A Radiatively Cooled ADS Beam Window

Caroline F. Mallary

Advisor: Germano S. Iannachione

Physics MQP

April 26, 2007

Abstract

The purpose of this MQP is to examine candidate materials for the beam window of an ADS reactor. ADS stands for Accelerator Driven System, a means of spallating atomic nuclei to produce controlled nuclear fission, or to transmute existing nuclear waste into species that are easier to handle. Due to the high power necessary for ADS accelerators, the integrity of the accelerator's beam window has been a concern. Progress has been made on liquid-metal cooled ADS designs, especially in the ORNL and JAERI research centers, but problems have become apparent, too. My project was to investigate potential materials for an ADS beam window that can radiatively cool itself.

Section 0: Introduction

0.1 What is ADS?

Sustainable energy production has become an increasingly important issue in the world today. The burning of fossil fuels has many environmental and political consequences, and alternatives to them are being sought. While renewable sources such as wind and wave power deserve attention, nuclear power is the only emission-less option that is independent of location and environment. Traditional fission plants are not favored by many people because of perceived problems with safety, and the long isolation time required for reactor waste. ADS is a nuclear option that addresses both these issues.

ADS stands for Accelerator Driven System. In ADS, an accelerator is used to produce some of the neutrons necessary for nuclear fission. Because these neutrons are not produced by chain reaction, their production can be stopped at any time: No active intervention is necessary to prevent them from being created. What this means is that the chain reactions in an ADS core can never get out of hand, because they depend on the accelerator to sustain them. An ADS core is sub-critical, and it can't run on its own. This addresses one of the biggest popular concerns about nuclear power. An ADS reactor will never suffer accidental meltdown.

Because ADS can run on a sub-critical core, it can burn materials that are not currently fissionable. Namely, it can burn the waste of other reactors (Fig. 1). There is a great deal of energy stored in nuclear waste (Fig. 2). It is this energy that makes it so damaging to the environment. ADS causes some of the energy stored in minor actinides and long-lived fission products (MAs and LLFPs) to be released immediately. It can then be capture in a power-generating plant, instead of a mountain. It is believed that 95% of MAs and LLFPs can be transmuted, and that an ADS could process 250 kg of nuclear waste every 300 days. ADS could reduce the geological storage time for nuclear waste from millions of years to less than 500 years[1]. This lifetime is feasible for a geological storage facility: Architects have been building structures that last this long for millennia.
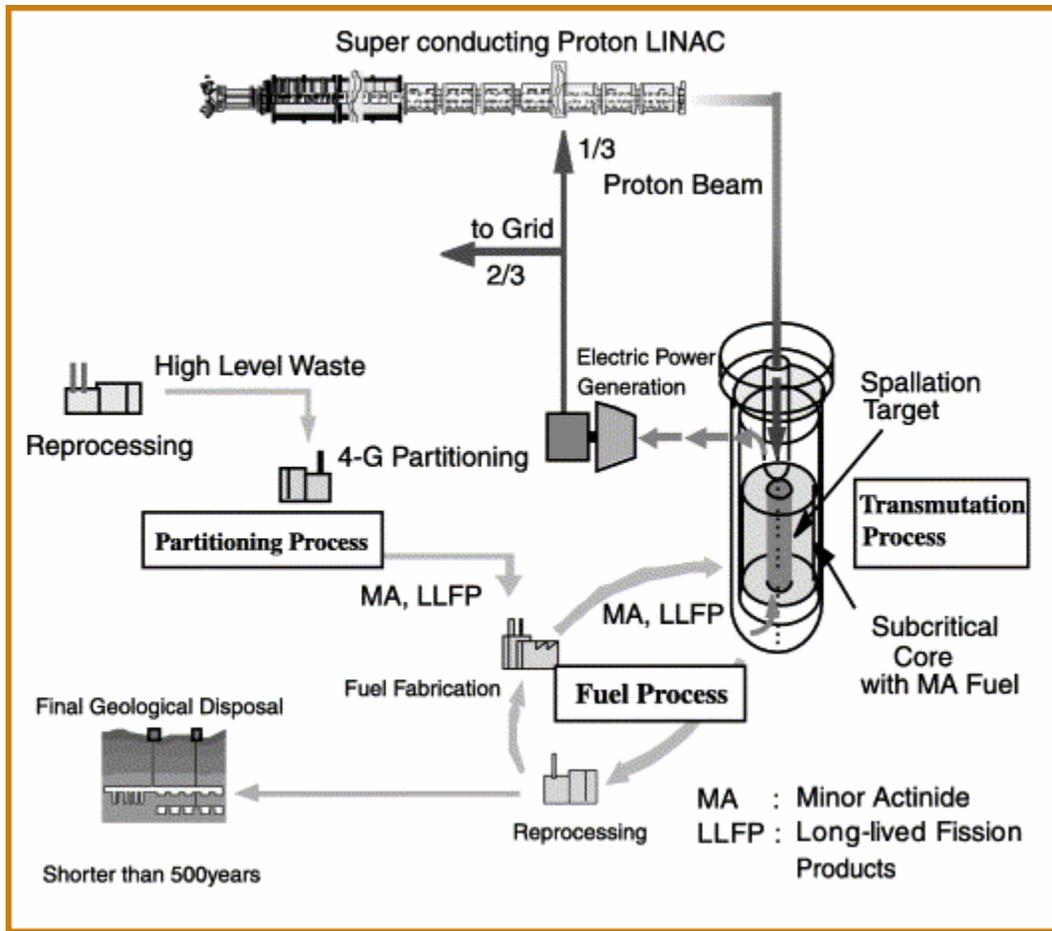
Figure 1. Concept of a Power & Transmutation system for long-lived radioactive nuclides by JAERI. From Y. Kurata, T. Takizuka, T. Osugi, H. Takano, *JNM* **301**, 1, (2002)

The mechanism by which the accelerator produces the needed neutrons is called spallation. Ions, usually protons, are accelerated to extremely high energies (~1 GeV) and aimed at a "target". The target material may be chosen on several different criteria, but the most important is having a large number of neutrons. Spallation is a fancy word for smashing the target nuclei to bits. When the nuclei are smashed, dozens of neutrons may be freed and continue on into the core. They must be moderated first, as some will have very high energies. However, it is not necessary to thermalize them. An ADS would most likely be fast-neutron reactor, because fast neutrons are not as easily absorbed as thermal ones. This would ensure that more waste was burned than created.
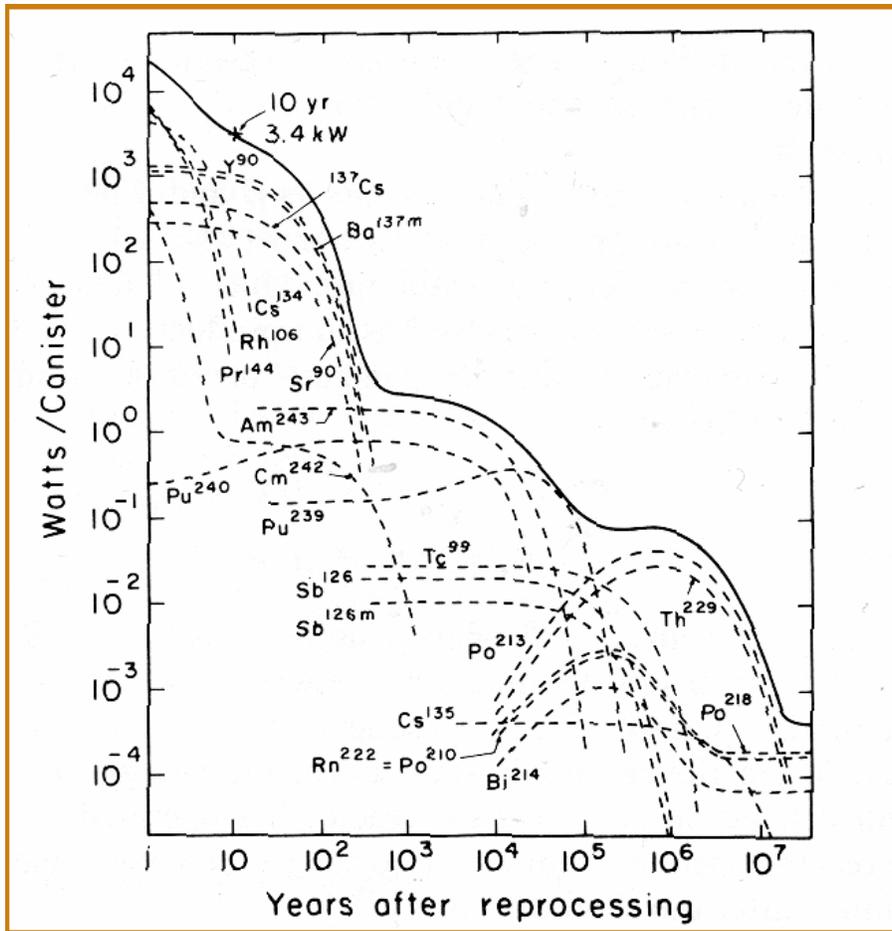
Figure 2. Radioactive power from decay of fission products and actinides. This decay-power results from the waste of 1 mo. of operation of a 1000-MW power plant. Solid curve is the sum of contributions of individual isotopes. From B.L. Cohen, *Rev. Mod. Phys* **49**, 1 (1977) via [2].
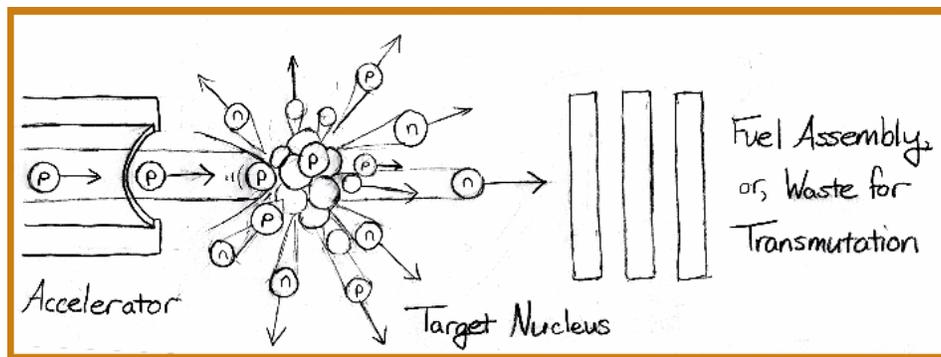

Figure 3. Spallation. Hand-drawn.

Interest is ADS has been building quietly for a long time. There are two facilities already built which focus on ADS. Both Oak Ridge Nat'l Laboratory (ORNL), and J-PARC in Japan have beams online for this purpose. Oak Ridge's Spallation Neutron Source (SNS) has been operating since June 2006, and J-PARC's spallation facility came online in October 2006. J-PARC will begin work with minor actinides in its Transmutation Experimental Facility in the near future. In Europe, the MEGAPIE beam at SINQ in Switzerland is used for spallation experiments, but a fulltime European Spallation Source has not yet been built

0.2 The Beam Window

Figure 4.
One of the window designs considered for SNS.
Note the domed central portion.
From *Proceedings of the Particle Accelerator Conference*, ORNL team, 2003.

_____

ADS is in the process of evolving, and one of the engineering problems thus far has been the beam window. The ADS accelerator must be kept at high vacuum, and some structure has to be responsible for keeping air and other gases out. The problem is that this same structure must also allow the beam to get out. Very high proton fluxes are necessary to produce enough neutrons to run a sub-critical fission reactor. One such estimation, done in Section 5 of this report, requires a flux of 10^{17} protons per second, each at 1 GeV. This amounts to a 16 MW beam. A beam like this is capable of melting most things that are put in front of it, and does extensive radiation damage to whatever remains solid.

Radiation damage to the window is not dealt with broadly in this report, because the extent and form of it depends on many factors. These include nuclear and chemical

properties of the material itself, temperature at which irradiation occurs, characteristics of radiation, and even spatial orientation of the crystals in the material. In short, it is experimental work. One such experiment was done in conjunction with this report, and is described in Section 6.

The main focus of this report is the mechanical properties of materials from which the window could be made. It is hoped that a radiatively cooled window, which is run hot, would be able to anneal much of the radiation damage done to it. This may be possible because of the type of damage that can do be done to it. A brief overview of radiation damage is given below.

Several different types of damage can occur. The crystal structure of the window is damaged by the high-energy ions (protons) passing through it. The atomic dislocations cause hardening and eventually embrittlement. This may be similar to the phenomenon which causes overwrought metals to become work-hardened. Irradiation-swelling of the material can also occur when dislocations result in large voids in the material, or transmutation precipitates form bubbles in it. Transmutation products are an issue: The protons may spallate or "side-swipe" the window nuclei themselves. They break nucleons off the window nuclei, or even shatter them. Additional radiation damage then occurs due to neutron bombardment, gammas, and free charged particles. In Oak Ridge's SNS, hydrogen and helium bubbles are common in used 2mm steel windows [3,4]. This causes them to be brittle. Sulfur, phosphorous, and other elements will also replace some of the iron, and have a similar embrittling effect. The transmutations cannot be prevented by running the window hot, but transmutation gases may be able to escape the window. Additionally, a design in which the beam is split and sent through multiple windows reduces radiation damage. If the windows are radiatively cooled, and therefore outside the core, a multi-beam concept is more feasible than with some other schemes.

A common unit of radiation damage is dpa, or displacements per atom. I was unable to determine why this unit is used, as it is material- and energy-specific, and physically means nothing without the application of algebra. Additionally, this algebra has obscure

material properties as inputs. The eventual result of conversion is the number of incident ions necessary to make one displacement likely. I was unable to determine how to use dpa to express damage done by neutrons. Anyone wishing to know how to work with dpa should consult [5], and be aware that $(dE/dx)_{nuclear}$ is equivalent to nuclear stopping power times material density. Information on nuclear stopping powers can be found at [6].

The mechanical properties of the window are a major issue, and it is the issue most addressed in this report. The beam can deposit an incredible amount of heat into the window, and it is necessary to get rid of this heat. There are several proposed schemes..

Windowless designs duck the issue entirely. In windowless designs, the spallation target is right up against the accelerator vacuum. However, there may be problems with evaporation of target material into the vacuum. This is especially true if a liquid metal target is used.

Gas cooling, which was briefly considered when this project began, is probably unable to remove heat from the window fast enough to be a primary cooling mechanism. It will be seen later in this report that heat deposition in the window can be phenomenal. Over a 100,000 W may be put into a window less than a millimeter thick, if the beam is run continuously. Most designs feature a pulsed beam, which allows some time for heat to disperse.

In J-PARC and Oak Ridge, the solution to heat in the window is liquid-metal cooling. This liquid metal is mercury in both cases.   Lead-bismuth eutectic alloy has been considered as well. Using liquid metal has some advantages. It is coolant for both the window and the core, and it is the target as well. Being a liquid, target radiation damage is not a concern: Neutron brightness can be maintained easily. Liquid metal ADS is currently seen as the most promising design.

There are disadvantages to liquid metal, though. The proximity of the window to the target increases radiation damage from neutrons. Liquid metal puts more pressure on the window and is highly corrosive: Its contact with the window is chemically damaging. This damage is exacerbated by conditions peculiar to ADS.

The pulsed beams in SNS and J-PARC are very intense. They cause instantaneous rates of temperature change on the order of 10^7 K/s [7], though the window itself may be only 10 K above surroundings.   This results in shock waves in the mercury. The shock waves can tear voids into the mercury, a phenomenon known as cavitation. When these voids collapse, they shoot high-pressure jets of liquid mercury at the window. Some of the deep "pitting" caused by this can be seen in Fig. 5.
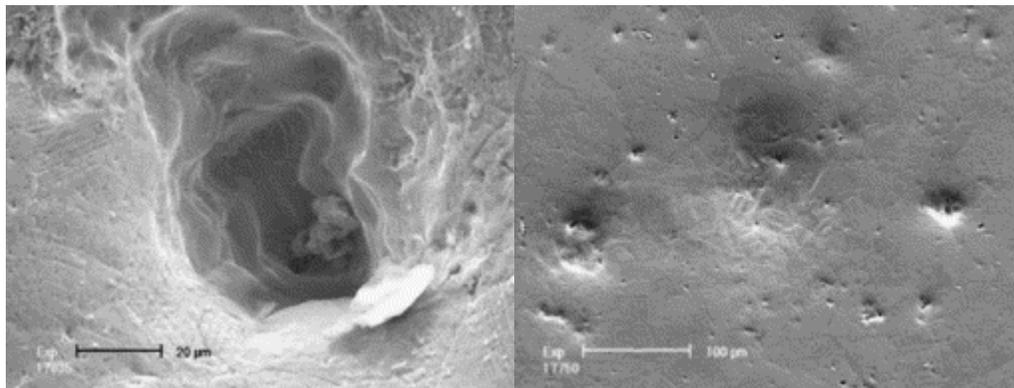

Figure 5. Pitting in an annealed 316LN window (SNS). From: J. Hunn, B. Riemer, C. Tsai, *JNM* **318**, pg. 102, (2003)

One more potential solution to heat in the window is radiative cooling. This is described in the next section.

References for Section 0:

[1] Y. Kurata, T. Takizuka, T. Osugi, H. Takano. *JNM* **301**, 1, (2002)

[2] K. Krane. *Intro. To Nuclear Physics*, Wiley, 1988

a means of spallating atomic nuclei to produce controlled nuclear fission, or to transmute existing nuclear waste into species that are easier to handle.

[3] P. Vladimirov, A. Moslang, *JNM* **356**, 1-3 (2006) p. 287-299

[4] R.L. Klueh, N. Hashimoto, M.A. Sokolov, P.J. Maziasz, K. Shiba, S. Jitsukawa. *JNM* **357,** 1-3 (2006) p. 169-182.

[5] Lee, Hunn, Hashimoto, Mansur. *JNM* **278**, 2-3 (2000) p. 266-272

[6] Isotopes Project, Lawrence Berkeley Laboratory: (get authors, dates) http://ie.lbl.gov/interact/abs3c.pdf

[7] John R. Haines. *Target Systems for the Spallation Neutron Source,* PowerPoint

+In-line References

Section 1.0: Radiatively Cooled Window - Description

This project is meant to demonstrate the feasibility of a radiatively cooled ADS beam window.

Radiative cooling comes naturally to an object that is much hotter than its surroundings. The formula for power emitted is:

$$P = a\sigma(T^4 - \text{ambient temperature}^4)$$

where P is power in Watts, T is the temperature of the object, and a is the emissivity of the material. Emissivity is a unit-less number that is always less than 1. Emitted power rises rapidly with increased temperature. Therefore, a radiatively cooled beam window is run hot.

At elevated temperatures, metals lose strength and stability. At some point, the window will no longer be able to hold back an atmosphere of pressure. Strength can be improved by increasing thickness, but this also increases the amount of heating by the beam. Designing a radiatively cooled beam window is therefore an optimization problem. A particular window material with be able to tolerate the most proton flux at a particular operating temperature and thickness.
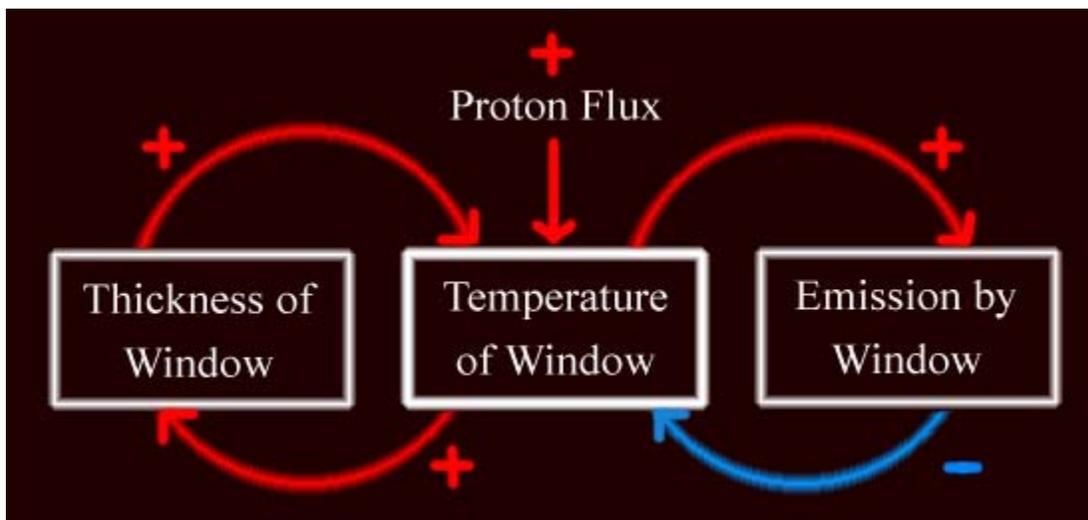

Figure 1. Dynamics of a radiatively cooled beam window.

Section 1.1: Considerations

Heating by the beam is given by **1**:

$$H = F_p \times S(E_0) \times z \times \rho \times k$$

where
  $H = $ Heating, in Watts
  $F_p = $ Flux of protons through window, in protons / second
  $S(E_0) = $ Stopping power of window material, for protons of specified energy,
      in $\text{MeV} \Big/ \text{proton} * \text{g/cm}^2$
  $\rho = $ Density of window material, in g/cm^3
  $k = 1.602 * 10^{-13}$ Joules / MeV
  $z = $ Thickness of the window (average), in cm

This formula was adapted from one found in the Journal of Nuclear Materials, Volume 356, Issues 1-3, "Irradiation conditions of ADS Beam Window and Implications for Window Material". That formula gave the rate of heating in Watts per gram, and appeared as: $H = F_p * S(E_0) / \rho * k$, which appears to have a units error. This version of the formula is justified as follows:

Each proton that passes through the window will, on average, lose a certain amount of energy to the window, heating it. This energy is described by the stopping power, $S(E_0)$, used in the formula above.. Although it is being assumed here that the proton does not actually collide with nuclear material in the window, it does pass through the window's electron cloud. The electron cloud exerts a pull on the proton, and some of the proton's energy is used to overcome it. For a 1 GeV proton, the energy lost to the window is typically about 1-2 MeV/(g/cm^2). This unusual unit becomes more understandable when used in a formula.

For instance, a radiative steel window may be 0.0020 cm thick, and it has a standard density of 7.8 g/cm^3. So, one square centimeter of the window has volume of 0.0020 cm^3. Its mass is (7.8 g/cm^3) × (0.0020 cm^3) = 0.016 g. That is, there are 0.016 grams of material present in each square centimeter of the window, or 0.016 g/cm^2.

The stopping power of steel is about 1.6 MeV/(g/cm^2) for a 1 GeV proton[1, 2]. So, each 1 GeV proton loses (1.6 MeV/(g/cm^2)) × (0.016 g/cm^2) = 0.026 MeV of energy to the steel window.

If the proton flux is 10^{17} protons per second striking the window, then a total of 0.026 × 10^{17} MeV is deposited in the window each second. This is equivalent to 420 Joules per second, or 420 Watts.

The stopping power of a material may be calculated using a semi-empirical formula provided by the Isotopes Project of Lawrence Berkeley Laboratory[2]. The stopping power decreases when the proton beam's energy is increased. At the energies relevant to ADS, the stopping power comes almost entirely from the material's electron cloud. There is a nuclear component of stopping power, but it is negligible, and left out of the formula below.

2   The formula is:

$$S_e = \left[A_9 + A_{10}\left(\frac{\ln E}{E}\right) + A_{11}\left(\frac{\ln E}{E}\right)^2 + A_{12}\left(\frac{E}{\ln E}\right) \right] \times \frac{0.6022}{M} \times 10^3$$

Here, M is the window material's atomic mass, in amu, E is proton energy in keV, and the A_n coefficients are empirical constants which are listed on the site by proton energy and the material which the protons strike. The complete table of A_n is available there[2]. An abbreviated version, containing only elements likely to be of interest for an ADS beam window, is on the next page.

Table 1.

Material	Z	M (amu)	A$_9$	A$_{10}$	A$_{11}$	A$_{12}$	E (keV)	Stopping Power (MeV/(g/cm^2))
Titanium	22	47.88	7.50E-02	2.96E+03	-5.56E+05	1.81E-07	10^6	1.62
Vanadium	23	50.94	8.17E-02	3.05E+03	-4.62E+05	1.81E-07	10^6	1.62
Chromium	24	52.00	8.17E-02	3.15E+03	-5.51E+05	1.89E-07	10^6	1.61
Iron	26	55.85	8.79E-02	3.35E+03	-6.06E+05	2.01E-07	10^6	1.60
Nickel	28	58.69	9.44E-02	3.53E+03	-6.12E+05	2.10E-07	10^6	1.62
Zirconium	40	91.22	1.33E-01	4.88E+03	-1.01E+06	2.91E-07	10^6	1.46
Niobium	41	92.91	1.36E-01	4.97E+03	-1.02E+06	2.95E-07	10^6	1.46
Molybdenum	42	95.94	1.36E-01	5.17E+03	-1.42E+06	3.24E-07	10^6	1.45
Tantalum	73	180.9	2.31E-01	7.93E+03	-2.20E+06	4.74E-07	10^6	1.25
Tungsten	74	183.9	2.28E-01	8.16E+03	-2.95E+06	5.21E-07	10^6	1.24
Rhenium	75	186.2	2.35E-01	8.11E+03	-2.44E+06	4.93E-07	10^6	1.24

It can be seen that higher **Z** elements trend to lower stopping powers. The actual best material for the window is likely to be an alloy, due to the improved mechanical properties available. In cases where one element is predominant, such as carbon steel, it is sufficient to use the stopping power of the predominant element, in this case, iron. High-carbon steel is only about 2% carbon, and ignoring carbon's contribution only leads to a loss of one significant digit. Likewise, the stopping power of an alloy containing similar-**Z** metals can probably be estimated by inspection. In other cases a weighted average can be used to find the stopping power of the alloy.

While the higher-**Z** materials have lower stopping powers, they also have higher densities. Density is one of the factors used to determine how fast the window heats up. The higher densities of the higher-**Z** materials limit the benefit of their lower stopping powers.

The window's tensile strength at operating temperature must be sufficient to hold back an assumed 1 atmosphere (0.101325 MPa) of pressure. The calculations to determine if this is possible are demonstrated below.

Tensile strengths of any sort are given in units of Force/Area. It is therefore necessary to determine how the atmosphere exerts force on the window, and on what area it is exerted. While the entire window is responsible for keeping the atmosphere out of the accelerator, the greatest load is on the edges of the window, where it is supported. If the edges can hold up, the rest should, too.

A hemispherical window design allows for the best support. The increased surface area of a hemisphere compared to a circle does not increase total force on the window. This is because the radial components of force cancel out. The component of force pushing the window back into the accelerator is proportional to the accelerator's circular opening.

The radial atmospheric forces do serve a function, however: They allow the window to hold its shape, much like a balloon.

Figure 2. A hemispherical window.
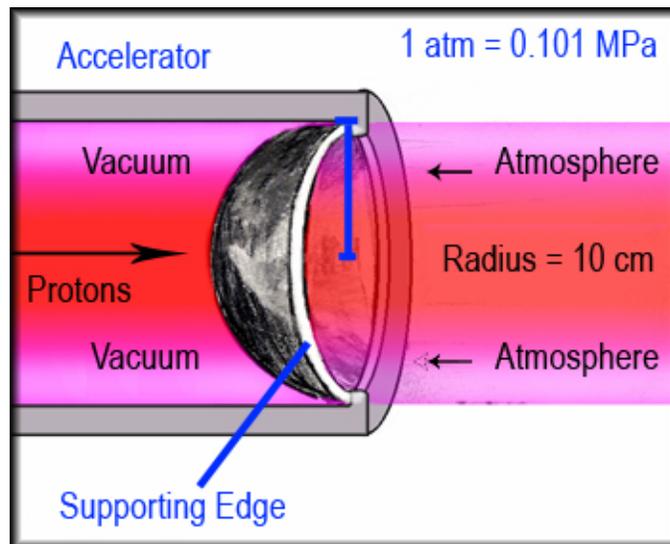
3   The total force on the supporting edge of the window is given by:

$$\text{Force}_{total} = \pi R^2 \times (\text{Ambient Pressure})$$

4   The area of the supporting edge is:

$$\text{Perimeter Area} = 2\pi R \times (\text{Thickness of Edge})$$

The thickness of the window at the edge may be slightly greater than the thickness of the window at the center. This is the case at Oak Ridge, where the edge is about 150% the thickness of the center. The radius of the window is in the vicinity of 10 cm.

5   The total amount of pressure that the window's edge must hold back is:

$$\text{Load} = \text{Force}_{total} \Big/ \text{Perimeter Area}$$

If the tensile strength of the material is greater than the load, then the window will hold, at least for a short time. In practice a safety factor would be put into the thickness.

Section 1.2: Assumptions for Optimization

In order to determine the best candidate materials for a radiatively cooled window, alloys based on 12 elements were examined. The maximum proton flux that a window of each alloy could withstand was determined based on the following assumptions:

1. The window is in all cases 10cm in radius, and must hold back 1 atmosphere (0.101325 MPa) of pressure. The load-bearing edges of the window are 1.5 times thicker than the window's center. This is similar to the windows used in Oak Ridge's SNS. The window is hemispherical, but the amount of inward force on it is proportional to the amount of it facing outwards, so the window may be approximated as a flat disc. Material densities used are room temperature densities, on the assumption that any decrease in density at high temperature will come with an equivalent increase in window thickness.

2. The ambient temperature is 300K, and the only source of heat in the window is the 1 GeV proton beam. Depending on reactor design this may or may not be a reasonable assumption: According to models generated by P. Vladimirov and A. Moslang for their 2006 article in the Journal of Nuclear Materials[1], one sixth of the heating may come from gamma rays and another 0.5% from neutron bombardment. However, this estimate is for an ADS design in which the window is in direct contact with the liquid-metal target. Here, heating by the reactor, and cooling by convection, are assumed to be negligible: The window is outside the nuclear core. It is assumed the window may be blackened so that it radiates 90% as much power as a blackbody of the same temperature. The window radiates from both its inside and outside surfaces; however, only the amount of it facing directly inward or outward radiates effectively. For radiative purposes, the window may again be approximated as a flat disk. All elemental stopping powers are calculated based on data provided by LBNL's Isotopes Project[2], and all alloy stopping powers are taken to be a weighted average of their major constituent elements.

3. The beam is operated in continuous mode, so the cyclic fatigue properties of the materials are not relevant. The beam has a "uniform" profile, in that it causes uniform heating. In fact, a uniform profile will not cause uniform heating, because the protons near the edges of the hemispherical window see more material in their way. The flux per square centimeter given in the tables below is the maximum flux that can be tolerated at the center of the window, and total flux can be generated from it using these geometric considerations. Total flux is about 163 times the central flux. The calculations are done in Section 3. Beam current in mA is gotten by multiplying total flux by 1.602×10^{-16}.Coulombs. Radiation damage from the beam or backscattered neutrons is not addressed quantitatively here.

In a radiatively cooled design, the window must emit as much power as it absorbs as heat from the protons. This is necessary for it to remain in equilibrium. Radiative designs lend themselves to hot windows: If the window were at 300K like its surroundings, its net emission would be 0 W. For each candidate material, there is an ideal range of operating temperatures, where the proton flux is maximized, the window is thick enough to be strong and durable, and thermal equilibrium is maintained.

Recall that the formula for heating is:

$$H = F_p \times S(E_0) \times z \times \rho \times k$$

where
$\quad H = \text{Heating, in Watts}$
$\quad F_p = \text{Total flux of protons through window, in protons / second}$
$\quad S(E_0) = \text{Stopping power of window material, for protons of specified energy,}$
$\qquad \text{in } \text{MeV} \big/ \text{proton} * \text{g/cm}^2$
$\quad \rho = \text{Density of window material, in g/cm}^3$
$\quad k = 1.602 * 10^{-13} \text{ Joules / MeV}$
$\quad z = \text{Thickness of the window, as seen by incoming protons, in cm.}$

F_p is not actually known, because the flux is not uniform, and z varies considerably (Assumption 3). However, it is assumed that the beam profile can be manipulated so that these factors offset each other, making Heating uniform. This makes it unnecessary to know either F_p or z at the moment. The flux per square centimeter, f_p, is presumably maximum at the window's center, so if that can be found, we will be able to determine what the rest of the window can tolerate. A conversion from the central flux per square centimeter, f_{p0}, to total flux, F_p, is done in detail in Section 3. Under the assumptions made there, $F_p \approx 163\, f_{p0}$.

Because this is a radiatively cooled design, Heating must equal Emitted power. The maximum flux f_{p0} through a square centimeter at the center of the window is then given by:

$$f_{p0} = \text{Emitted} / (S(E_0) \times z \times \rho \times k \times \pi R^2)$$

where z is now specifically the thickness at the center of the window, and $1/\pi R^2$ is a conversion factor from *F_p*, the total flux the window would experience if it were a flat disc of z thickness, to f_p.

There are now two different unknowns to deal with: z and Emitted. Emitted power, by Assumption 2, is a function of temperature (and window radius, a constant here), and z is a function of material strength (also radius and ambient pressure, both constant here). Both temperature and material strength may be regarded as independent variables. This may seem backwards: Temperature is high because of the protons, and materials lose strength because of high temperatures. However, material strength is a bounding condition: The window *has* to be strong enough, or it serves no purpose. Strength dictates the beam power possible. Temperature is also "independent", in that it is plugged into a formula to determine Emitted power, with no explicit relation to strength.

Section 2: Materials - Maximum Possible Flux

In materials science, there is seldom an exact formula for how strong a material is under different conditions. Fortunately, there is a large amount of data available on the subject. In each of the 12 alloy-bases studied, data sheets were found that gave the tensile strength of materials at high temperatures. Tensile strength tends to degrade if materials are kept at elevated temperatures for a long time, and where possible, long-term tensile strengths were used.

Below is a sample table that shows which formulas were used in calculating the maximum flux each alloy can tolerate. These formulas are derived from the assumptions described in Section 1.

T(K)	UTS (MPa)	Emitted Power (W)	Center Thickness (mm)	Max Proton Flux/cm^2
Input	Input	$0.90 \times 2 \times \sigma \times \pi R^2 \times$ $(T^4 - (300K)^4)$	$\dfrac{Safety \times R \times atm}{2 \times S(E_0) \times 1.50}$	$\dfrac{Emitted}{\rho \times S(E_0) \times z \times \pi R^2}$

T is Temperature in Kelvin, UTS is Ultimate Tensile Strength in megapascals (sometimes given for long times at temperature), W is Watts, σ is the Stefan-Boltzmann constant, R is the window radius, and atm is the ambient pressure. All these formulas must include the necessary conversion factors so that units agree. Very often raw data was given in units of kpsi (thousand lb. / in^2), and had to be multiplied by 6.892 to convert it to MPa. Similarly, temperatures were given in several scales.   Density and stopping power for each material are given in the blurbs near its respective table. Beam current in mA can be obtained from proton flux/cm^2 at the center by multiplying by:

$$1.602 \times 10^{-19} \, C/p \times 1000 \, mA/A \times (163.1 = Total \; flux/(flux/cm^2))$$

For each alloy-base, an effort was made to find the most suitable alloy for an ADS beam window. In some cases, such as vanadium-based alloys, data was limited, in others, such as steel, there was so much data a thorough search would be outside the scope of this project.

The alloys examined were found to fall into two categories: Low temperature, lower beam-current alloys, and refractory alloys. The low-current alloys have the six atomically lightest bases. They have a distinct advantage, in that they may be run in air at the relevant temperatures. Their ideal operating temperatures, where the most proton flux could be tolerated, were no higher than 650°C, and they typically tolerated fluxes on the order of 10^{14} protons/cm^2/s, or several mA total beam current. Because these protons are at 1 GeV the corresponding beam power would be the same number of MW as mA. (e.g., 7.5 mA implies 7.5 MW) This may be sufficient for a power-generating ADS reactor.

In this section, calculations are done to find maximum total proton flux possible. The maximum tolerable flux is inversely proportional to window thickness. In some cases, especially among the refractory metals, the max flux may be much higher than is necessary for a power-generating ADS, and the window thinner than is easily fabricated. For example, an Inconel-718 window can cope with 73.3 mA of beam current at 0.012 mm of center thickness. This is about the same thickness as light aluminum foil. Inconel-718 is difficult to machine, and a decent-size power-generating ADS may only need 16 mA of beam current. This window would be better if it were:

(73.3mA / 16 mA) × 0.012 mm = 0.055 mm thick, with 16 mA of beam current

There is now a factor of 4.58 safety in the window's thickness, in addition to the factor of 4 safety that was present in this window before.

In Section 4, I will give window thicknesses and relative strengths of some windows whose beam current restricted to 16 mA. In the Power Generation Section, I will justify the assumption that a 16 mA beam can sustain a decent-size (660MW) power plant.

The refractory alloys can generally cope with 10 times as much beam current as the low-temperature alloys, or be 10 times thicker for the same current. This thickness, aside from making fabrication easier, reduces the air-pressure stress on the window.

Unfortunately, actual air cannot be used in the reactor, because all the refractory alloys burn in oxygen at operating temperatures. It may be possible to run them in a nitrogen atmosphere, doped with a small amount of hydrogen to stick to any oxygen that leaked in. Helium is another possibility, but is more likely to leak through the window itself.

The refractory alloys may also cope with radiation damage better, as higher temperatures may allow transmutation gasses to escape more easily, and some radiation damage may anneal away. However, since the refractory alloys get their high-temperature strength from structural resistance to heat, this requires experimentation.

The advantages that may be offered by refractory alloys must also be balanced against the fact that it may be more difficult to blacken them to emit properly. The requirements of blackening are somewhat less stringent because hot metals emit at a higher percentage of blackbody to begin with.

Section 2.1: Low-Temperature Alloys

1. Aluminum (Z=18).

Aluminum H16, H18, and 2014-T6 were examined, and 2014-T6
(90.4 - 95% Al, 3.9 - 5% Cu, 0.2% Cr max, 0.7% Fe max) was found to have the best
properties of these alloys. Aluminum alloys are inferior to other low-current alloys for
ADS purposes. However, they are suitable for other types of accelerators: The MIT-
Bates linear electron accelerator has a window of 0.05mm thick, Teflon-coated aluminum
foil. (This accelerator also makes use of a concept that could be useful in ADS: There
are several incomplete windows inside the accelerator, which are differentially pumped
so as to make a transition from 10^{-9} atmospheres in the main accelerator to 10^{-5}
atmospheres behind the aluminum window. Source: Jan Vanderlaan, MIT-Bates.)

Fig. 19. Tensile strength of 2014-T6 at various temperatures
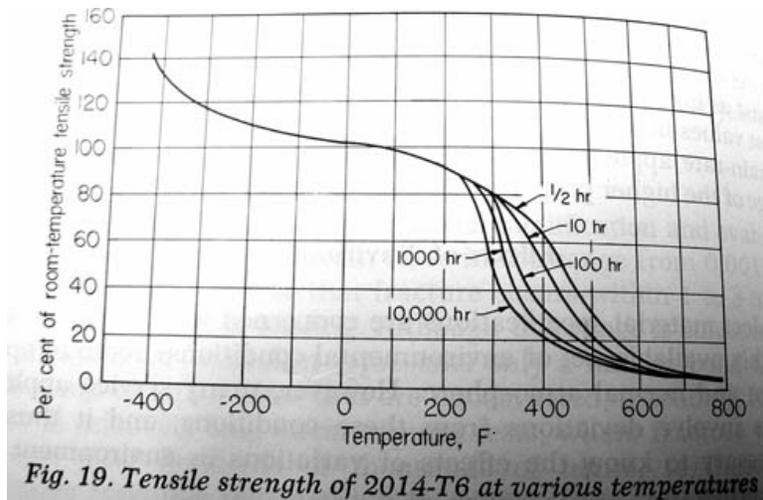
T(K)	UTS (MPa) 10,000 hr	Emitted Power (W)	Center Thickness (mm)	Max Proton Flux/cm^2
300	483	0	0.007	0.00E+00
366	415	32	0.008	1.12E+14
422	145	76	0.023	9.30E+13
477	101	141	0.033	1.21E+14
533	63	233	0.054	1.24E+14
589	29	359	0.117	8.83E+13

Table Set 1. Density = 3.96g/cc, Stopping Power= 1.75 MeV×cm^2/g. Values obtained
and converted from the image above. The best thickness for this window is 0.054 mm,

about the thickness of heavy-duty aluminum foil. There is a maximum total beam current of 3.24 mA, or 3.24 MW beam power for 1 GeV protons. One beam like this might sustain a small power plant at full power (see Power Generation Section), but not a large one. Although 10,000 hr strengths of Al 2014-T6 were used, the lifetime has little tolerance for error at the best operating temperature of 533K (= 260°C = 500°F).
Source: W.F. Kehler. *Aluminum*, 1980

2. Titanium (Z=22).

Various Titanium alloys were examined; only "beta" titanium alloys were found to have good strength. Of these, Crucible Beta III (Ti-11.5%Mo-6%Zr-4.5%Sn) was found to have the best properties at temperatures below 730°F. Titanium alloys are superior to Aluminum alloys in terms of the flux they can tolerate.

Table 4 – 100-HOUR CREEP AND CREEP STABILITY – Sheet
(Specimens solution treated 1400°F, descaled and pickled, aged 950°F 8 hr, creep loaded as shown for 100 hr and finally tensile tested without removing oxide)

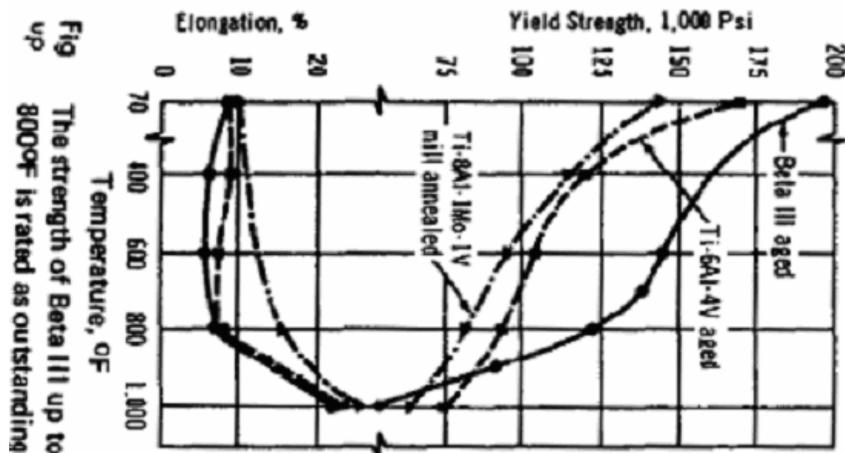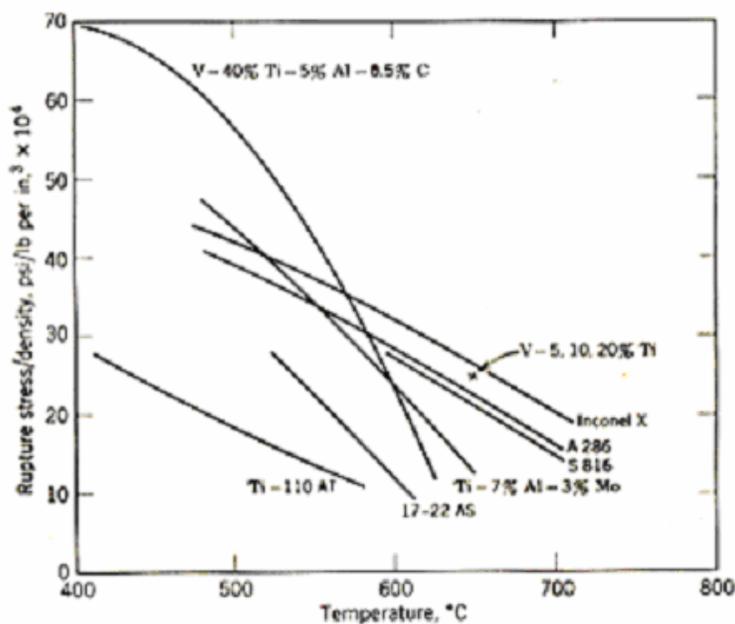| Creep Temp °F	Creep Stress psi	Plastic Creep %	Tensile Strength psi	Yield Strength (0.2% Offset) psi	Red. of Area %
As-aged tensile properties			195000	183000	30
500	130000	0.07	199000	192000	43
600	115000	0.16	202000	195000	33
700	35000	0.05	202000	186000	22
	75000	0.17	206000	198000	30
800	10000	0.04	205000	192000	25
	23000	0.20	204000	193000	31

Fig up

The strength of Beta III up to 800°F is rated as outstanding

T(K)	Creep Stress (MPa) 100 hr	Emitted Power (W)	Center Thickness (mm)	Max Proton Flux/cm^2
533	896	233	0.015	3.77E+14
589	793	359	0.017	5.14E+14
644	517	526	0.026	4.91E+14
700	159	742	0.085	2.13E+14

Table Set 2. Density = 5.09g/cc, Stopping Power= 1.6 MeV×cm^2/g. Values obtained and converted from the image above. Because only short lifetime data was available, a safety factor of 4 was put into the thickness of the window, which reduced the Max Proton Flux by a factor of 4. As this material will only creep 0.16% in 100 hours without the safety factor (see upper left of image), a reasonable lifetime can be expected from this window if radiation damage can be tolerated. The best center thickness is 0.017 mm (similar to standard aluminum foil), at an operating temperature of 589K (= 316°C = 600°F). Proton flux can be four times higher than through the Aluminum-base window, with a maximum total bean current of 13.4mA. Source: *ASM Online Data Sheets.*

3. Vanadium (Z=23).

Data on Vanadium-based alloys was somewhat incomplete; no very long lifetime stresses were available. Of the alloys for which 100hr rupture data was available, V-40%Ti-5%Al-0.5%C was found to have the best properties. Based on a graph on pg.93 of *The Metallurgy of Vanadium*, an alloy with a somewhat lower percentage of titanium may perform better. The 0.5%C increases strength, as does the 5% aluminum, which gives the alloy better stress-rupture properties than an equivalent amount of chromium. Pure Vanadium is a hard material, like Chromium. Calculations indicate that if sufficient lifetime and fabricability can be demonstrated, then Vanadium-based alloys could be a good choice for a lower-current, air-operated window, as this alloy is able to tolerate a beam current of 38.7 mA.

Figure: Rupture stress/density, psi/lb per in.^{3} $\times 10^4$ vs Temperature, °C. Curves labeled: V–40% Ti–5% Al–0.5% C; V–5, 10, 20% Ti; Inconel X; A 286; S 816; Ti–110 AT; Ti–7% Al–3% Mo; 17-22 AS.

T(K)	UTS (MPa) 100 hr Rupture	Emitted Power (W)	Center Thickness (mm)	Max Proton Flux/cm^2
673	920	632	0.015	9.96E+14
773	772	1119	0.018	1.48E+15
873	283	1836	0.048	8.89E+14

Table Set 3. Density = 5.3g/cc, Stopping Power= 1.62 MeV×cm^2/g. Values obtained and converted from the image above. Because only short lifetime data was available, a safety factor of 4 was put into the thickness of the window. The best center thickness is 0.018 mm, at an operating temperature of 773K (= 500°C = 782°F), and a maximum total beam current of 38.7 mA. This proton flux is almost 3 times higher than through the Titanium-base window. Source: Rostoker. *The Metallurgy of Vanadium,* 1958.

4. Chromium (Z=24).

Chromium is more typically found as an additive than a base in alloys. It is particularly good at improving corrosion resistance, such as the "stainlessness" of stainless steel. Only one majority-chromium alloy was found with sufficient data to analyze it as a window material, a 50Cr-50Ni alloy. Stress-rupture-data for 1,000 hrs was used with a safety factor of 2 in the window thickness. This translates to half as much stress and proton flux. Because halving the stress tends to extend the window lifetime by

a factor of about 10 (see below), this window may last 10,000 hrs. This alloy is somewhat inferior to Titanium Crucible Beta III, but may improve if used at lower temperatures.

Table 2 – TYPICAL STRESS-RUPTURE PROPERTIES

Test Temperature °F	Stress, psi for Rupture In	
	100 hours	1000 hours
1200	20000	10000
1300	13000	6000
1400	7500	3500
1500	5000	2500
1600	3000	2000
1700	2500	1500
1800	1500	1000
1900	800	500

T(K)	UTS (MPa) 1,000 hr	Emitted Power (W)	Center Thickness (mm)	Max Proton Flux/cm^2
922	69	2290	0.098	3.66E+14
977	41	2901	0.163	2.78E+14
1033	24	3625	0.280	2.03E+14
1089	17	4476	0.392	1.79E+14

Table Set 4. Density = 7.92g/cc, Stopping Power= 1.6 MeV×cm^2/g. Values obtained and converted from the image above. The best thickness for this window, based on the data available, is 0.098mm, which in this material may be thick enough to make it rather stiff. The best temperature is 933K (= 649°C = 1200°F) with a maximum total beam current of 9.6mA. There is a safety factor of 2 in window thickness, which may extend lifetime to 10,000 hrs.   Lower operating temperatures may allow for thinner windows and better performance. This window has similar flux tolerance to Titanium Crucible Beta III, but removes more energy from the proton beam. Source: *ASM Online Data Sheets*.

5. **Iron (Z=26).**

   There is an immense variety of steels, and extensive documentation on their properties. Because of this, a full survey of steel types is outside the scope of this project. The familiarity of steel makes it attractive as a window material: The most suitable steel found can tolerate 85% as much proton flux as the best Vanadium alloy, and has stress-

rupture data available to 100,000 hrs. (In actual ADS conditions, this lifetime would be significantly shortened by radiation damage, but that is outside the scope of this section.) Steel is also much less susceptible to fatigue than other metals, making pulsed-beam operation more viable. Pitting-resistant 316 LN steel is in use at the liquid-metal-target window at Oak Ridge's pulsed SNS[3], and the low-activation steel F82H has been considered by researchers at Switzerland's SINQ[4]. No high-temperature tensile data for F82H could be found, although data for type 316 was available. For a radiatively cooled, solid target window, it was found that the steel Sanicro 31HT was slightly superior to type 316 (Eastern 316, data from ASM Online Data Sheets). This difference may evaporate if the higher nickel content of 31HT is found to lead to premature embrittlement, so data for both steel types has been provided in the tables below. The steels Enduro HCN, Enduro 317 and Sandvik 253 MA were also examined, but were not as good as 316 or 31HT.

Type 316 (Eastern): 10-14Ni, 16-18Cr, 2-3Mo, max 2Mn, max 1Si, max 0.08C, max 0.045P, max 0.30S. Density = 8 g/cc. Maximum total beam current is 27.2mA.

T(K)	UTS (MPa) 10,000 hr 1% Creep	Emitted Power (W)	Center Thickness (mm)	Max Proton Flux/cm^2
811	167	1360	0.020	1.04E+15
866	125	1780	0.027	1.03E+15
922	88	2290	0.039	9.21E+14
977	54	2901	0.062	7.26E+14
1089	19	4476	0.175	3.97E+14

Type 31HT (Sanicro): 30-35Ni, 19-23Cr, 0.15-0.60Al, 0.15-0.60Ti, max 1.5Mn, max 1Si, max 0.75Cu, max 0.10C , max 0.015P, max 0.015S. Density = 8.1 g/cc. Maximum total beam current is 28.4mA.

T(K)	UTS (MPa) 100,000 hr Rupture	Emitted Power (W)	Center Thickness (mm)	Max Proton Flux/cm^2
866	134	1780	0.025	1.09E+15
922	85	2290	0.040	8.94E+14
977	56	2901	0.061	7.40E+14
1033	35	3625	0.096	5.82E+14
1089	23	4476	0.149	4.65E+14

Table Set 5. Stopping Power= 1.6 MeV×cm^2/g in both cases. Lower operating temperatures may give the metal some better characteristics, but higher temperatures may give it higher radiation tolerance, and the thicker windows may be easier to fabricate. This matter requires more investigation, as it has been found that irradiation temperature has a significant effect on the form of radiation damage in some steels[4]. The best thickness for the Type 316 window, based on the data available, is 0.020mm, with a maximum total beam current of 27.2 mA and an operating temperature of 811K (= 538°C = 1000°F),. The best thickness for the Type 31HT window, based on the data available, is 0.025 mm, with a maximum total beam current of 28.4 mA and an operating temperature of 866K (= 593°C = 1100°F). Note that 316's strength data is for 1% Creep in 10,000 hrs, while 31HT's data is for rupture in 100,000 hrs. These windows do not have flux tolerances as high as that of the best Vanadium alloy, but are better studied and have more reliable lifetime data. The thinnest of these windows is similar in thickness to aluminum foil, the thickest is somewhat stiff. Source: *ASM Online Data Sheets*.

6. Nickel (Z=28).

.Unfortunately, extensive data could not be found on the most interesting Nickel-base alloy examined, Inconel-718. Inconel-718, should its lifetime prove sufficient, can tolerate more flux than the best Vanadium alloy (nearly twice as much, if Inconel-718's long-term strength is assumed to be ¼ of its short-term strength at relevant temperatures). Inconel-718 is the window material being used in a 1MW pulsed beam at the liquid-metal J-PARC in Japan[5]. Inconel-718 may contain up to 1% cobalt. It was not determined if the cobalt can or should be removed from metal used in a beam window.

One potential problem with this and other nickel alloys is that they are very hard, and the window thicknesses which allow them the highest flux are no thicker than light aluminum foil. Foils like this cannot be rolled, although it may be possible to use other techniques such as powder-melting the window onto a mold. It its possible to make the windows thicker, but then they will absorb more heat, and have similar considerations to refractory windows. If fabrication of thin nickel-alloy windows is impractical then windows of other materials are more attractive.

Other Nickel-base alloy studied include Kubota KHR48N, Udimet 901, MO-RE 40-MA, and Inco-Alloy 330. This is just a sampling, and there are likely to be others worthy of consideration for the window. Of this sampling, none could tolerate as much flux as the Inconel-718, but lifetime data was available. The Udimet and Kubota alloys were nearly equal: Udimet came out ahead in the temperatures for which data was available, but Kubota may be superior if allowed to operate at a lower temperature. Its lifetime data is also more reliable.

Nickel alloys may function at greater than 650°C (923K), but oxidation could become a real issue.

Inconel-718: 50 – 55 Ni, 17 – 21 Cr, 17 Fe, 4.75 - 5.5Nb, 2.8 - 3.3 Mo, 0.65 - 1.15 Ti, 0.2 - 0.8 Al, Max 1 Co, Max 0.35 Si, Max 0.3 Cu, Max 0.08 C, Max 0.015 S, Max 0.015 P, Max 0.006 B.   Density = 8.19 g/cc, Stopping Power= 1.62 MeV×cm^2/g. There is a safety factor of 4 in the window's thickness, reducing the flux by the same factor. This is because of the lack of lifetime data. Maximum total beam current is 73.3mA, corresponding to a beam power of 73.3MW. Source: matweb.com.

T(K)	UTS (MPa) Short-Term	Emitted Power (W)	Center Thickness (mm)	Max Proton Flux/cm^2
300	1375	0	0.010	0.00E+00
923	1100	2301	0.012	2.81E+15

Udimet 901: 42.0Ni, 36.0Fe, 13.0Cr, 5.6Mo, 2.9Ti, 0.2Al, 0.15B, 0.03C, max 0.1Mn, max 0.1Si. Density = 8.26 g/cc, Stopping Power= 1.60 MeV×cm^2/g. There is a factor of 2 safety which is likely to extend the lifetime past 10,000 hrs. Flux tolerance peaks near 650°C (922K, 1200°F), with a window thickness of 0.013 mm. Maximum total beam current is 69.8mA. Source: *ASM Online Data Sheets*.

Table 4 – STRESS-RUPTURE DATA – Bar
(Solution treated and aged)

Test Temperature	Stress, psi for Rupture in	
°F	100 Hours	1000 Hours
1000	130000	112000
1200	92000	76000
1300	70000	53000
1400	44000	30000
1500	24000	12000

Table 5 – TYPICAL CREEP STRENGTH
(Solution treated and aged)

Test Temperature	Stress, psi, for 1% Creep in
°F	100 Hours
1000	105000
1200	73000
1400	27000
1500	13000

T(K)	UTS (MPa) 1,000 hr Rupture	Emitted Power (W)	Center Thickness (mm)	Max Proton Flux/cm^2
811	772	1360	0.009	2.34E+15
922	524	2290	0.013	2.67E+15
977	365	2901	0.018	2.36E+15
1033	207	3625	0.033	1.67E+15
1089	83	4476	0.082	8.24E+14

Kubota KHR48N: 45 – 50 Ni, 25 – 30 Cr, balance Fe, 4.0 – 6.0 W, 0.4-0.6 C, max 1.5 Mn, max 1.5 Si, max 0.03 S, max 0.03 P.   Density = 8.03 g/cc, Stopping Power= 1.59 MeV×cm^2/g. No safety factor is needed; stresses used are the minimum at which rupture can occur after 10,000 hrs, and are not much higher than the average stresses for rupture after 100,000 hrs. (Note, however, that these are extrapolated values; see source image below.) Flux tolerance may be higher at lower temperatures, for which there was no data. Maximum beam current for available data is 32.7mA. Source: *ASM Online Data Sheets*.

Table 2   Stress Rupture and Creep Rupture Properties

Temperature		Rupture stress																Creep stress, average, 0.001 %/h	
		100 h				1000 h				10,000 h				100,000 h					
		Average		Min		Average		Min		Average		Min		Average		Min			
°C	°F	MPa	ksi	MPa	ksi	MPa	ksi	MPa	ksi	MPa	ksi	MPa	ksi	MPa	ksi	MPa	ksi	MPa	ksi
760	1400	...	...	...	...	...	...	...	...	90.5	13.13	74.6	10.82	70.9	10.28	58.6	8.50	...	...
816	1500	...	...	...	...	85.6	12.41	70.6	10.24	65.8	9.55	54.5	7.90	49.6	7.19	41.2	5.98	...	...
871	1600	82.9	12.03	68.5	9.93	62.8	9.11	52.0	7.54	46.4	6.73	38.5	5.58	33.4	4.84	27.9	4.05	...	...
927	1700	61.6	8.93	51.0	7.39	44.7	6.49	37.2	5.40	31.6	4.58	26.4	3.83	21.6	3.13	18.1	2.63	30.0	4.35
982	1800	44.5	6.45	37.0	5.36	30.8	4.47	25.8	3.74	20.6	2.99	17.4	2.53	13.3	1.93	11.3	1.64	21.7	3.15
1038	1900	31.1	4.51	27.1	3.93	20.5	2.97	17.2	2.50	13.0	1.88	11.0	1.60	7.93	1.15	6.76	0.98	14.3	2.07
1093	2000	21.1	3.06	17.8	2.58	13.1	1.90	11.1	1.61	7.86	1.14	6.76	0.98	4.62	0.67	4.00	0.58	8.89	1.29
1150	2100	13.8	2.00	11.7	1.70	8.14	1.18	6.96	1.01	4.69	0.68	4.00	0.58	...	...	...	...	4.62	0.67
1205	2200	8.76	1.27	7.52	1.09	4.96	0.72	4.27	0.62	...	...	...	...	...	...	...	...	2.14	0.31

Long-term creep-rupture properties were extrapolated from Larson-Miller parameter versus stress plots. Creep and rupture stress values are subject to periodic revisions as the results from long-term tests become available.

| T(K)	UTS (MPa) min 10,000 hr Rupture	Emitted Power (W)	Center Thickness (mm)	Max Proton Flux/cm^2
1033	74.6	3625	0.045	1.25E+15
1144	38.5	5466	0.088	9.7E+14
1255	17.4	7928	0.194	6.36E+14

Table Set 6. Inconel-718 is the best of all the low-current, air-operated window materials if it has sufficient lifetime and can be fabricated. J-PARC's spallation neutron

source, whose first beam came on line in late 2006, will be able to give data on this material's behavior when exposed, long term, to beam irradiation: Its window is made from Inconel-718.

Many other Nickel-based alloys are promising. An advantage of the Kubota KHR48N alloy is that data shows it has decent flux tolerance even when the window is thick and stiff. The stress-to-rupture in a minimum of 10,000 hrs is close to the stress-to-rupture in an average of 100,000 hrs.

7. Zirconium (Z=40).

Zirconium which has been purified of Hafnium is known as "reactor grade". Its known suitability for such environments makes it worth looking at. Zirconium is not as strong at elevated temperatures as some of the other metals examined.
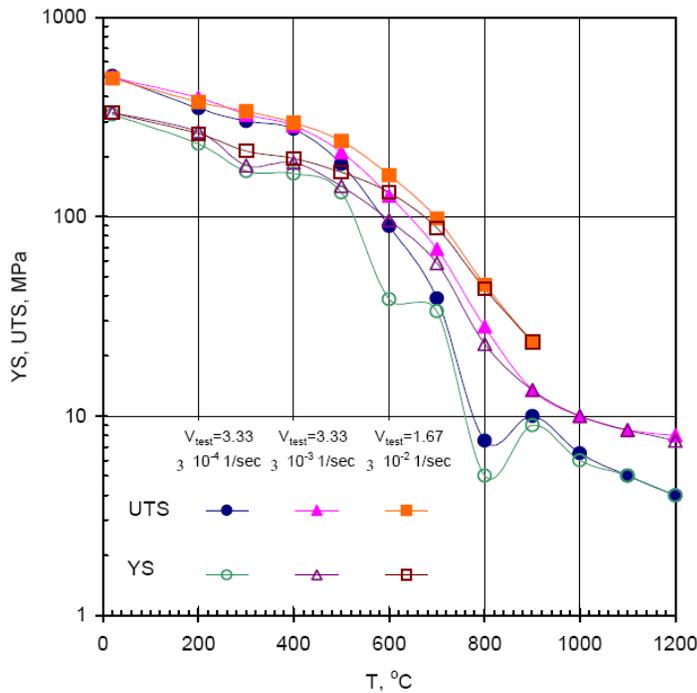
T(K)	UTS (MPa)	Emitted Power (W)	Center Thickness (mm)	Max Proton Flux/cm^2
773	230	1119	0.059	3.97E+14
873	150	1836	0.090	4.25E+14
1073	27	4224	0.500	1.76E+14
1273	10	8394	1.351	1.29E+14
1089	8	15068	1.689	1.86E+14

Table Set 7. Zr-2.5Nb: Density = 6.53 g /cc, Stopping Power = 1.46 MeV×cm^2/g. This alloy has its flux peak between 500 and 600°C, and a thickness of 0.06 to 0.09 mm. Maximum total beam current is 11.1 mA at 600°C = 873K. Uncertainties are introduced by the uncertainty in strength (UTS), which is as high as 50 MPa due to the difficulty of "eyeballing" values from the logarithmic graph above. This is exclusive of uncertainties in the measured strength, which, as with most metals examined, is seldom mentioned and may be unknown. Source: Rodchenkov & Semenov. *High Temperature Mechanical Behavior of Zr-2,5 % Nb Alloy*, SMiRT Conference, 2003.

Section 2.2: Refractory Alloys

The image below is from the NASA Technical Note *Stress-Rupture and Tensile Properties of Refractory-Metal Wires at 2000°F and 2200°F (1093°C and 1204°C)* by Petrasek and Signorelli, Lewis Research Center, 1969. This document made a detailed study of many of the alloys also examined in this section. The purpose of the document was to find suitable wires to act as reinforcing fibers in superalloy composites. Although examining such fiber-reinforced materials was outside the scope of this project, they ought to be considered for a radiative ADS window due to their higher strength. A downside of composite materials is the heating irregularities they may introduce.
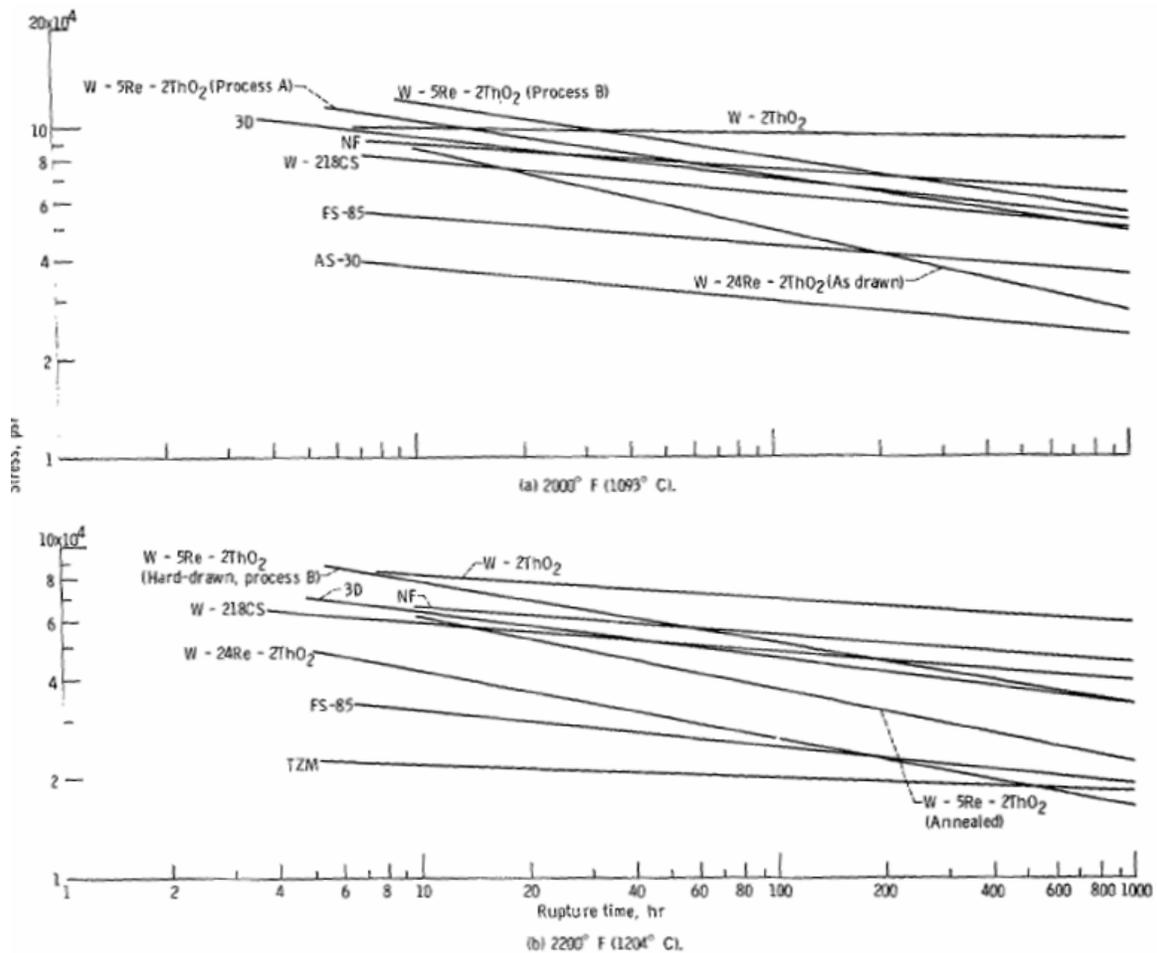

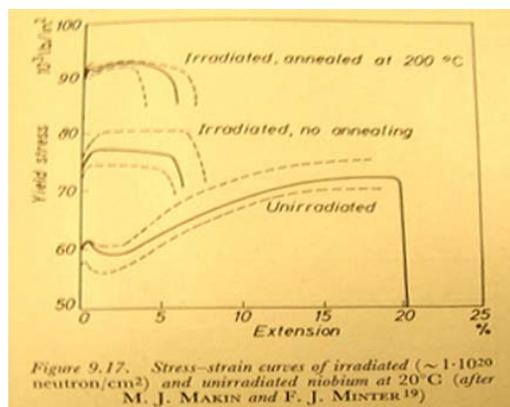Figure 8. - Stress to cause rupture of wires at 2000° and 2200° F (1093° and 1204° C).

NASA Graph.

Although refractory alloys cannot be run in air, the entire reactor need not be in an inert atmosphere. It may be possible to put an extremely thin, non-pressure-bearing window some ways in front of the refractory window for the sole purpose of keeping a bubble of inert gas in front of the window. This would require more examination. A similar dummy-window could be used to keep mercury vapor away if a liquid-metal target were being used.

8. Niobium (Z=41).

Niobium, sometimes known as Columbium, is considered the first of the refractory alloys. Its strongest alloys, however, contained other refractory alloys. The best such alloy in terms of high temperature mechanical properties was a Molybdenum and Zirconium–modified ternary alloy, containing about 70% Nb, 20% W, 7% Ti, and 3% Mo. The Zr is a 1% addition that is important for strength, but it is not known which other element(s) were reduced to make room for it. Because density data was not available for this alloy at the time of writing, the density of the alloy Nb-28Ta-10W (also known as FS-85) was used in calculations instead. FS-85 is examined here as well, and is shown on the NASA Graph above.

Niobium is known to irradiation-harden: Yield stress may be increased by 30% when irradiated to 10^{20} neutrons/cm^2 at 20°C and then annealed at 200°C. This increase in hardness, and corresponding increase in both strength and brittleness, was looked for in Vanadium in the Experimental section of this report.

Figure 9.17. Stress–strain curves of irradiated ($\sim 1 \cdot 10^{20}$ neutron/cm^2) and unirradiated niobium at 20°C (after M. J. MAKIN and F. J. MINTER[19])

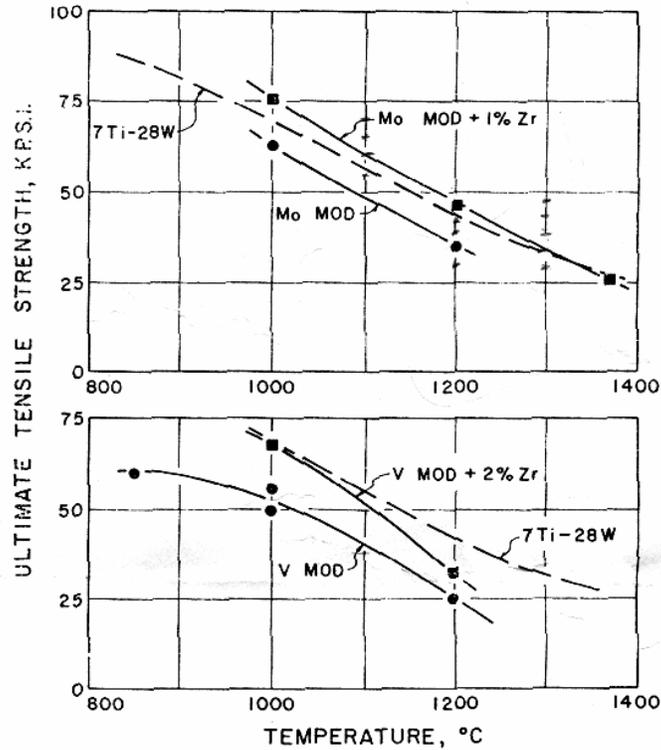**Source: Miller. *Tantalum and Niobium*, New York Academic Press, 1959**

Fig. 5. Effect of small zirconium additions on the tensile strengths of the vanadium and molybdenum modifications. Included for comparison is the strength-temperature curve of the 65% Cb-7% Ti-28% W alloy.

Source: Sisco (ed.). *Columbium and Tantalum*, Wiley, 1963

FS-85: 72 Nb, 28 Ta, 10 W. Stopping Power = 1.34 MeV×cm^2/g. Short-term strength data was available for this alloy on matweb.com, for room temperature and for 1588K (= 1315°C = 2400°F). A safety factor of 4 was inserted into thickness to make up for the lack of long-term data there. This may, based on NASA's graph for 2200°F, extend the lifetime past 1,000 hrs and possibly past 10,000 hrs: The material's 2400°F short-term strength is equivalent to 23 kpsi, and it is only being stressed at a quarter of that. The material's 2200°F, 1,000-hr strength is about 18 kpsi. This window can tolerate 92.0 mA of beam current.

T(K)	UTS (MPa) Short-Term	Emitted Power (W)	Center Thickness (mm)	Max Proton Flux/cm^2
300	580	0	0.023	0.00E+00
1588	160	20364	0.084	3.52E+15

Nb-7Ti-20W-3Mo-1Zr: Stopping Power = 1.43 MeV×cm^2/g. There is a safety factor of 4 in this window due to lack of lifetime data. The density of FS-85 was used, but Stopping Power was, as usual, a weighted average of the elemental Stopping Powers. As the calculations stand, this window can tolerate a maximum flux of 126 mA. Source: Sisco (ed.). *Columbium and Tantalum*, **Wiley, 1963**

T(K)	UTS (MPa)	Emitted Power (W)	Center Thickness (mm)	Max Proton Flux/cm^2
1273	517	8394	0.026	4.39E+15
1373	414	11368	0.033	4.76E+15
1473	317	15068	0.043	4.84E+15
1573	234	19604	0.058	4.65E+15
1673	145	25092	0.093	3.68E+15

Table Set 8. Density used for both alloys is 10.16 g/cc, the density of FS-85. The Mo-MOD + 1% Zr alloy appears to be the best of the Niobium alloys, but there is some uncertainty in its density and lifetime. The data for FS-85 is more reliable. Other alloys examined included Cb-22 and Cb-15W-5Mo-1Zr.

9. Molybdenum (Z=42).

Molybdenum-TZM, which contains 0.5% Ti and 0.08% Zr, was found to be a very good refractory alloy. It may be in the recrystallized or stress-relieved (annealed) condition. The recrystallized condition is about twice as strong but probably more prone to embrittlement. It is very stable for long periods of time at wide ranges of high temperatures. At 1200°C, near where it tolerates the maximum flux, it is the most stable of all alloys found ([10], pg. 14). However, it should be noted that oxidation can cause it to fail catastrophically; that is, an oxygen leak would cause the window to burst rather than erode. The distinction may or may not be trivial given the thinness of the window.

Molybdenum and Molybdenum Alloys vs Selected Superstrength Alloys: A Comparison
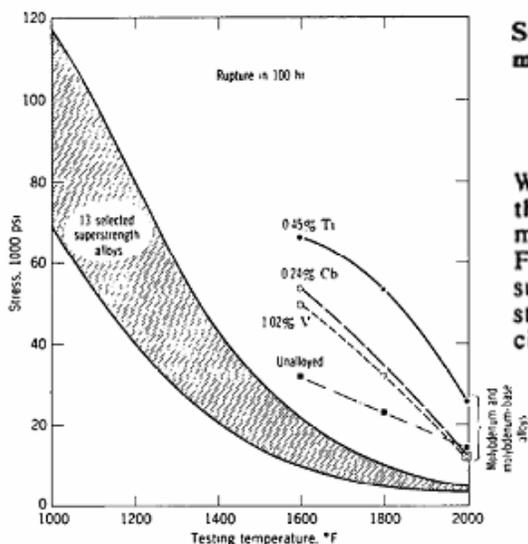
Stress-to-rupture properties of molybdenum and molybdenum alloys.

Within the temperature range of 1600-2000 °F, the creep-to-rupture strength of the unalloyed metal itself is superior to that of the best of Fe-base, Co-base, Ni-base, or other commercial superalloys. At 2000 °F, Mo-base alloys have strength properties superior to those of commercial high-temperature alloys at 1600 °F.

T(K)	UTS (MPa)	Emitted Power (W)	Center Thickness (mm)	Max Proton Flux/cm^2
300	1068	0	0.013	0.00E+00
700	1137	743	0.012	8.43E+14
922	841	2291	0.016	1.92E+15
1144	744	5470	0.018	4.06E+15
1366	620	11152	0.022	6.91E+15
1589	379	20397	0.036	7.72E+15
1811	172	34454	0.078	5.93E+15
1922	103	43727	0.131	4.51E+15

Table Set 9. Moly-TZM: Density = 10.16 g /cc, Stopping Power = 1.45 MeV×cm^2/g. There is a factor of 4 safety which may be excessive given the stability of Moly-TZM at high temperatures. The maximum flux shown on this table occurs at 1589K = 1316°C = 2400°F, but it is probably almost as high if not higher at 2200°F, for which data was not available from the source used for the rest of the table. This window can mechanically tolerate up to 202 mA. Other alloy examined included binary Molybdenum-Rhenium alloys. Source (Image & Data): *ASM Online Data Sheets.*

10. Tantalum (Z=73).

Tantalum, while strong, is unstable at high temperatures. It loses strength rapidly if held at elevated temperatures for a long time. Because of this, Tantalum-based alloys were deemed unsuitable for use in an ADS beam window.

Table 3 – REPRESENTATIVE STRESS-RUPTURE PROPERTIES OF 0.040-in. SHEET

Condition	Test Temperature °F	Stress psi	Time to Rupture hours
Stress relieved, 3 hr at 2250°F	2000	52200	1.5
		50000	2.4
		47000	6.0
		45000	20.0
	2400	38000	0.1
		28000	2.8
		25000	4.0
		22000	16.4
Recrystallized, 1 hr at 2730°F	2700	19000	1.9
		10500	4.8
	3500	8000	0.5
		5000	3.4

Image Source: *ASM Online Data Sheets.*

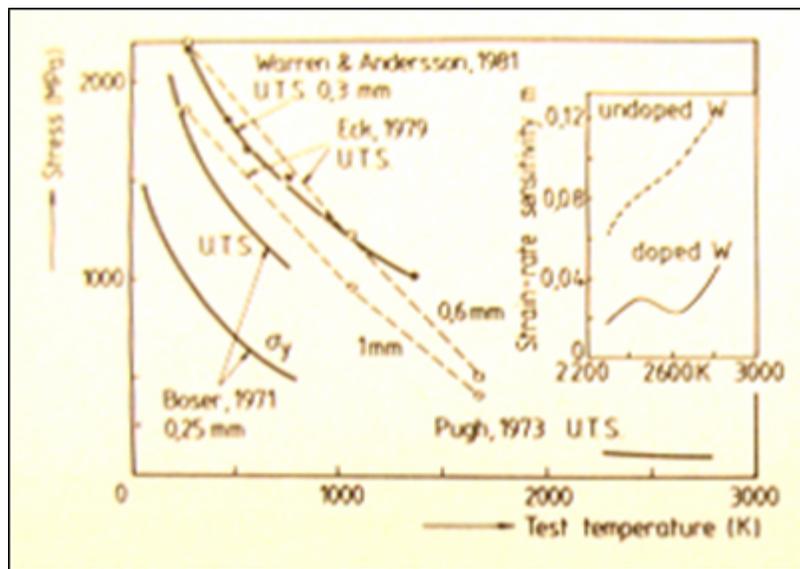| T(K)	UTS (MPa)	Emitted Power (W)	Center Thickness (mm)	Max Proton Flux/cm^2
1073	469	4224	0.029	1.39E+15
1273	345	8394	0.039	2.04E+15
1473	234	15068	0.058	2.49E+15
1673	172	25092	0.078	3.05E+15
1873	110	39434	0.123	3.06E+15

Table Set 10. Ta-10W: Density ~16.7 g/cc, Stopping Power = 1.25 MeV×cm^2/g. This is the alloy for which the table above gives data. Since density data was not available for this alloy, pure density was used in calculations, so flux estimations are probably optimistic. There is a factor of 4 safety, but it may be insufficient unless the 10% W significantly improves stability. The maximum flux tolerated, according to calculations, is 80 mA. Data Source: cabot-corp.com

11. Tungsten (Z=74).

Tungsten was one of the more interesting refractory alloys. Thoriated tungsten (2% ThO_2) is used in car headlights because of its good vibration resistance. Although Thorium can become fissile, this is probably not relevant, as the proton beam is capable of spallating even very stable elements like [56]Fe. NASA deemed thoriated Tungsten to be the best material for its wires in the study referred to previously [10]. It found that W-$2ThO_2$ was more stable than Rhenium-containing Tungsten alloys at high temperatures.. Alternately doped "non-sag" Tungsten, which is use in standard incandescent bulbs and contains 60-200 ppm of Potassium, has good properties as well. This is because the Potassium forms small bubbles which add stiffness to the material, a very similar phenomenon to H or He embrittlement.

Doped Tungsten: Essentially pure. Source: Pink & Bartha (ed.). *The Metallurgy of doped/non-sag tungsten*, **Elsevier Applied Science, c.1989. Data is guessed from Boser's and Pugh's data on image below and is somewhat uncertain.**

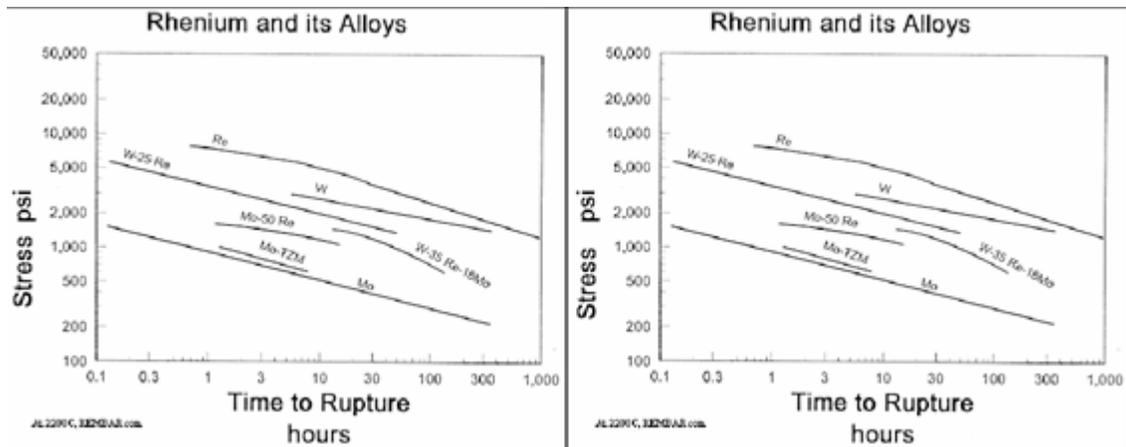| T(K)	UTS (MPa)	Emitted Power (W)	Center Thickness (mm)	Max Proton Flux/cm^2
300	1650	0	0.008	0.00E+00
2250	100	82148	0.135	5.05E+15
2800	80	197052	0.169	9.69E+15

Thoriated Tungsten: 98W, 2ThO$_2$. Source: Avallone & Baumeister (ed.). *Marks' Standard Handbook for Mechanical Engineers*, McGraw & Hill, 10th Ed.

T(K)	UTS (MPa)	Emitted Power (W)	Center Thickness (mm)	Max Proton Flux/cm^2
1477	276	15251	0.049	2.58E+15
1922	207	43718	0.065	5.55E+15
2200	172	75038	0.078	7.95E+15

Table Set 11. Stopping Power and Density of pure Tungsten used in both cases, 1.24 MeV×cm^2/g and 19.3 g/cc, respectively. There is a factor of 4 safety in both cases. Doped Tungsten may reach a maximum beam current of 253 mA, but must do this at a very high temperature. In Section 4, conversions to a greater thickness and 16 mA (16 MW) beam are done for ideal operating temperatures. Doped Tungsten run at its ideal of 2800K would heat the window so much that it would start to cut noticeably into beam efficiency. There is no real need to run a doped tungsten window this hot, unless it is the only way to anneal radiation damage. The Thoriated Tungsten may be a better choice and according to [10], it is very stable at least up to 1477K, where it can mechanically tolerate 67.5 mA. The calculated maximum for WThO$_2$ is 208 mA.

12. Rhenium (Z=75).

Of the more exotic refractories studied, which included Osmium and Iridium, Rhenium was the only one for which sufficient data could be found. Rhenium is very hard and very expensive, and of all 12 metals reported in this paper, it was the only one which may be best in its unalloyed form.

T(K)	UTS (MPa) 1,000 hr Rupture	Emitted Power (W)	Center Thickness (mm)	Max Proton Flux/cm^2
1873	14	39434	0.490	6.13E+14
2473	8	119897	0.891	1.03E+15

Table Set 12. Pure Rhenium: Density = 21.03 g/cc, Stopping Power = 1.24 MeV×cm^2/g. There is a factor of 2 safety in the window thickness which will hopefully extend the mechanical lifetime past 10,000 hrs. Data is taken from the graph above; it is visually extrapolated for 1873K = 1600°C. At this temperature it will take a beam current of exactly 16 mA, enough to run an ~660MW power plant. (See Power Generation Section). It is capable of a 26.7 mA (26.7 MW) beam, which is less efficient: It loses about 0.5% of its power to the window. Lifetime data for other temperature was not found. Data Source: REMBAR.com

Section 3: Total Flux Calculations

To convert from flux per square centimeter at the center of the window, f_{p0}, to total proton flux, F_p, assumptions must be made about the geometry of the window. It has been stated that the window is 1.5 times as thick near its edge as at its center, that it is hemispherical, and that it is being evenly heated by the protons. Since heating by the protons is proportional to the thickness of window they must pass through, flux must be reduced near the edge to account for greater thickness. This makes calculating total flux through the window somewhat involved. In the calculations below, the perpendicular thickness of the window, $z(r)$, is assumed to increase linearly with r, so $z(10cm) = 1.5 \times z(0cm)$. The thickness of the window that protons must pass through is labeled s. At $r = 0cm$, $s = z$. The flux at r, $f_p(r)$, is inversely proportional to s, with a constant of proportionality α.

$$S = \frac{Z(r)}{\cos\theta} = \frac{Z_0\left(1+\frac{r}{20}\right)}{\cos\left(\sin^{-1}\frac{r}{16}\right)} = \frac{Z_0\left(1+\frac{r}{20}\right)}{\sqrt{1-\frac{r^2}{100}}}$$

$$r = 10\sin\theta$$
$$\theta = \sin^{-1}\frac{r}{10}$$

$$Z(r) = Z_0 + \frac{(1.5Z_0 - Z_0)}{10 - 0}r$$
$$= Z_0\left(1 + \frac{r}{20}\right)$$
$$[\text{Z is linear with r}]$$

$$f_p' = \frac{\alpha}{Z_0} = \frac{\alpha}{S(0)} \;;\; f_p(r) = \frac{\alpha}{S(r)} \;;\; F_p = \int_{r=0}^{r=10} \frac{2\pi r \, \alpha \, dr}{S(r)}$$

α = some constant

$$\Rightarrow F_p = \frac{2\pi\alpha}{Z_0}\int_{r=0}^{r=10} \frac{r\sqrt{1-\frac{r^2}{100}}}{\left(1+\frac{r}{20}\right)}dr = 2\pi f_{p_0}\int_{r=0}^{r=10} \frac{r\sqrt{1-\frac{r^2}{100}}}{\left(1+\frac{r}{20}\right)}dr$$

The Integrator program at wolfram.com (integrals.wolfram.com) was used to solve this integral. A screen shot is below. The program took x as input instead of r, and the constants at the front of the integral were left off.

$$\int^x \frac{\sqrt{1 - \frac{x^2}{100}}}{\frac{x}{20} + 1} \, dx =$$

$$2\left(\frac{1}{2} \sqrt{100 - x^2} \ (x - 40) - 350 \sin^{-1}\left(\frac{x}{10}\right) + 200 \sqrt{3} \ \tan^{-1}\left(\frac{2(x+5)}{\sqrt{300 - 3x^2}}\right)\right)$$

Using the limits of integration $x = 0$ and $x = 10$, and returning the constants $2\pi f_{p0}$ to the front of the integral, the solution becomes $F_p \approx 163.1 \, f_{p0}$

Section 4: Materials - A 16 mA Beam and Thicker Windows:

Aluminum, Titanium, Chromium and Zirconium –based windows are all unsuitable for a 16 mA beam.

A V-40Ti-5Al-0.5C window can mechanically tolerate 38.7 mA at 0.018 mm of center thickness and an operating temperature of 500°C. This thickness includes a factor of 4 safety to compensate for the fact that only 100-hr rupture strengths were available. If the beam were 16 mA, then this window could be 0.044 mm thick and still operate at 500°C. Assuming it was still hemispherical and 1.5× as thick at the edge, the safety factor would be increased to 9.68 from 4 due to the greater thickness. (An additional factor of 2.37)

A 31HT steel can mechanically tolerate 28.4 mA of current at 0.025 mm of center thickness and an operating temperature of 593°C. This thickness includes no safety because rupture strengths used are for 100,000 hrs. According to [1], operating at temperatures above 420°C significantly reduces irradiation hardening of some types of steel. In a 16 mA beam, this window could be 0.045 mm thick and still maintain the same operating temperature. There would now be a safety factor of 1.78 in the window's thickness.

Inconel-718's mechanical behavior in a 16 mA beam was discussed in the beginning of the Materials- Maximum Possible Flux section. Because of the difficulty of making Nickel-based windows thin, I will state the 16 mA behavior of the Nickel window that was thickest to begin with. Kubota KHR48N can tolerate 32.7 mA at 0.045 mm of thickness and an operating temperature of 760°C. There is no safety due to long lifetime data. In a 16 mA beam, this window could be 0.092 mm thick and would have a safety factor of 2.04.

A Nb-20W-7Ti-3Mo-1Zr window can tolerate roughly 126 mA of beam current at 0.043 mm thickness and an operating temperature of 1200°C. In a 16 mA beam this window could be 0.339 mm thick and maintain the same operating temperature. There would be a

safety factor of 7.88 in window thickness in addition to the factor of 4 safety already there to account for a lack of lifetime data. In a 32 mA beam the window could be 0.169 mm thick with a safety of 3.94 in addition to the factor of 4 already there.

A Moly-TZM window can tolerate 202 mA at 0.036 mm of thickness and 1316°C. In a 16 mA beam this window can be 0.455 mm thick and remain at the same operating temperature. This thickness, 0.455 mm, can be produced by commercial methods and is very stiff: There is an sample of 0.5 mm pure Molybdenum in the WPI Physics Department at the time of this writing. With 0.455 mm of thickness, there is now a safety factor of 12.6 in the window, in addition to the factor of 4 that was already there. Alternately, a 32 mA beam could have a 0.227 mm window and a factor of 6.31 additional safety. This kind of flexibility also allows for some flaws in the initial assumptions (Section 1). For example, even if emission cannot be made to reach 90% of blackbody, there is room for the window's thickness, and therefore heating, to be reduced.

A Ta-10W window could possibly tolerate 80 mA at 0.123 mm thickness and 1600°C for an unknown but probably limited period of time. This thickness could be increased to 0.615 mm in a 16 mA beam, which would put an additional safety factor of 5 in the window.

A Thoriated Tungsten window can tolerate 208 mA of beam current at 0.078 mm and 1927°C. In a 16 mA beam this window could be as thick as 1.01 mm without running any hotter or taking any more power from the beam. (Power loss to window is ~0.5% in this case, 75kW.) The additional safety factor gained by doing this would be 13.0. Alternatively, a 0.507 mm window could work with a 32 mA beam and a factor of 6.49 safety. Since it is possible and common to make $WThO_2$ thinner than 1mm, the $WThO_2$ window in a 16 mA, power-generating ADS could be made thinner and run significantly below capacity. In this way it is similar to Moly-TZM, and has similar flexibility in initial assumptions.

The Rhenium window was already well-suited to a 16 mA beam at 1600°C in Section 2.2. If run at 2200°C, it can be 0.891mm thick and take 26.7 mA of current. At this temperature, it could be 1.49 mm thick in a 16 mA beam. This makes for an additional safety factor of 1.67.

Conclusions: Thoriated Tungsten and Moly-TZM make the mechanically best refractory ADS beam windows. Low-temperature, oxygen-tolerant windows must be thinner, and are limited by the difficulty of manufacture. For this reason a steel window may come out ahead, although the Vanadium alloy has technically more tolerance for flaws in the initial assumptions.

The ability of these materials to withstand or anneal radiation damage remains to be seen. Pure Vanadium was tested for radiological effects in the Experimental section.

Section 5: Power Generation

The assumption that a 16 mA beam is sufficient for power generation is justified here.

Assume:

1. Each proton from the beam will spallate 1 target nucleus and yield 30 neutrons. The actual number of neutrons per spallation event is a spread that depends on the target material and proton energy, assumed here to be 1 GeV.

2. There is a reactor core which, without the neutrons produced by spallation, is at 97% criticality. The spallation neutrons bring it to exactly 100% criticality. This means that at any instant, 3% of the free neutrons are from spallation.

3. Each neutron that causes a fission event releases 200 MeV of energy. Again, the actual energy is a spread that depends on fuel material, but 200 MeV is a commonly used number[8].

4. The beam is running at 16 mA, or 10^{17} protons/second total flux. Since these are 1 GeV protons it is a 16 MW beam. The beam is 15% power-efficient. The power plant has 30% thermodynamic efficiency. The system must be able to tolerate miscellaneous loses that bring nuclear efficiency as low as 80%.

Calculations:

1. (30 free neutrons/proton) $\times$ (10^{17} protons/second) / (0.03) = 10^{20} free neutrons/second
2. At least 80% of these cause a fission from which the energy can be captured. 0.80 fissions / neutron $\times$ 200 MeV/fission $\times$ 10^{20} neutrons/second = 1.6×10^{22} MeV/second.

3. Plant is 30% efficient: $0.30 \times 1.6 \times 10^{22}$ MeV/s $\times 1.6 \times 10^{-13}$ J/MeV $= 770$ MW.

4. 770 MW produced – 16 MW / 0.15 power to the beam $= 660$ MW

Some math done on an electric bill shows that a house can be run on an average of a kW of power. If a third of power consumption is residential this 16 mA ADS is capable of powering a small-to-midsize city.

References for Sections 1-5:

[1] P. Vladimirov, A. Moslang, *J. Nucl. Mater.* **1-3** (2006) p. 287-299

[2] Isotopes Project: Stopping Powers. http://ie.lbl.gov/interact/abs3c.pdf

[3] L.K. Mansur, *J. Nucl. Mater.* **1** (2003) p. 14-25

[4] X. Jia, Y. Dai, M. Victoria, *J. Nucl. Mater.* **1** (2002) p. 1-7

[5] H. Nakashima, *Radiological Safety for the J-PARC Project* (2006), p. 29
http://eurisol.wp5.free.fr/WP5/Documents/5th_meeting_Munchen_2006/JPARC.pdf

[6] NIST Center for Neutron Research: http://www.ncnr.nist.gov/resources/n-lengths/

[7] NASA Technical Note *Stress-Rupture and Tensile Properties of Refractory-Metal Wires at 2000°F and 2200°F (1093°C and 1204°C)* by Petrasek and Signorelli, Lewis Research Center, 1969

[8]   Krane. *Intro. to Nuclear Physics*, Wiley, 1988

+ In-line References.

Section 6: Experiment- Irradiation Hadening in Vanadium

Abstract

Many metals undergo hardening in the presence of radiation. Irradiation hardening initially leads to increased tensile strength, but eventually to brittleness. This has been a problem in ADS prototypes. The purpose of this experiment was to see if irradiation hardening could be observed in Vanadium. Samples that had been irradiated in WPI's Washburn reactor and unirradiated samples were measured for Knoop hardness. Data taken suggests that irradiation hardening did occur in the irradiated samples. The unirradiated samples had a mean microhardness of 151 ± 2 HK. The irradiated samples had a mean microhardness of 157 ± 3 HK when a very outlying data point is included, and a mean microhardness of 160 ± 2 HK when that point is excluded. In the first case, statistical variation could be responsible for the irradiated vs. unirradiated hardness difference 6.6% of the time. When the outlier is excluded, statistical variation could only produce this difference less than 0.2% of the time. This suggests that the hardness difference is statistically significant, and that Vanadium may be too sensitive for use in an ADS beam window.

Introduction

The phenomenon of irradiation hardening has been observed in many metals, including steel[1], tungsten[2], tantalum[3], niobium[3], and nickel[4]. The swelling, transmutations, and dislocations that cause hardening would seem to be possible in all metals, given sufficient radiation. Some metals are more susceptible than others. For example, a small amount of rhenium improves the swelling resistance of tungsten at least up to 1073K [2]. It is not desirable that an ADS beam window material be overly sensitive to radiation of any sort. Even if not inside the core, damage from the proton beam can produce many free neutrons, alphas, betas, and gammas. A material that showed mechanical changes after small radiation exposures is probably not a good structural material where larger exposures are expected.

Exposures that cause irradiation hardening may be expressed in several ways. Sometimes damage is recorded, and expressed in terms of dpa, a difficult and obscure unit to work with. Other times exposure is expressed in terms of the neutron flux per square centimeter. This may be some 10^{20} n/cm^2 [2,3]. The 10 kW Washburn reactor cannot practically deliver this much neutron flux, at least not in the thermal range. Its thermal flux is about 10^{11}n/cm^2/s [5]. An MQP poster by Christopher Patterson and Timothy Tully, "Optimization of Thermal Neutron Flux in the WPI LEU Reactor Core" suggests that there is much more fast neutron flux (Fig.1). However, the fast neutrons may not contribute significantly to radiation damage, as they do not absorb well and could not knock a metal atom out of its lattice unless extremely energetic. The gamma-radiation in the core is about 10^6 Roentgen/hr. Alpha radiation is a unmeasured quantity, but certainly present. Beta radiation is also unmeasured, but it alone is visible: The Cherenkov radiation it causes takes the form of blue light (Fig. 2). It is possible for all these types of radiation to have structural effects on a material, but it is unknown how much damage they each contribute. The reactor was operated by Steve Laflamme and student assistants.

The microhardness of the irradiated and unirradiated vanadium samples were measured using a Knoop hardness tester. The Knoop tester makes a small, diamond-shaped indentation in the material being tested. The ratio of the force on the indenter and the area of the indent gives the Knoop microhardness. The testing machine was operated by Boquan Li.

Each sample vanadium sample was about 2.5cm × 0.5cm × 0.05 cm. The samples were cut from a single sheet by Roger Steele. The sheet was documented as 99.7% pure vanadium.
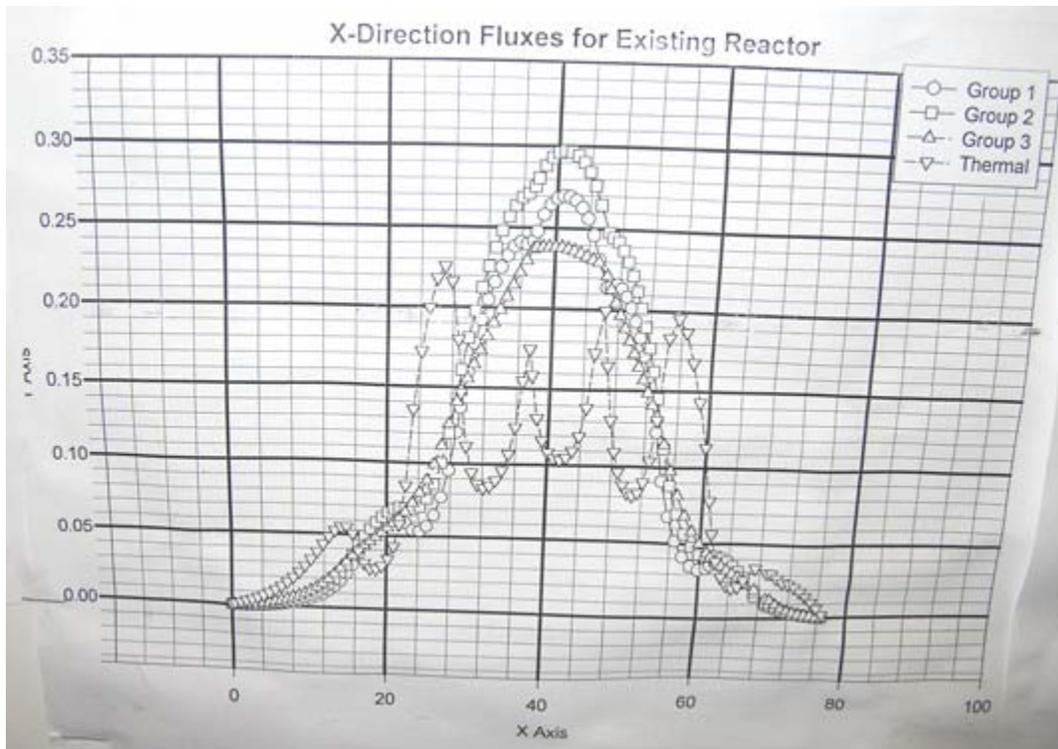
Figure 1. From "Optimization of Thermal Neutron Flux in the WPI LEU Reactor Core" MQP poster by Christopher Patterson and Timothy Tully. The neutron flux is graphed as a function of location in the core, with neutrons separated into 4 energy spectra: thermal and 3 fast. The image suggest the presence of many more

fast neutrons than thermal. Unfortunately, the lack of units labels or explanations

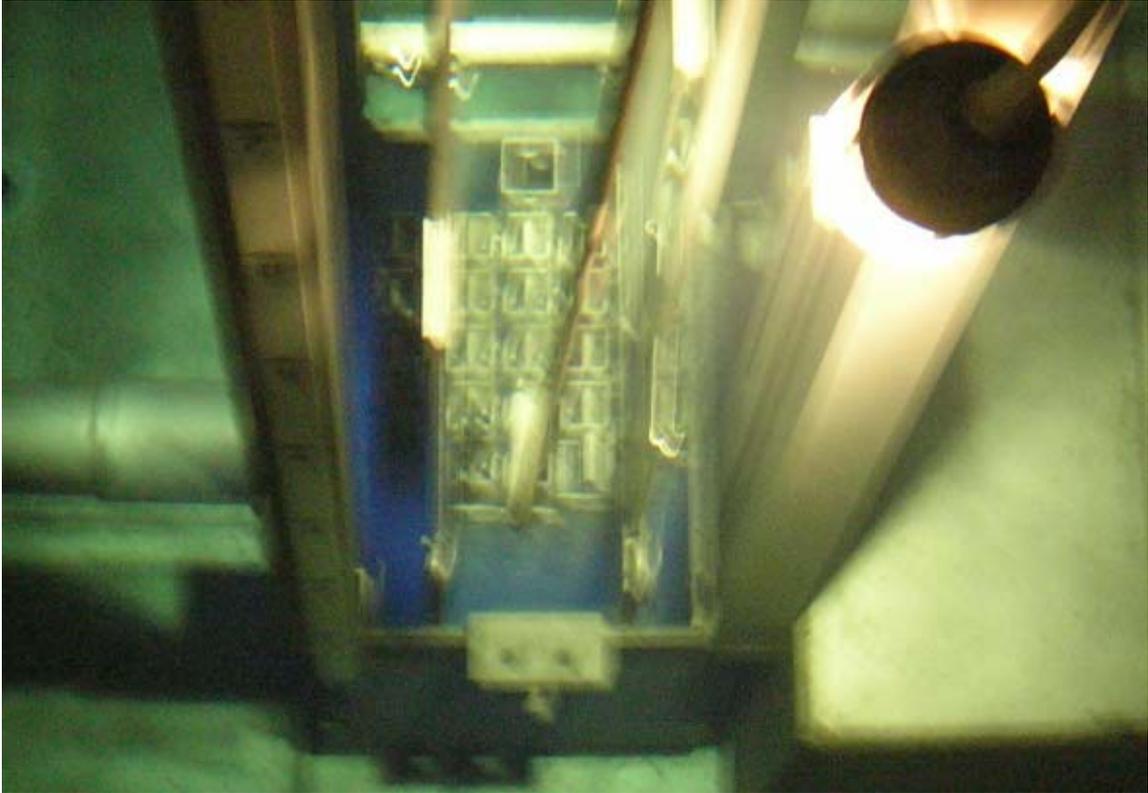**makes this data difficult to quantify.**

Figure 2. The blue glow, called Cherenkov radiation, is evidence of beta particles (fast free electrons) in the Washburn reactor. The area where samples are inserted is glowing brightly.

Procedure

A single 5 × 5 cm, 0.5 mm thick sheet of 99.7% pure vanadium "foil" was obtained from Sigma-Aldrich. This piece of metal was very stiff, not foil-like. There was some blue-green oxidation on its surface, and many abrasion scratches. These scratches later made hardness testing more difficult, and ought to have been polished off. The metal was also not perfectly flat. Four appropriately-sized strips were cut from this sheet by Roger Steele. Each took 15 to 20 minutes on a cutting wheel.
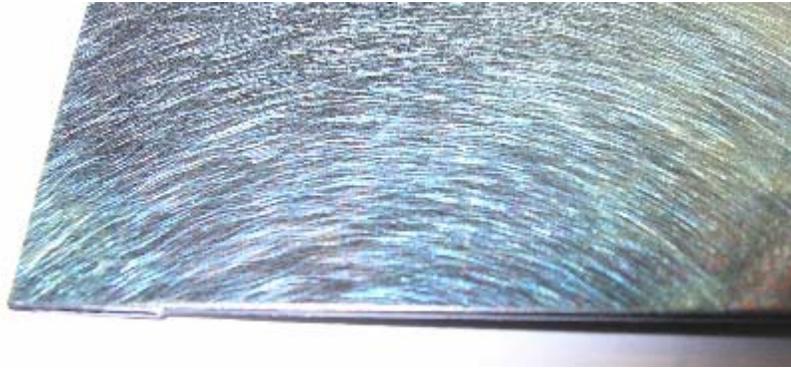

Figure 3. Uncut Vanadium.

The samples were brought down to the nuclear lab and weighed.

Table 1. Sample masses.

Sample 1	Sample 2	Sample 3	Sample 4
0.3625 g	0.4052 g	0.3603 g	0.4097 g

Samples 1 and 2 were selected for irradiation and put in small specimen jars. Sample 2 was put in the jar marked "D". Samples 3 and 4 were visually distinguishable. The samples were wiped but not washed beforehand.

The Washburn reactor was started shortly after 1:00 pm and was at full power by 1:42 pm. "Full Power", according to the digital monitor, is 9600 ± 100 W. The reactor

remained at full power throughout the irradiation. Pool temperature also remained fairly constant at 76 ± 1 °F.

The samples were fed down into the core at 1:47 pm. They remained there for exactly an hour, at which point the reactor was shut down by dropping the blades into the core. After this hour, the neutron flux through them totaled about 3.6×10^{14} n/cm^2. They had been exposed to 10^6 Roentgen of gamma-rays. Their expected activity level was 106.3 mCi [5], far too hot to be removed from the pool. For this reason they were allowed to remain in the pool until the next day. The half-life of radioactive ^{52}V is 3.76 minutes, so about 370 half-lives had passed when the experiment began again the next day.

It was apparent upon removing the samples from the pool that they were still hot. A gamma intensity of 0.38 ± 0.02 mR was measured, and the Geiger counter beeps were so continuous as to overlap each other. Sample 1 was placed in the 8192-channel Germanium-crystal gamma spectrometer. This spectrometer is known to have background peaks corresponding to ^{40}K (1463.03 keV), ^{214}Pb (87.06 and 77.22 keV) , and ^{238}U (122.98 keV). Sample 1 showed ^{40}K, and all 6 of the peaks corresponding to ^{187}W, which has a half-life of 23.9 hrs. From this it was determined that the unspecified 0.3% impurity in the vanadium sample was significantly tungsten. ^{24}Na was also detected; it is believed this came from handling the sample barehanded prior to irradiation. There was a peak at 511.32 keV that was believed to be an "escape peak", corresponding to the mass-energy of the electron, and another at 2237.15 keV, which is the energy of an ^{24}Na peak minus the electron mass energy. ^{214}Bi is a natural decay product of uranium and believed to be part of the background There was a lot of indeterminate noise in the sample too, which could correspond to any number of elements. When reading these spectrographs, it is on the researcher to determine which possible elements are plausible [5]

An unirradiated V sample, Sample 3, was placed in the spectrograph as well to ensure that no radioactivity had been present prior to irradiation. Surprisingly, radioactive ^{187}W
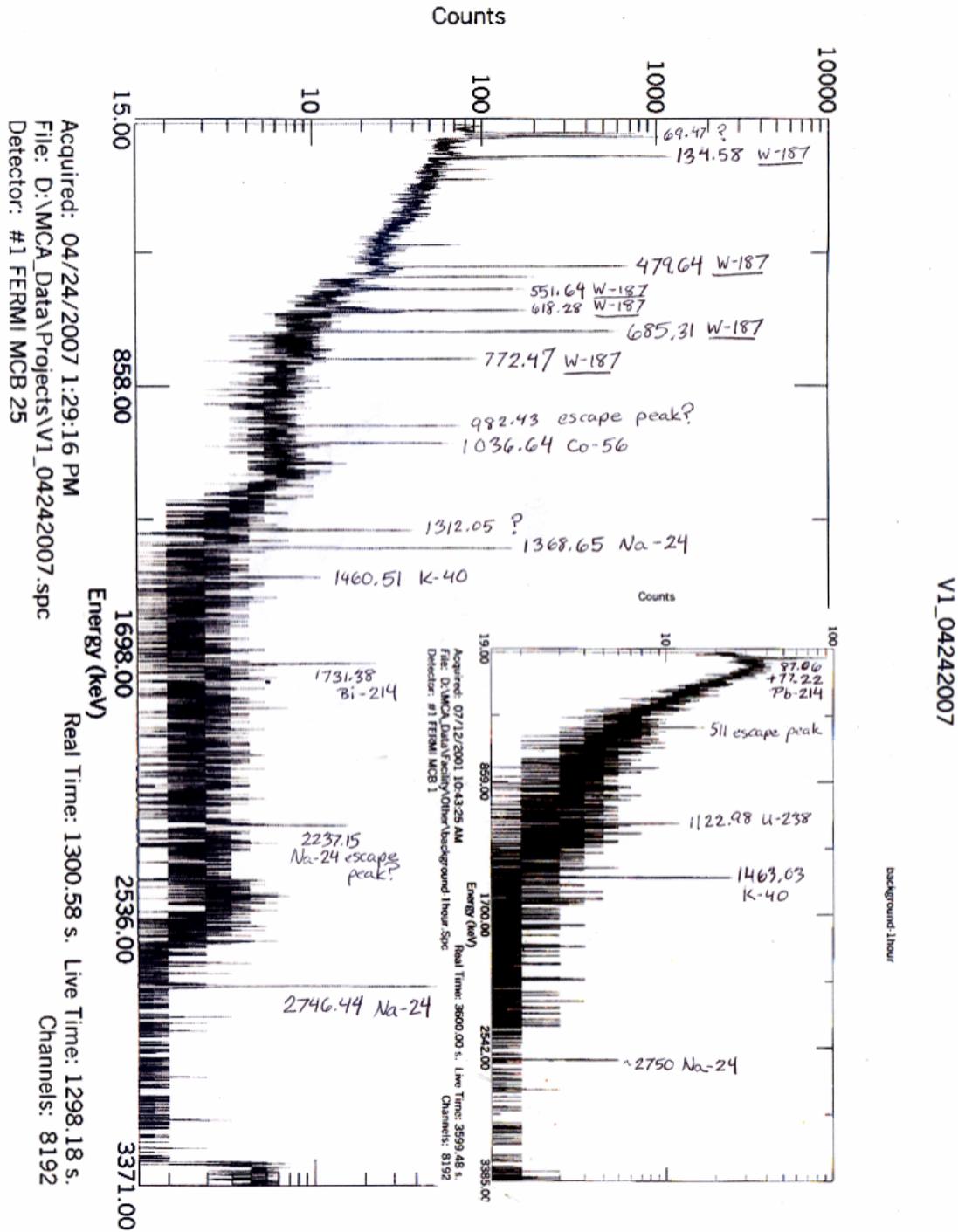
Figure 4. Spectrograph of irradiated V sample, 22 hrs 43 min after completion of irradiation. Note W impurities, underlined. Inset is background count. Numbers given are energy in keV as recorded by the spectrograph. They are known to have a calibration uncertainty of ±1 keV near the center of the spectrum, but up to 8 keV

at higher energies. Generally less counts at higher energies because of the inefficiency of the spectrometer there.

_____

peaks showed there as well. When Sample 3 was placed in a different tray than Sample 1 had been in and counted again, [187]W did not appear. This demonstrates the need for care when dealing with radioactive samples, as they can contaminate the things they touch.

Because of the [187]W activity and [24]Na activity, the samples were not cleared for temporary release from the facility until the next day, when they had had more time to decay.

The next day Samples 1 and 2 were signed out of the facility. Their activity was now estimated to be <0.1 μCi. They were brought upstairs to have their microhardness tested.

Unquantified uncertainty was introduced by the fact that the samples were not perfectly flat and not polished. Since 1 and 2 could not be machined after irradiation, we made do with the rough surfaces. The relationship of the surface scratches to the Knoop indentation can be seen in Fig. 5.

There were 6 indents made on Sample 1, 4 on Sample 2, and 8 on Sample 3. It was decided that results from Samples 1 and 2 should be taken as equivalent data, and that there was no need to indent Sample 4, as its treatment was no different from that of Sample 3. The results are given in Table 2.

Boquan Li, who operated the hardness tester, believed that the outlying point mentioned in the abstract was a fluke due to the sample not being flat. It is marked with an asterisk in the table. This point does lie 2.35 standard deviations from the mean, and more than means Chauvenet's Criterion for disregarding data. Two sets of statistical analysis were done for the results of this experiment: One with the outlying point included, and one with it excluded.
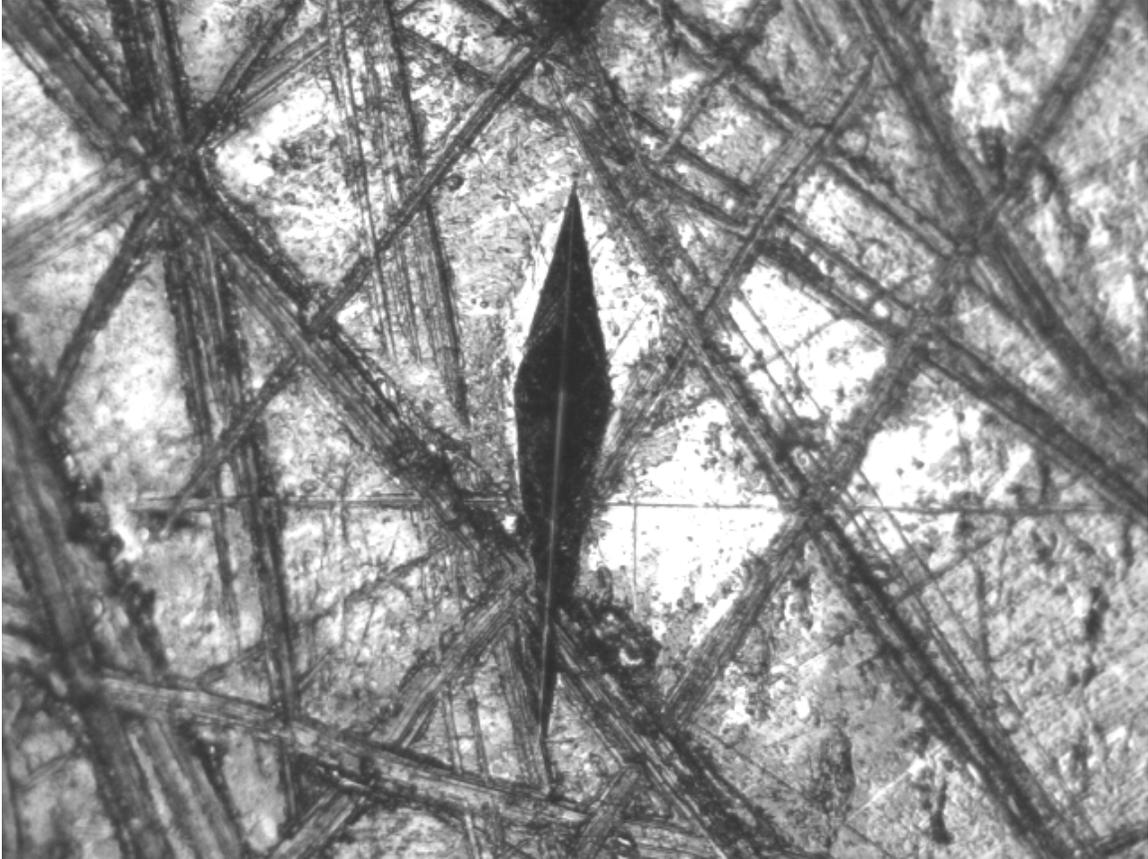
Figure 5. Optical micrograph of the surface of Sample 3, showing scratches and Knoop indentation. The Knoop indentation is about 220 microns long.

Table 2. Knoop Hardness. There is an unknown visual uncertainty.

Sample 1, Irradiated			Sample 2, Irradiated			Sample 3, Unirradiated		
#	HK	Indent length	#	HK	Indent length	#	HK	Indent length
1	134.8*	(no record)	1	163.4	208.6 μm	1	157.1	212.8 μm
2	163.0	(no record)	2	161.7	209.7 μm	2	140.2	225.2 μm
3	163.0	209.5 μm	3	151.9	216.4 μm	3	150.6	217.3 μm
4	149.8	217.8 μm	4	155.9	213.6 μm	4	154.7	214.4 μm
5	159.1	211.4 μm				5	147.1	219.9 μm
6	171.1	203.5 μm				6	148.6	218.8 μm
						7	150.5	217.4 μm
						8	155.9	213.6 μm

Analysis

Samples 1 and 2 were analyzed together as 9 or 10 data points, depending on whether or not the outlier can be excluded. At first glance, it certainly seems that more hardness variation is present in the irradiated samples. Nonetheless, the outlier is very far out. The mean hardness of the irradiated samples is calculated at 157 ± 3 if it is included (mean HK^*_{rad}, rounded), and 160 ± 2 if it is not (mean HK_{rad}, rounded). The standard deviation of HK^*_{rad} is $\sigma^*(rad) = 9.6$, and the standard deviation of HK_{rad} is $\sigma\ (rad) = 6.3$. All analysis was done using the $1/(N-1)$ definition of σ, the standard deviation.

According to Chauvenet's Criterion if the number of measurements, N, times the probability of getting a measurement like the outlier is less than ½., then the data may be rejected. The outlier is $2.35\sigma^*(rad)$ from the mean, and the probability of getting a measurement like this is $(1 - 0.9812) = 0.0188$, according to the chart in Appendix A of [6]. $N \times 0.0188 = 0.188 < $ ½ , so the point certainly meets the criterion. Nonetheless calculations are done both with and without it.

The unirradiated measurements were more obedient. The mean hardness, HK_0, was 151 $\pm\ 2$; the standard deviation of the measurements, $\sigma(0)$, was 5.1.

The difference between HK^*_{rad} and HK_0 is, using the un-rounded numbers:
$(157.33 - 150.59) \pm \surd(\ 3.19^2 + 1.82^2) = 6.74 \pm 3.76$. Since $6.74\ /\ 3.76 = 1.84$, the difference is outside 1.84σ, where σ here represents the probability that this difference could be obtained through random statistical variation. According to the chart in [7], there is a 6.6 % chance of this, or equivalently, that it will happen one time in fifteen.

The difference between HK_{rad} and HK_0 is, using the un-rounded numbers:
$(159.83 - 150.59) \pm \surd(\ 2.21^2 + 1.82^2) = 9.24 \pm 2.86$. Since $9.24\ /\ 2.86 = 3.23$, the difference is outside 3.23σ, where σ here represents the probability that this difference could be obtained through random statistical variation. According to the chart in [6],

there is less than an 0.2 % chance of this, or equivalently, that it will happen less than one time in five hundred.

These results suggest that the difference in hardness is significant and that irradiation hardening did occur.

Conclusions

The appearance of radiation hardening in this sample was not entirely expected, because thermal neutron flux through these samples was about 3×10^5 times less than neutron flux through samples cited as irradiation hardened[2,3]. One possible conclusion is that alphas, betas, gammas, and fast neutrons do more damage to vanadium than was previously thought. In the first three cases, ionization of sample atoms may be the culprit.

The probable presence of irradiation hardening after 1 hr in the Washburn reactor casts doubt on vanadium's suitability as an ADS beam window material, because the radiation conditions in ADS are likely to be much more stringent. The exception would be if vanadium is capable of annealing most radiation damage at its ideal operating temperature of 500°C (see Materials Section).

Suggestions for other experiments along these lines include:

1. The samples should be polished to a mirror-sheen and tested for flatness prior to irradiation.
2. The experimenter should be familiar with the operation of a Knoop hardness tester, and be able to use it without supervision. This will allow many more data points to be collected.
3. In the case of statistically significant results, the irradiated and unirradiated samples could be heated to see if the hardness difference was reduced by annealing. This experiment would require special precautions because of the possibility of radioactive material evaporating from the samples.
4. Other materials could be tested. In particular, alloy systems may not have been studied.
5. The exact mechanisms of hardening could be studied.

References for Section 6

[1] **Journal of Nuclear Materials**

Volume 356, Issues 1-3 , September 2006, Pages 287-299

Vladimirov, Moslang. "Irradiation Conditions of ADS Beam Window..:"

[2]   He, Tang, Hesegawa, Abe "Microstructural development and irradiation hardening

of W and W-Re…" September 2006. stacks.iop.org/NF/46/877

[3] Miller. *Tantalum and Niobium*, New York Academic Press, 1959

[4] **Materials Science and Engineering A**

Volumes 309-310, 15 July 2001, Pages 198-202

Rodney, Martin, Brechet. "Irradiation Hardening by Interstitial Loops…"

[5] Steve Laflamme, WPI Reactor Operator

[6] Taylor. *Intro. To Error Analysis*, 2nd Ed., University Science Books, 1997