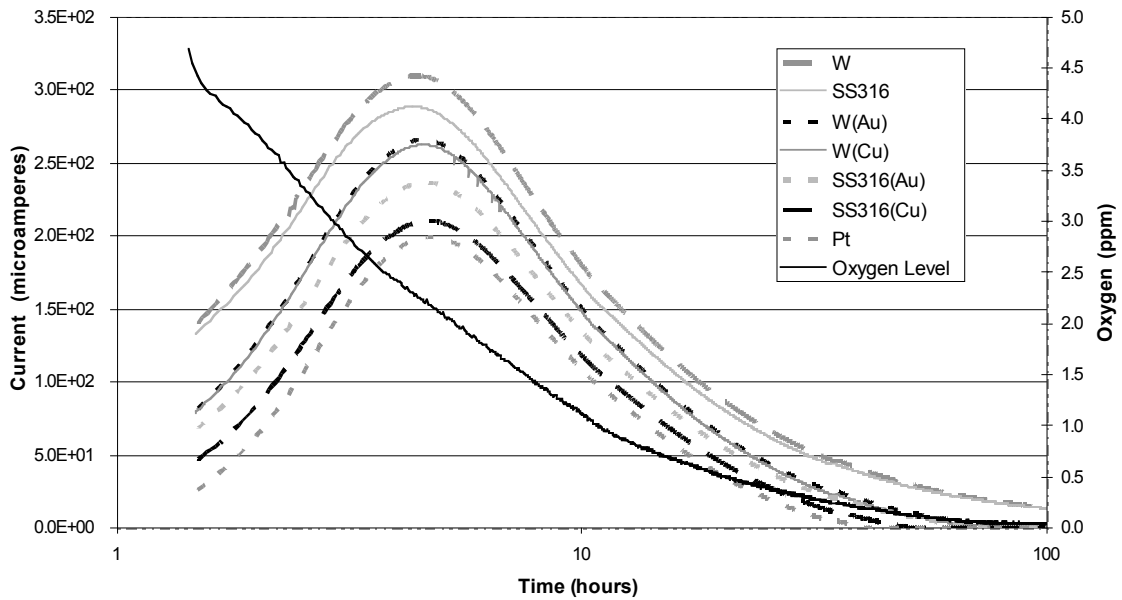
- [6] Hydrogen in Metals I & II, Topics in Applied Physics, G. Alefeld and J. Volkl, Ed., Springer-Verlag, New York, 1978
- [7] J. P. Hirth, Met Trans. 11A (1980) 861-890
- [8] A. R. Troiano, Trans of the ASM. 52 (1960) 151-177
- [9] P. McIntyre, Stress Corrosion Cracking and Hydrogen Embrittlement of Iron Base Alloys, NACE, Houston, Texas, 1977, 788-797
- [10] R. Kirchheim, Acta Metallurgica. 29 (1981) 835, 843.
- [11] T. Matsumoto, J. Eastman and H. K. Birnbaum, Scripta Met. 15 (1981) 1033-1037
- [12] T.M. Devine, Scripta Met. 10 (1976) 447-450.
- [13] Varian, Inc. 3130 Hansen Way, Palo Alto, CA 94304 USA. Tel: 650.213.8000 .
- [14] Illinois Instruments, Inc. 2401 Hiller Ridge Rd., Johnsburg, IL 60050 USA. Tel: 815.344.6212.
- [15] Agilent Technologies, Inc. 395 Page Mill Rd., Palo Alto, CA 94306 USA. Tel: 877.424.4536.
- [16] Keithley Instruments, Inc. 28775 Aurora Rd., Cleveland, OH 44139 USA. Tel: 440.248.0400.
- [17] National Instruments Corp. 11500 N. Mopac Expwy., Austin, TX 78759 USA.
- [18] R. Frauenfelder, J. Vac. Sci. Technol. 6 (1998) 388-397.

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**Figures:**



**Figure 1: Experimental setup showing the HMC, wires, gas and electrical systems**



**Figure 2: Current deposited on each wire over time with oxygen levels shown.**

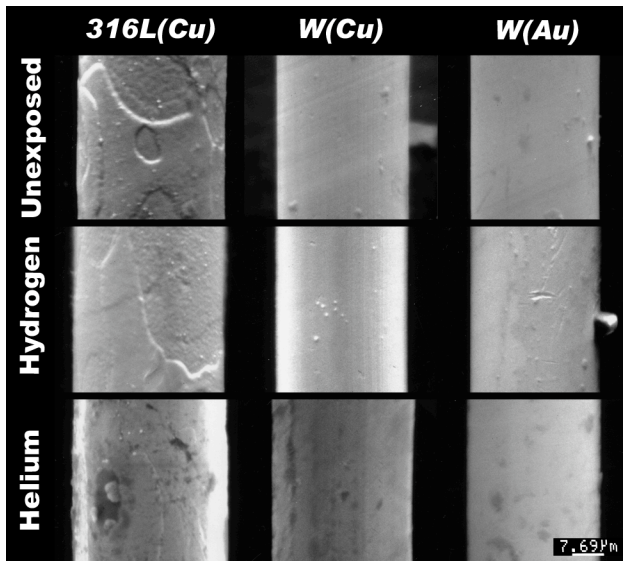


Figure 3: SEM images of pre and post exposed wires. Note, the scale applies to all wires.

Table 1: Wire types and diameters

Wire Type	Diameter ( $\mu\text{m}$ )
Platinum	43
Stainless Steel 316L	25
Gold Coated Stainless Steel 316L	25
Copper Coated Stainless Steel 316L	20
Tungsten	20
Gold Coated Tungsten	30
Copper Coated Tungsten	25

Table 2: Charge per unit length (Coulombs/centimeter) deposited on wires from each experiment

Run	Pt	W(Cu)	W(Au)	W	SS316L	SS316L(Cu)	SS316L(Au)
Hydrogen I	N/A	0.3	0.5	1.9	1.8	0.2	N/A
Hydrogen II	0.6	0.9	1.0	1.7	1.6	0.7	0.8
Helium	0.4	0.2	0.4	0.4	0.8	0.9	0.3

**Table 3: Average yield tensile strengths pre- and post-exposure  $\pm$  standard deviations with sample size in parenthesis for each wire and gas type**

	<b>Average YTS (MPa) <math>\pm</math> Standard Deviation</b>		
	<b>Pre Exposure</b>	<b>Hydrogen</b>	<b>Helium</b>
Platinum	287 $\pm$ 5 (6)	284 $\pm$ 6 (3)	278 $\pm$ 8 (3)
W_Cu	2162 $\pm$ 49 (3)	2146 $\pm$ 88 (5)	2067 $\pm$ 258 (3)
W_Au	2093 $\pm$ 17 (4)	2109 $\pm$ 53 (8)	2053 $\pm$ 30 (3)
W	2964 $\pm$ 64 (8)	3168 $\pm$ 94 (7)	3251 $\pm$ 191 (3)
SS	847 $\pm$ 16 (3)	823 $\pm$ 30 (5)	839 $\pm$ 29 (3)
SS_Cu	1873 $\pm$ 89 (10)	1921 $\pm$ 116 (7)	1978 $\pm$ N/A (1)
SS_Au	1349 $\pm$ 28 (4)	1430 $\pm$ 74 (7)	1238 $\pm$ 99 (2)