Assessing Criteria to Pick Ideal Moderators for Nuclear Fission Reactors

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Abstract

The goal of this study is to find the best and most feasible compound(s) to use in a nuclear reactor as a moderator in order to achieve a chain reaction with maximum efficiency. To obtain a chain reaction with the most energy output per volume material used, neutrons need to be able to collide with radioactive material in a manner that will maximize the probability of fission. For this to happen, the cross section for fission must be exceptionally large. This is attainable by slowing down the speed of neutrons by use of a moderator. A number of criteria must be met for a moderator to be considered the most feasible. For one, it must be able to quickly thermalize neutrons from the MeV range down to a few eV. In this paper, we will define thermalization as reducing a neutron's energy from 2 MeV to 0.025 eV [3]. Second, it mustn't have a high affinity for absorbing neutrons. And lastly, it must be cheap and abundant. If these criteria are met, we have found a good moderator.

Various studies have been conducted in this field, and the results vary depending on the type of reactor at the center of the experiments. For fast reactors, the most common coolant used at is $Helium_(g)$ (4He) due to its low density. As a result, only a few collisions between high-energy neutrons and moderator occur, which leads to the sustained high-energy level of the neutrons, which is favorable for the compound nucleus formation process. In thermal reactors, the common consensus is that light water, heavy water, and graphite are best at slowing down high-energy neutrons to room temperature in the least number of collisions. Water and graphite are further aided by their relatively high density and favorable collision kinematics.

This study will primarily explore to substances: Zirconium Hydride and Yttrium Hydride, and their abilities to act as moderators for slow water reactors.

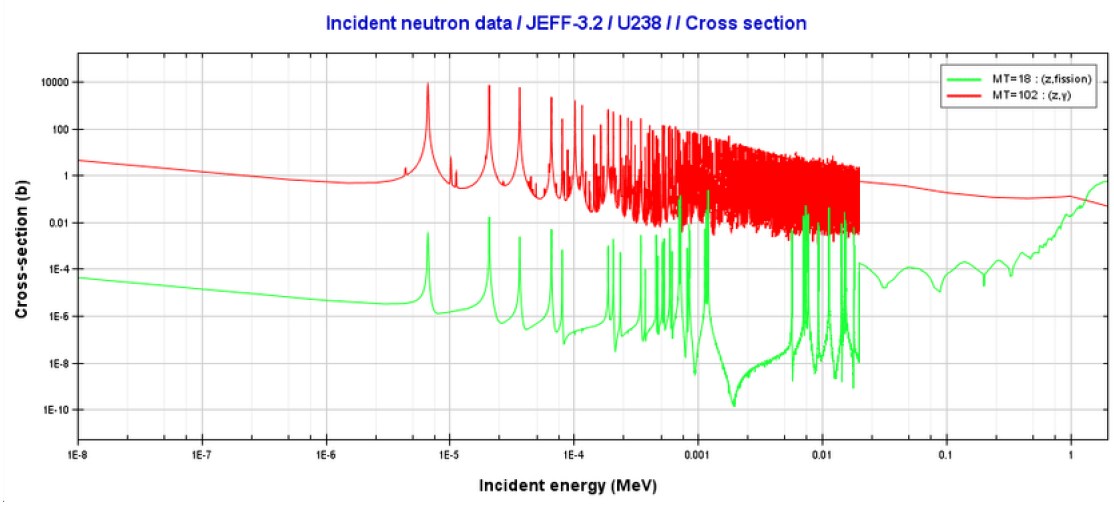
Contents

1 Metrics for picking ideal moderators

In order to definitively compare different compounds, some mathematical quantities have to be introduced. They will serve as unbiased method of comparing one moderator's quantitative abilities to another. Some important relations to note are the Resonance Escape Probability, Moderating Ratio, Logarithmic Energy Decrement, and Mean Free Path, Attenuation Length

1.1 Resonance Escape Probability

The resonance escape probability defines the likelihood that a high-energy neutron will survive to become a low-energy neutron. The most common fuel atom, U-238, has many resonance peaks from 6 eV to 200 eV which hinder many mid-energy neutrons from reaching full thermalization. This process is unfavorable because U-238 is a fertile material, meaning that it does not undergo nuclear fission directly as opposed to U-235. In order to maximize the resonance escape probability, we need to obtain a lot of collisions in a very short time span, and each of these collisions must result in a substantial energy loss.



1.2 Moderating Ratio

The moderating ratio is a direct measure of a material's effectiveness of a moderator. It is defined by (1), where ξ is the Logarithmic Energy Decrement(1.3), Σ_s is the scattering cross section, and Σ_a is the absorption cross section.

$$\xi * \Sigma_s / \Sigma_a \tag{1}$$

We can define the scattering cross section as the probability that a neutron will be expelled out of a nuclei at a lower energy than when it was absorbed. The absorption cross section can be defined as the probability that a neutron will be captured by a nucleus. Since scattering cross sections will decrease the energy of incident neutrons they are favorable and hence multiplied. to ξ. Absorption events are unfavorable because they remove the total amount of neutrons that are available to create fission events, hence it is divided. Not all absorption is detrimental, however, as some neutron capture can prevent runaway reactions from occurring so we must look at more than just the Moderation Ratio and the absorption cross sections.

1.3 Logarithmic Energy Decrement

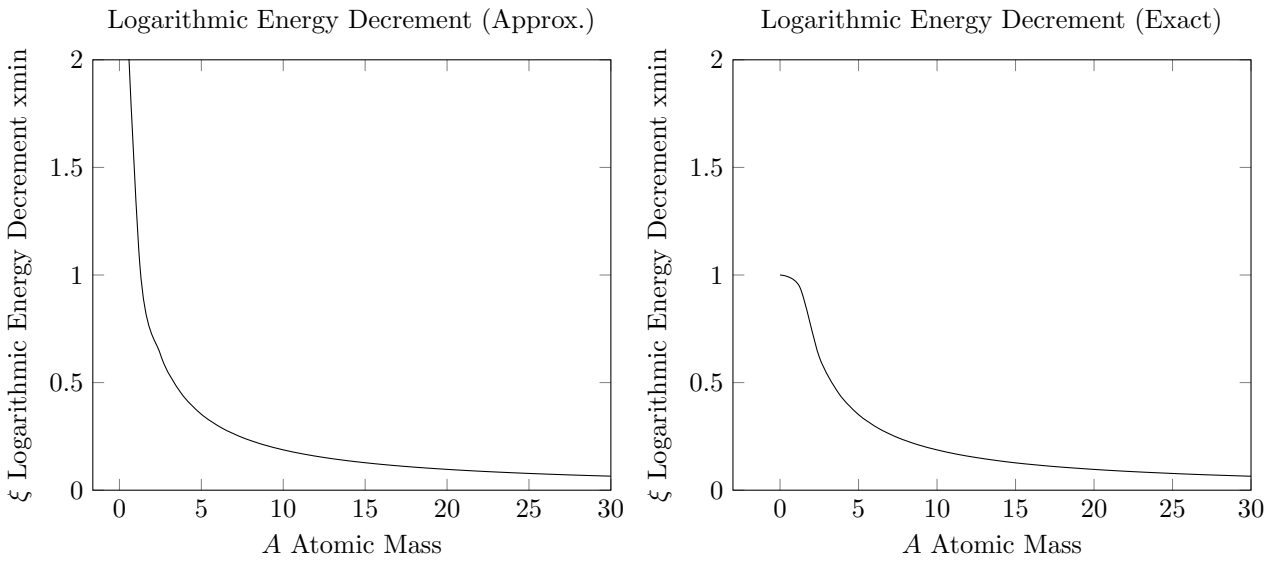
As mentioned in the supertopic, the logarithmic energy decrement is another metric of measuring moderator effectiveness. It is a value from 0 to 1 that describes how a moderator particle will slow down a neutron. It is not dependent on the energy of the incident neutron, it is an intrinsic property of the nucleus. It is given by 2a

$$\xi = 1 + \frac{\alpha}{1-\alpha} \ln \alpha \tag{2a}$$

$$\alpha = \frac{(A-1)^2}{(A+1)^2} \tag{2b}$$

It can be approximated by 3

$$\xi = \frac{2}{A + 2/3} \tag{3}$$



We can use ξ to calculate the number of collisions a moderator needs to undergo in order to reach a certain energy given that it starts at another energy. This equation is given by 4

$$n = \frac{1}{\xi} \ln \frac{E_0}{E_n} \tag{4}$$

We can also calculate the amount of time it takes for a neutron to thermalize using 2a and the scattering cross section.

$$t = \frac{\sqrt{2mc^2/E_f}}{c\xi\Sigma_s} \tag{5}$$

1.4 Mean Free Path

The intensity of a neutron beam colliding with a thick slab can be given by the following equation

$$I(x) = I_0 e^{-N\sigma_t x} \tag{6}$$

where I_0 is the intensity of the beam before it enters the sample, N is the atom density, and x is the distance traveled. We define the macroscopic cross section as $\Sigma_t = N\sigma_t$. In a compound like water, the macroscopic cross section is simply the addition of the individual cross sections of each element. We can define the probability of interacting within x as

$$P(x) = I(x)/I_0 = e^{-\Sigma_t x} \tag{7}$$

The mean free path is defined as the distance traveled by a neutron between two collisions (λ). We can calculate it by finding the average value of x –the distance traveled without a collisions over dx.

$$\int_0^\infty x * P(x)\, dx \;\; = \int_0^\infty x * e^{-\Sigma_t x}\, dx \;\; = 1/\Sigma_t \tag{8}$$

A shorter mean free path is more desirable as it means that reactions are likely happening more quickly.

1.5 Attenuation Length

What is sometimes more useful is the attenuation length, given by

$$e^{-x/\lambda} \tag{9}$$

λ is not to be confused with mean free path; these are two different quantities. This lambda is a material property and is energy dependent. It is the distance that a neutron has traveled into a material when the probability that a particle has not been absorbed equals 1/e, or around 63%.

2 Types of Moderators

2.1 Moderator Compounds Used in Fission Reactors

Our current norms for nuclear moderators are based upon the type of reactor. Slow reactors will often use light water, heavy water, or graphite, while fast reactors commonly use moderator and use compounds that act as coolants such as liquid sodium and helium. Fast reactors, or breeder reactors, are designed with the purpose of refining material, more specifically, turning U-238 into P-239 – a weapons grade material.

Common slow reactor moderators			
Compound	Neutron Scattering Cross Section (barns)	Neutron Absorption Cross Section (barns)	Moderating Ratio
Light Water H_2O	49	0.66	68
Heavy Water D_2O	10.6	0.0013	4150
Graphite (C)	4.7	0.0035	212

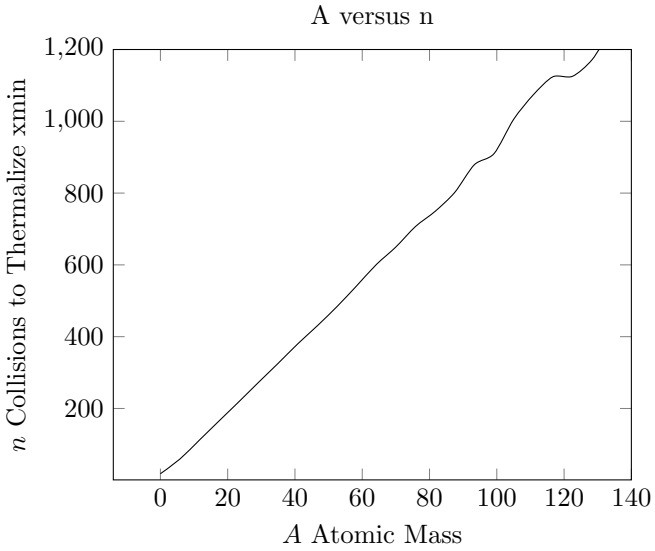
We see that light water has the greatest scattering cross section of the three; the probability of a scattering collision is large. But as a trade off, light water has an absorption cross section 200-500x larger than other proposed moderators.

Mathematically, it would be logical to use heavy water in every slow reactor across the world, but practically we run into issues. Light water is much more cheaper than heavy water and is much more abundant. So even though the moderating ratio of heavy water is 80x less than that of light water, it costs much more than regular light water. Graphite runs into the similar problems as even though it has a higher moderating ratio than water, it is simply not as cost effective as light water. Light water can act as both a coolant and a moderator, while graphite needs a separate coolant due to having a low specific heat (Water: 4184 J/kg· K, Graphite: 707J/kg · K)

The following table gives the number of collisions required to thermalize a high-energy neutron for common compounds found in a nuclear reactor.

Average number of collisions required to thermalize a high-energy neutron (2 MeV to 0.025 eV)		
Element	Atomic Weight	Number of Collisions [5]
Water	18	20
Hydrogen	1	27
Deuterium	2	31
Heavy Water	20	36
Helium	4	48
Beryllium	9	92
Carbon	12	119
Uranium	238	2175

From the table we see that water is one of the best moderators due to its very low number of collisions required for thermalization. The reason hydrogen falls behind water is because water contains two hydrogens, which are the most favorable element for moderation. The graph below compares the atomic mass of an element to the number of collisions that would be required for thermalization. There is a roughly linear relationship as atomic mass increases.



As a coolant, Helium plays a unique and necessary role in breeder reactors with its chemical and physical qualities. Since it is a noble gas, it is chemically inert and will not react with surrounding chemicals. It also has a high specific heat of around 5193 $\frac{J}{kg*K}$, a value even higher than water. Lastly, at high neutron energies, it has a large elastic scattering cross section and a relatively low neutron capture cross section – making it perfect for the environment of a breeder reactor. The best way to visualize helium's role in a breeder reactor is as a low-efficiency moderator that will control the temperature of a cage.

2.2 Propose New Moderators

Metal hydrides have become an appealing option for nuclear moderators. Compounds such as Yttrium Hydride (YH_x) and Zirconium Hydride (ZH_x) can act as alternatives to water. With their ability to pack hydrogen at high densities, these compounds are able to moderate fast neutrons down to thermal energies quickly. These metal hydrides are able to exist at these high temperatures at a decent H/Metal ratio without extreme pressures, meaning that they can act as great moderators.

"Properties of metal hydrides having potential nuclear applications" [7]		
Compound	Slowing Down Power	Moderating Ratio
TiH_2	1.85	6.3
ZrH_2	1.45	55
LiH	1.2	3.5
YH_2	1.2	25
ThH_2	1.0	5.2
$ThZr_2H_7$	1.55	14
$ThTi_2H_6$	1.8	6

They also have favorable absorption cross sections. Yttrium's absorption cross section for thermal neutrons (0.025 eV) is around 1.28 barns while Zirconium's isotopes average less than 1 barn.

2.3 Advantages and Drawbacks Proposed Moderators

Compared to elements like Boron that have cross sections that stretch for hundreds of barns radially, the two metals will do very little to interfere in the thermalization process. Additionally they are impacted very little by impurities. The process to create pure yttrium involves hydrofluorination and calcium reduction, the by products of which do not significantly impact Yttrium's ability to moderate.

$$Y_2O_3 + 3F_2 \rightarrow 2YF_3 + 1.5O_2 \tag{10a}$$

$$2RX_3 + 3Ca \rightarrow 3CaX_2 + 2R \tag{10b}$$

Heat of Reaction, Free Energy of Reaction, and Equilibrium Constant for the Reaction 10a [4]			
Temperature (K)	Heat of Reaction (Kcal/mol Y_2O_3)	Free Energy of Reaction (Kcal/mol Y_2O_3)	Equilibrium Constant
298	-374	-359	$3.98 * 10^{263}$
400	-373	-354	$6.82 * 10^{193}$
500	-372	-350	$1.15 * 10^{153}$
600	-371	-345	$8.61 * 10^{125}$
700	-370	-341	$4.01 * 10^{106}$
800	-369	-337	$1.40 * 10^{92}$
900	-368	-333	$8.61 * 10^{80}$
1000	-367	-329	$9.93 * 10^{71}$

As shown, the preparation of Yttrium oxide for calcium oxidation is a very thermodynamically favorable. The heat released by the reaction is so great that it trumps the tiny bit of entropy lost in the reaction, resulting in a very large gibbs free energy, and therefore a very large equilibrium constant. This implies that the reaction is spontaneous and does not require an energy input to yield products, further emphasizing the ease of conducting the reaction to refine Yttrium. Since the reaction is so dangerously spontaneous, it occurs at an unimaginably great speed. Most of the final product is generated by 10 minutes. Distilling water, however, takes a lot more time as it typically enters the flask droplet by droplet. Completion times may vary.

The only issue with reaction 10a is its release of too much energy. The huge amount of heat that is manumitted is difficult to control and may cause damage to the setup. Additionally, the fluorine gas must be kept at really high pressures (due to Le'Chatelier) and is naturally corrosive, meaning that there are many safety measures that must be taken: including the automation of some parts of the reaction and the remote activation of other parts. As a result, the cost to built a facility to refine Yttrium is way more expensive than purifying water. The heat released in the reaction can get so high that some of the fluorine product will melt, but this process is largely insignificant. The process of distilling water does not need as much safety features and is relatively inexpensive.

The reduction of YF_3 in reaction 10b occurs at a temperature of around $1550°C$ for around 5 minutes. According to the study conducted by Daane and Spedding [1], the conducted 3 trials: one with 3.5 grams of YF_3 and another with 35 grams of YF_3. Eventually they were able to reach a yield of $99\% \pm \frac{1}{2}\%$ with both amounts of YF_3. The tantalum content (impurity) was able to be reduced to around 500ppm at the cost of 5% yield. Which the methods mentioned in the study, we can harvest largely pure Yttrium at high quantities at a time, making the metal more accessible to be used in nuclear reactors.

Any impurities created in the process of these two reactions is largely negligible and do not impact the ability of YH_X as a moderator.

The process of hydrogenating Yttrium can be challenging, however. In the hydriding process of Yttrium, hydrogen enters the lattice structure of the Yttrium, causing it to expand. At the same time, the hydrogen forms a hydride case around the Yttrium, and the Yttrium's expansion causes the whole structure to crack. By heating up Yttrium [8] (1000°) and funnelling H_2 through a vacuum chamber and then a very small retort, the reaction was able to be controlled and produce crack-free Yttrium Hydride that is usable for moderation. When comparing Y to Zr, it's important to take into account their behaviors at high temperatures like those of nuclear reactors. At $1,100°$C Y has 2.6 times the hydrogen content as Zr (when at equilibrium with 1 atm H_2) [7]. In short, ZrH_X requires much more processing through methods, such as metal cladding, self-protecting oxide, or nitride layers to even compete with YH_X. Yttrium will retain its hydrogen at a much higher rate, which gives it favorable kinematics. At higher temperatures, Y's desorption of hydrogen becomes non-negligible.

To combat thermal expansion and other processes that are prevalent at high temperatures, the previously mentioned metal cladding 2.3 technique can be used, however at the temperatures of a nuclear reactor, the cladding metals themselves start to expand at exorbitant rates. Glass-enamel coating might be a more sound option as results indicate that its permeability to hydrogen is 10x less than regular cladding, but the results are largely not reproducible which lowers its credibility. As temperatures increase, the evasiveness of hydrogen in the metal-cladding system increases 2.3, a problem not present in water as the chamber is pressurized. The nitride and oxide layers are a little better, but still not great as hydrogen losses are reduced, but its overall impact is very small [6].

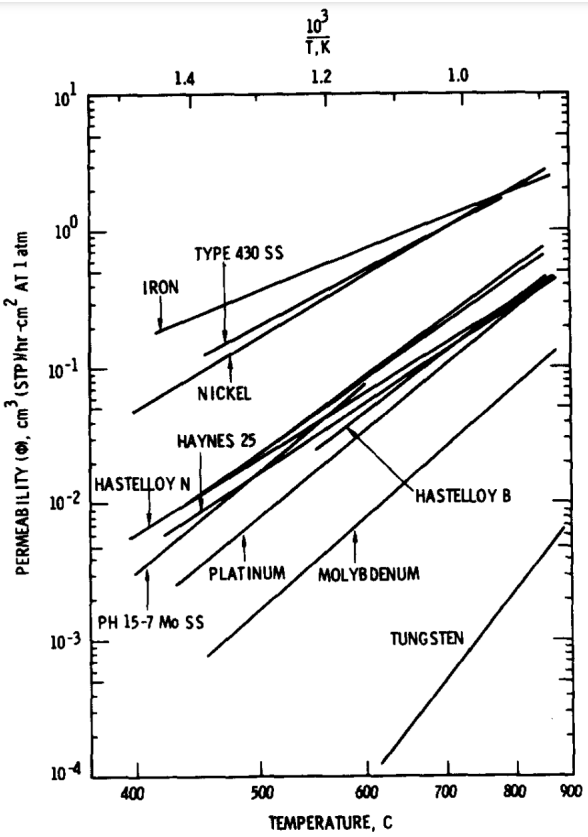


Figure 1: Permeability of metal-cladding metals to hydrogen [6]

However, what the nitride and oxides layers can do is increase the thermal conductivity.

2.4 Comparing Current Norms to Proposed Norms

In crack free Yttrium and/or other heavy metals, we can observe its true density of hydrogen and compare it to other moderators. We see that Yttrium comes close to water when it comes to atomic density, and ZrH_2 is even better than water, reaching a density of up to 7.7 in a perfect lattice. Combined with Zirconium's and Yttrium's relatively low tendency to absorb thermal neutrons, they are both great choices for nuclear reactors. The only barrier that might stop them is the price. Zirconium of about 99.6% purity costs around $150 per kg, and Yttrium costs around $35 per kg [2], both of which are massive compared to purified water.

Density of Hydrogen in Various Compounds [7]	
Compound	$10^{22} atoms/cm^3$
TiH_2	9.1
ZrH_2	7.3
LiH	5.8
YH_2	5.8
ThH_2	4.9
H_2O	6.6
$ThZr_2H_7$	7.7
$ThTi_2H_6$	8.8

Issues arise when we take into account the temperature dependency of various quantities. In order for water to be effective it mustn't boil, therefore it must be kept at a very high temperature. Zr and Y, however, have a few previously mentioned methods (other than pressurizing the cell) to reduce their

permeability of hydrogen, on top of their high resistance to fluctuations in temperature.

Comparing their moderating ratios, water has an MR of 68, ZrH_2 has an MR of 55, and YH_2 has an MR of 25. While YH_2 is the lowest of them all, it is arguably the best choice as a moderator. The different MRs of Zr and Y at different H concentrations was not collected, so optimizing the ratio of Y to H is still in the question. Additionally, it survive up to $1200°C$ without much effect on its ability to moderate, while Zr breaks at around $900°C$ and water requires extreme pressurization at its boiling point.

3 OpenMC Simulation

The following section will detail a pin-cell simulation ran in OpenMC. The liberties taken are as follows: the reactor is estimated as a sphere, the cladding is made of Zirconium for all trials, and the fuel is 3% enriched UO_2. 100 trials were run per test case and the values were calculated accordingly.

Test Case 1: Control (No Moderator)

```
============================>     RESULTS     <============================

k-effective (Collision)     = 0.58371 +/- 0.00195
k-effective (Track-length)  = 0.58401 +/- 0.00196
k-effective (Absorption)    = 0.59054 +/- 0.00304
Combined k-effective        = 0.58570 +/- 0.00172
Leakage Fraction            = 0.00000 +/- 0.00000

============================>     TALLY 1     <============================

Cell 2
  U235
    Total Reaction Rate                 0.893149 +/- 0.00243989
    Fission Rate                        0.153673 +/- 0.000606184
    Absorption Rate                     0.20162 +/- 0.00086256
    (n,gamma)                           0.0479467 +/- 0.000260573
```

Test Case 2: Light Water

```
============================>     RESULTS     <============================

k-effective (Collision)     = 1.40457 +/- 0.00465
k-effective (Track-length)  = 1.40890 +/- 0.00553
k-effective (Absorption)    = 1.40147 +/- 0.00369
Combined k-effective        = 1.40293 +/- 0.00339
Leakage Fraction            = 0.00000 +/- 0.00000

============================>     TALLY 1     <============================

Cell 2
  U235
    Total Reaction Rate                 0.733635 +/- 0.00276656
    Fission Rate                        0.550799 +/- 0.00226682
    Absorption Rate                     0.661246 +/- 0.00267846
    (n,gamma)                           0.110447 +/- 0.000427015
```

Test Case 3: ZrH_2

```
============================>     RESULTS     <============================

k-effective (Collision)     = 1.14884 +/- 0.00429
k-effective (Track-length)  = 1.14576 +/- 0.00471
k-effective (Absorption)    = 1.15367 +/- 0.00349
Combined k-effective        = 1.15111 +/- 0.00311
Leakage Fraction            = 0.00000 +/- 0.00000

============================>     TALLY 1     <============================

Cell 2
  U235
    Total Reaction Rate                 0.60299 +/- 0.00232696
    Fission Rate                        0.448116 +/- 0.00192403
    Absorption Rate                     0.539103 +/- 0.00223698
    (n,gamma)                           0.0909877 +/- 0.000324006
```

Test Case 4: YH_2

```
============================>     RESULTS     <============================

k-effective (Collision)     = 1.20280 +/- 0.00359
k-effective (Track-length)  = 1.20114 +/- 0.00485
k-effective (Absorption)    = 1.19991 +/- 0.00283
Combined k-effective        = 1.20071 +/- 0.00272
Leakage Fraction            = 0.00000 +/- 0.00000

============================>     TALLY 1     <============================

Cell 2
  U235
    Total Reaction Rate                 0.640991 +/- 0.00241382
    Fission Rate                        0.467738 +/- 0.00199181
    Absorption Rate                     0.565666 +/- 0.00233426
    (n,gamma)                           0.0979279 +/- 0.000359543
```

To prevent a runaway reaction, a combined k-effective closest to 1 is most desirable as it means that the neutrons in each generation are not multiplying out of control, but rather replacing each other to maintain the total number of neutrons. Zr and Y both have a k-effective closer to 1 than light water.

Since light water is producing more neutrons per generation, it is producing more fission events and therefore yields a higher fission rate. Conversely, since Zr is producing the least amount of neutrons, it will have the smallest fission rate

According to the outputs of the simulation, light water is likely the optimal moderator in terms of power output, but Zr and Y are safer to be used in a fission reactor.

4 Conclusion

If we take all of these factors into account, YH_X seems to be the best moderator for slow reactors. With current technology that allows us to make quality yttrium hydride in desired quantities, the

issue of cost is hardly an issue. If yttrium hydride were to become more widely recognized as a more suitable moderator, more purifying facilities would be built and eventually, the cost would be driven down, resulting in a compound that would be economically comparable to water. Barring cost, the moderators ranked in order from least suitable to most suitable go as follows: ZrH_X, H_2O, and YH_X.

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