Cross Calibration for using Neutron Activation Analysis with Copper Samples to measure D-T Fusion Yields

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Abstract

We used a dense plasma focus with maximum neutron yield greater than 10^{12} neutrons per pulse as a D-T neutron source to irradiate samples of copper, praseodymium, silver, and lead, to crosscalibrate the coincidence system for using neutron activation analysis to measure total neutron yields. In doing so, we counted the lead samples using an attached plastic scintillator, due to the short half-life and single gamma decay. The copper samples were counted using two 6" NaI coincidence systems and a 3" NaI coincidence system to determine the total neutron yield. For the copper samples, we used a calibration method which we refer to as the "*F* factor" to calibrate the system as a whole and used this factor to determine the total neutron yield. We concluded that the most accurate measurement of the D-T fusion neutron yield using copper activation detectors is by using 3 inch diameter copper samples in a 6" NaI coincidence system. This measurement gave the most accurate results relative to the lead probe and reference samples for all the copper samples tested. Furthermore, we found that the total neutron yield as measured with the 3 inch diameter copper samples in the 6" diameter NaI systems is approximately $89 \pm$ 10 % the total neutron yield as measured using the lead, praseodymium and silver detectors.

Introduction:

Motivation:

 The National Ignition Facility (NIF) at Lawrence Livermore National Laboratory is the epicenter for research done in inertial confinement fusion (ICF). At NIF, the research into fusion is being done by laser-driven ICF, powered by the 1.8 MJ ultraviolet laser system with peak power of 500 TW. This system is focused onto a 2 mm diameter pellet, enclosed in a cylindrical gold hohlraum with the pellet filled with a fuel mixture of deuterium and tritium, either as a gas or a thin cryogenic layer^[1]. One of the methods used to determine the yield of the pellet implosion is neutron activation analysis. For total neutron yields greater than 10^8 , nuclear activation is commonly used due to its ability to accurately measure high neutron yields^[2]. In nuclear activation analysis, the high-energy neutrons produced by fusion interact with a material, resulting in the production of an unstable nuclear state with a measurable decay product.

 One of the most common materials for neutron activation analysis of D-T fusion neutrons is copper, which due to its high threshold energy, decreases the number of neutrons from other reactions and scattering that could be detected. Both naturally occurring isotopes of copper undergo a (n,2n) reaction resulting in a positron emitter. Upon the annihilation of the positron, this produces two 511 keV gamma rays that can be detected with a coincidence system using two NaI(Tl) detectors. The primary isotope of copper that we are interested in is Cu-63 due to the production of Cu-62 in the reaction ${}^{63}Cu(n,2n){}^{62}Cu$ and the short half-life of 9.74 minutes for Cu-62. This compares to the half-life of 12.7 hours for the Cu-64 produced in the reaction ${}^{65}Cu(n,2n)$ ⁶⁴Cu^[3,4]. As a result of the shorter half-life and greater atom fraction, the activity of the Cu-62 radioisotope will be greater than that of the Cu-64, so the number of counts detected will be more statistically significant above background. On the other hand, the activity of the

Cu-64 would only be marginally significant above the natural background radiation that registers in the detectors. The second reason that the ⁶³Cu(n,2n)⁶²Cu(β ⁺) reaction is of interest is that all the Cu-62 that is present in the sample comes from that reaction, whereas there is Cu-64 formed by neutron capture in Cu-63^[3] along with the Cu-64 from the $(n,2n)$ reaction, thereby complicating the interpretation of the ${}^{65}Cu(n,2n)$ ⁶⁴Cu results.

Theory:

The fusion of deuterium and tritium has been considered the most viable method for achieving ignition since the beginning of research into nuclear fusion. This is because it has the greatest cross section for fusion of most viable fuels, and the cross section peaks at the lowest energy, such that temperatures greater than 4-5 keV can achieve ignition . The fusion of deuterium and tritium results in the production of an alpha particle and a 14.1 MeV neutron^[5].

 $d + t \rightarrow 4$ He + n + 17.6 MeV

Although fusion is possible at thermal temperatures, to produce a measurable number of fusion neutrons, the energy of the system must be increased. This can be achieved by accelerating particles or adding heat to the overall system. This increase is necessary because the fusion cross section is nonlinear with respect to particle energy with a thermal cross section less than 10^{-4} barns. By accelerating deuterons with an ion beam and directing the beam upon a tritium doped target, such as Erbium tritide, the cross section grows significantly reaching a peak at deuteron energy of 107 keV with the cross section being 4.95 barns^[6].

 In contrast to the use of deuterium and tritium, the other two commonly used fuel mixtures are far less advantageous for the attempt to reach ignition conditions in nuclear fusion

reactions. These fuel mixtures are pure deuterium, which reacts in a D-D reaction producing either tritium or helium-3, and deuterium and helium-3 which fuse to produce helium- $4^{[7]}$.

$$
d + d \rightarrow t + p + 4.03 \text{ MeV}
$$

$$
d + d \rightarrow {}^{3}\text{He} + n + 3.27 \text{ MeV}
$$

$$
d + {}^{3}\text{He} \rightarrow {}^{4}\text{He} + p + 18.3 \text{ MeV}
$$

The benefits of these reactions are that the energy output in neutrons is significantly lower than that for D-T and that they do not require a tritium containment system. However, the amount of energy necessary to achieve fusion is significantly higher, and the fusion reaction rate is a fraction of that for D-T. For acceleration using an ion beam, with deuteron energy of 100 keV, the cross sections for both D-D and D- 3 He fusion are around 20 millibarns, or $1/250th$ the cross section for D- $T^{[6]}$. As a result, experiments for using these fuels to achieve ignition are nonexistent, but the reactions are used to understand the fundamental physics of nuclear fusion and its associated engineering challenges.

The two common methods to irradiate samples with neutrons with energy of 14.1 MeV, such as D-T fusion neutrons, are the use of an ion beam or a dense plasma focus (DPF). Using an ion beam, such as the Sandia Ion Beam Laboratory, generally entails firing a deuteron beam at targets fabricated from a rare earth metal doped with tritium, such as erbium-tritide $(ErT_2)^{[8]}$. This method is effective because it allows for associated particle counting in the ion beam. Through the use of associated particle counting, a near absolute measurement of the ion flux and total yield can be made. The use of an ion beam also allows for a selection of the deuteron energy with a velocity selector. This allows for the focusing of only those deuterons with approximately optimum energy on the target, rather than having a wide range of deuteron energies and fusion cross sections.

 Similar to the use of an ion beam for producing the fusion neutrons necessary for calibration of the (n,2n) reaction in copper, a DPF will act as a neutron source by focusing and heating the plasma in either a *z* or *θ* pinch. However, using a DPF to irradiate the sources only allows for a known maximum energy and plasma current due to the construction of the device. As a result, the total yield cannot be calculated directly unless the DPF undergoes a perfect discharge at both maximum energy and plasma current^[9].

A DPF works by passing a strong electric current through a plasma, which in turn creates a strong magnetic field that compresses the plasma and locally heats the plasma to fusion temperatures^[10]. The Mather-type DPF is designed as a system with two coaxial electrodes, such that by inducing an intense electrical discharge between them, the induced magnetic field in the plasma radially compresses ("pinches") the plasma about the z-axis. This creates a *z* pinch at the open end, which is treated as a point neutron source, as shown in Figure $1^{[9]}$.

Figure 1: Diagram showing radial compression and formation of *z* pinch at open end of electrodes in plasma focus^[9]

DPF's made with deuterium or D-T plasmas are commonly used as neutron sources due to the relatively uniform heating within the plasma. As this is the case, to achieve D-T fusion, the average particle energy in the pinch needs to be greater than 10 keV due to the nature of the DPF as a pulsed device that rapidly compresses and heats the system. Because a DPF can produce controlled neutron fluxes ranging from the order of 10^4 neutrons per shot to greater than 10^{12} neutrons per shot depending on the size of the device, they are commonly used for neutron activation experiments, along with radiography and other uses if configured to release x-ray pulses. As D-T DPFs are short pulse 14.1 MeV neutron generators, with a pulse lengths ranging from a few nanoseconds to microseconds, they are extremely beneficial in calibration of diagnostics for ICF experiments as they will mimic the neutron output from ICF. The second major benefit in using a DPF for calibration of ICF diagnostics is the required energy for operation, as a DPF will have a significantly higher neutron yield for a given amount of input energy $^{[11]}$.

 Neutron activation analysis relies on the reaction between neutrons and atoms of particular isotopes to produce a radioisotope with a known decay sequence. In neutron activation analysis, either the decay of the radioisotope or the emission of a characteristic gamma ray for the transition from a nuclear excited state to ground state is measured, allowing for the determination that a neutron was absorbed by the material^[12]. For fusion neutrons, using neutron activation allows for the focusing of a detector on a specific frequency corresponding to the characteristic radiation of the sample material. Neutron activation analysis is also used for determination of material quantities in a mixture based off the measurement of a broad spectrum with peaks corresponding to specific material characteristics. This method can be used for a wide range of neutron energies, including those produced by nuclear fusion.

In using neutron activation for measurements of high-energy neutrons, such as those produced by D-T fusion, it is advantageous to use a material that has threshold energy as close to 14 MeV as possible, provided that the cross section is large enough for significant activation to occur. In doing this, the number of scattered neutrons that interact in the material will be decreased and neutrons produced by most other methods would likely be below threshold. It is also important to use a material that has a relatively short half-life, as a long lived radioisotope will result in a decreased activity and require a longer counting interval. For D-T fusion, the materials that meet these characteristics and are commonly used are copper, lead, indium, and praseodymium. All of these materials have threshold energy for a (n,2n) reaction greater than the neutron energy from the D-D reaction as is given in Table 1. Another material that is being used at NIF is zirconium due to its 12 MeV threshold and high cross section. However, it has a half-life of 3 days and requires a long count time to give results that are statistically significant relative to the background radiation. Sliver is also commonly used for the measurement of neutrons due to its high cross section for neutron capture. However, the threshold energy for silver is very low and it predominantly measures neutrons that have slowed to thermal energies, making it undesirable if other neutron sources are present.

Table 1: Important properties pertaining to neutron (n,2n) reactions in isotopes commonly used for measurement of D-T fusion neutrons

To use a sample for neutron activation analysis and yield determination, the sample needs to be able to have the radioactivity of the product measured. As this is the case, the sample should have a half-life on the order of minutes if it is to be counted outside the target chamber, or be connected to a probe if it has a shorter half-life. For materials that will be counted outside the

target chamber, having positron emitters is advantageous because it allows for the use of two detectors in a coincidence circuit, which in turn allows for the accurate counting of low activities even with large background radiation. The detectors in this case should have the discriminators in the coincidence circuit set for the detection of the characteristic 511 keV gammas that result from electron-positron annihilation. The use of a coincidence circuit for measuring the 511 keV gammas is more effective than using a single detector for materials that have multiple decay products. This is because the discriminators can be set to only measure a window around the 511 keV photo-peak. Then, by knowing the fractional decay rate for positron decay, the total decay rate can be more accurately measured^[15]. For the materials given in Table 1, the $(n,2n)$ reaction for Pb-208 results in the meta-stable state of Pb-207, which then emits a single gamma to decay to the ground state of Pb-207^[16]. The rest of the materials in Table 1 all undergo positron decay and are effectively counted using a coincidence system to measure the annihilation photons. Silver primarily undergoes β decay and gamma decay, and is counted with an attached detector similar to that for lead.

 The use of copper for the detection of fusion neutrons is beneficial because of its relatively high threshold energy for a (n,2n) capture. Copper is also commonly used because of the short half-life of copper-62, which allows for a distinct calculation of the neutron yield. Also in copper, the resultant isotopes undergo β^+ emission, such that the 511 keV gammas that are produced by positron annihilation can be measured in a detector with a coincidence system. Because the copper samples are naturally occurring copper, they are comprised of both Cu-63 and Cu-65 with atom percentages of 69.15% and 30.05%, respectively. As there are two distinct radioisotopes that will be produced by (n,2n) reactions in the copper samples, it is important to determine the relative activity of the two isotopes for yield calculations (Eq. 1)^[17].

$$
A = \lambda N, \qquad \lambda = \frac{\ln(2)}{T_{1/2}} \tag{1}
$$

In equation 1, *A* is the activity of the sample, *N* is the number of molecules of the radioisotope present in the sample, and *T½* is the half-life of the radioisotope. To determine the relative activity of the Cu-62 and Cu-64, it becomes trivial to take a ratio between the activities of the individual isotopes.

 The most common practice used for the modeling of nuclear fusion experiments is the Monte Carlo method. The Monte Carlo method is a statistical process based off the drawing of random samples from specified probability distributions. In Monte Carlo, the samples are drawn as part of a random walk process with the probabilities recalculated after the drawing of each sample and the collection of samples from the random walk building up the probability function. In these samples, the Monte Carlo calculation effectively gives an estimate of the expected value of the estimating random variables. In doing this for neutron transport, the superposition principle allows for Monte Carlo calculations to be run in the different energy regions in a multigroup calculation of neutron energy. This allows for the superposition of solutions to the transport equation for neutrons at different energies and a complete picture of the neutron's interactions without having to use analytical methods^[18].

Experiment:

 For the experiments at National Security Technologies (NSTec), a Mather-type dense plasma focus with output on the order of 10^{12} neutrons was used. The DPF was mounted in a tritium contaminated pit with a concrete shield over the top, and operated using a deuterium and tritium fuel mixture for producing 14.1 MeV fusion neutrons. The source was aligned with the copper, praseodymium, lead and silver materials in a line of sight for irradiation.

 To determine the calibration of the copper system relative to the lead probe, silver detector, and praseodymium samples, it is necessary to calibrate the copper detector system. The calibration of the detector system determines the relationship between the total neutron yield and induced activation in the samples. The methods that could be used to calibrate the detector system are a derivation of the system response with fundamental quantities and the detector efficiency or the "*F* Factor" method developed by G.W. Cooper, C.L. Ruiz, *et. al.*[19] where the entire system is calibrated based off the specific sample geometry, irradiation geometry, and counting geometry. With these quantities modeled into the system, the *F* factor gives a calibration factor for the number of counts recorded per neutron, provided there is knowledge of the number of neutrons incident on the sample. Having done this, the factor can be used for future experiments in the same geometry without the need to know counting efficiencies, cross sections, etc. For the calculations in this experiment, the *F* factor method is used to calibrate the copper detector system for determining total neutron yields from the initial sample activity.

 In calibrating the system, there are certain quantities, such as escape probability for gamma rays in the samples, that must be either measured or modeled. For these quantities, modeling was done using MCNPX, which is the Monte Carlo N-Particle eXtended code developed by Los Alamos National Laboratory. This code was used to model the geometries and detector efficiencies given in the system for the determination of the *F* factor and total yield.

 After determining the calibration of the detector for the different sizes of copper samples used in the experiment, we compared the calculated yield for each shot to that from the lead probe, silver detector, and praseodymium sample. We chose to use copper samples because the (n,2n) reaction has the highest threshold energy, which results in the detection of the least inscattering for neutrons, and the material is abundant and relatively inexpensive compared to

praseodymium and other materials. From this, we determined whether the copper samples gave an accurate measurement of the neutron yield or if there was a persistent difference in the measured yield that could imply an error in the calibration method.

Methodology:

F Factor Method[19]*:*

The *F* factor method calibrates the entire system as a unit, rather than determining the calibration factor for the detectors and extrapolating it to the system. This method is beneficial because it eliminates the need to determine the amount of attenuation and scattering that would occur from the specific geometry of the system. This is the case because the geometry is taken into consideration when the *F* factor is measured. As a result, the system is calibrated for that specific geometry and amount of attenuation.

 To measure the *F* factor for this experiment, we consider the case where the production of fusion neutrons is approximately steady state. As such, the number of atoms, $N(t)$, of the given radioisotope present at an irradiation time, *t*, can be represented by

$$
N(t) = \frac{R\left(1 - e^{-\lambda t}\right)}{\lambda} \tag{2}
$$

where *R* is the production rate for the choice radionuclide. As *R* is determined by the parent material and the neutron flux, it can be written in terms of material quantities as

$$
R = \frac{\phi \varepsilon_A M N_A \sigma(E)}{A_W} \tag{3}
$$

where ϕ is the neutron flux in neutrons/cm² s incident on the sample, ε_A is the natural abundance of the parent material in atom fraction, *M* is the mass of the sample, *NA* is Avogadro's Number (6.022×10^{23}) , A_W is the atomic weight of the material, and $\sigma(E)$ is the cross section for the

neutron capture reaction at energy, *E*, in square centimeters. With the value of *R* known, it is possible to calculate the number of atoms of the desired radioisotope at the time when it was removed from the system, denoted as *t0*.

 With the number of atoms of the desired radioisotope known, the number of counts that would be expected over a time frame in the detector can be determined from the number of atoms present multiplied by the detector's counting efficiency, *εD*, the branching ratio for the desired decay, ε_B , and the proportion of the gamma rays that escape the sample rather than being absorbed, ε_S . Therefore, we can determine that over an interval $t_1 - t_2$, the number of counts, *C*, including background counts, *B*, which would be expected, is:

$$
C = \frac{\phi \varepsilon_A \varepsilon_D \varepsilon_B \varepsilon_S M N_A \sigma(E)}{\lambda A_W} \left(1 - e^{-\lambda t_0}\right) \left(e^{-\lambda t_1} - e^{-\lambda t_2}\right) + B \tag{4}
$$

If we assume that the number of neutrons emitted is constant, we can define the total neutron yield, *Y*, as the flux multiplied by the irradiation time times the solid angle, with *d* the distance from the neutron source to the sample, such that:

$$
Y = \phi t_0 4\pi d^2 \tag{5}
$$

With this yield, we define the calibration factor, *F*, as:

$$
F = \frac{(C - B)t_0 4\pi d^2 \lambda A_W}{Y M (1 - e^{-\lambda t_0})(e^{-\lambda t_1} - e^{-\lambda t_2})}
$$
(6)

and from this we get:

$$
F = \frac{\varepsilon_A \varepsilon_D \varepsilon_B \varepsilon_S N_A \sigma(E)}{A_W} \tag{7}
$$

 By knowing the *F* factor, calculations of the total neutron yield become simple. Therefore, to determine the total yield for a short pulse experiment, we can approximate $(1 - e^{-\lambda t_0})$ as λt_0 , resulting in the equation for the total yield being:

$$
Y = \frac{(C - B) 4\pi d^2 A_W}{F M (e^{-\lambda t_1} - e^{-\lambda t_2})}
$$
(8)

Procedure:

 In order to make measurements of the fusion neutrons produced, it is first necessary to set-up and calibrate the detectors and coincidence circuit^[20]. As the primary quantity of interest for measurement is the dual 511 keV annihilation photons, it is reasonable to use a detector system that consists of two detectors to measure both photons. Also, because the irradiated source is radioactive and the background counts will be decreased if properly shielded, it is important to have the counting system shielded. Within the counting system, there are two 6" diameter NaI(Tl) detectors located on opposite ends of the lead pig, with the placement of the sample to be counted going in the center of the pig as seen in Figure 2.

Figure 2: 6" diameter NIF coincidence system for copper activation showing NaI detectors, lead "pig" for shielding, and slot for insertion of copper samples

The data from the detectors is sent through a preamp followed by amplifiers to increase the amplitude of the signal for transmission to the coincidence circuit. The signals are then analyzed using single channel analyzers (SCAs) to select events depositing 511 keV in the NaI scintillator. The SCAs then emit a logic pulse to the coincidence unit, and if a 511 keV event is measured by both SCAs in coincidence an output signal is sent to a multi-channel scaler where the counts were recorded in time bins of 1 minute.

Figure 3: Schematic diagram of coincidence system and electronics for measuring 511 keV photons emitted from positron decay in copper samples

 Historically, the system was calibrated using a positron source, such as sodium-22 which undergoes positron decay 90% of the time, but also releases a 1.274 MeV gamma ray 100% of the time. As a result, the higher energy gamma can result in the coincidence system missing counts due to summing events from the 1.274 MeV gammas moving the count outside the discriminator window, resulting in a smaller calculated counting efficiency. This happens because the deposition of energy from the 1.274 MeV gamma at the same time as one of the 511

keV gammas registers in the detector as a single photon with energy 1.785 MeV. If the summing events are not taken into account, the measurement of the counting efficiency is off by about a factor of two. However, by using the *F* factor method, the counting system is calibrated to a copper sample, such that a Na-22 source is only necessary for setting the coincidence windows and measuring the detector efficiency. Because the summing events involving the 1.274 MeV gamma emitted by Na-22 complicates the measurement of the efficiency, we used a Ge-68 source, which has almost no summing events and gives an almost absolute of the detector efficiency for modeling of the efficiency for copper sources in MCNP.

 To take data at NSTec, the copper samples were attached to a train car, which was in turn attached to a tape measure, such that the samples were lowered a given length into the chamber for irradiation as shown in Figure 4. Also noted in Figure 4 are the locations of the lead, silver, and praseodymium as all the samples were irradiated at the same time for each shot.

Figure 4: 2-D schematic diagram of building containing DPF and detector locations relative to the DPF. All samples had a direct line of sight to the bulls-eye on the DPF.

This method was used because the DPF was mounted in a tritium-contaminated pit, and there were no other access points to place the samples for irradiation. Further complicating the irradiation of the sources was the cover over the DPF pit, such that there was only a small solid angle where the sources could be placed while maintaining a direct line of sight to the bulls-eye on the DPF (Figure 5).

Figure 5: View into DPF Pit. The samples were aligned with a direct line of sight to the bulls-eye, which marked the approximate location of the pinch in the source.

 Also, because the building housing the DPF was a radiation area, we had to set the coincidence systems up in the trailers used for controlling the DPF and have the sample activity measured by health physics prior to removal.

The D-T experiments were done by using a dense plasma focus provided by NSTec as a 14.1 MeV neutron source. The DPF used was a Mather-type pinch with maximum neutron yield of greater than 10^{12} neutrons per shot. The DPF operated using 133 kJ of stored energy with a

peak plasma current of 1.5 MA. For storing this energy prior to discharge, it was based off of 9 -27μ F capacitors charged with a Marx generator with potential 35 kV. The neutrons produced by D-T fusion reacted with the copper samples, which were then removed from the chamber and inserted into the counting system. To allow for the extrapolation of the number of atoms of the radioisotopes formed back to the initial time, the MCS software was triggered externally when the DPF fired.

 For the individual shots, the copper samples that were used had dimensions of 1 cm diameter with 1 cm thickness, 1 inch (2.54 cm) diameter with $\frac{3}{8}$ inch (.9525 cm) thickness, or 3 inch (7.62 cm) diameter with $\frac{3}{8}$ inch (.9525 cm) thickness. For these samples, the counting efficiencies of the detectors were modeled based on the geometry in MCNP. The modeling was done by Dr. Gary Cooper, using a method that combined the factors for the detector efficiency, ε_D , and self absorption, ε_S into a single factor. In doing this, it was determined that in the 6" diameter coincidence system for the 1 cm sample, the counting efficiency was 12.0%, for the 1 inch sample, the counting efficiency was 10.12%, and for the 3 inch sample the counting efficiency was 8.01%. For calibration using the 3" NaI coincidence system, the counting efficiency for the 3 inch sample was calculated to be 3.45%. Using these values, the *F* factor was calculated for the different samples and used to calculate the neutron yield of the given shot using Eq. 8.

Data:

 Using the DPF from NSTec, a total of 11 D-T shots were taken with data counted on either the 6" NSTec coincidence system or the 3" NaI coincidence system, with one additional shot having data taken only on the 6" NIF coincidence system. For the data taken using the

different multi-channel scalers, I was tasked with the data analysis for the measurement of the yield using the *F* factor method for the copper samples. Along with measurement of the yield, I made a comparison of the yield measurements for the different coincidence systems and sample sizes. The data from the praseodymium and silver was taken and analyzed by Tim Meehan and Chris Hagan of NSTec, with the measurements using the lead probe analyzed by Alan Nelson of Sandia National Labs. For these experiments, either two or three copper samples were irradiated on each firing of the DPF source. These samples were either counted solely in one of the detectors or counted sequentially in two of the detectors to get a cross calibration between detectors and different sample sizes. For analysis, it was most effective to plot the data for both samples after subtracting an estimate of the background counts caused by natural background radiation or the long-lived isotope, Cu-64. Subtracting the long-lived isotope is appropriate for this calculation because the ratio of counts between the two radioisotopes formed by neutron activation is small. It is also important because the activation in the Cu-64 cannot be directly calculated, due the production of Cu-64 by neutron capture in the Cu-63. The threshold for neutron capture is very low, such that a measurement of the yield from Cu-64 would be artificially high as the production from the neutron capture is difficult to calculate. The ratio of activation between the isotopes can be approximated by applying Eq. 1 to both samples, getting:

$$
A_{62} = \lambda_{62} N_{62}, \quad A_{64} = \lambda_{64} N_{64}
$$

\n
$$
\frac{A_{64}}{A_{62}} = \frac{\lambda_{64} N_{64}}{\lambda_{62} N_{62}} \approx \frac{\lambda_{64} \phi_n \sigma_{65} \varepsilon_{B65} N_{65}}{\lambda_{62} \phi_n \sigma_{63} \varepsilon_{B63} N_{63}} = \frac{\lambda_{64} \sigma_{65} \varepsilon_{A65} \varepsilon_{B65}}{\lambda_{62} \sigma_{63} \varepsilon_{A63} \varepsilon_{B63}}
$$

\n
$$
\therefore \frac{A_{64}}{A_{62}} \approx \frac{.00091 \times 910 \times .3085 \times .19}{.0712 \times 450 \times .6915 \times .98} = .0022
$$

Subtracting the approximated activation of the Cu-64 from the total activation, the activation for the Cu-62 can be approximated, which is then used in producing a decay curve from the counting data. An example of this is given for data recorded on the 6" NSTec system for the second shot, shown in Figures 6 and 7. For these plots, the background was dominated by the decay of the long-lived isotope, such that for Figure 6, the background is approximately 16 counts per minute, and for Figure 7, the background is approximately 750 counts per minute.

Figure 6: Plot of measured exponential decay and number of counts for 1 cm diameter, 1 cm thick sample, for second D-T DPF shot using 6" NSTec coincidence system from shot time through removal from coincidence unit

Figure 7: Plot of measured exponential decay and number of counts for 3 inch diameter, 3/8 inch (.9575 cm) sample for second D-T DPF shot using 6" NSTec coincidence system from shot time through removal from coincidence unit In these plots, the number of counts recorded on the MCS is plotted on a semi-log scale with the exponential fit of the data demonstrating a strong linear relationship in both cases. As this is the case, the linearity of the plots satisfies the radioactive decay law, thereby validating the functionality of the detectors by the demonstration of the expected result.

Results:

 As the objective of this experiment was to cross-calibrate the copper activation systems relative to each other, the different sample sizes, and the lead probe, silver detector and praseodymium sample, the metric that is desired for observing this relationship is the total neutron yield. To calculate the neutron yield, I used the *F* factor method, calculating the factor for the different sample size and detector combinations as described earlier. I also used a measured *F* factor for the 1 cm samples that was determined by a previous experiment by G. W. Cooper, C. L. Ruiz, *et. al.*^[19]. This was done to compare the yield using that factor to the F

factor calculated for these samples. The yield calculations given in Table 2 are those using the *F* factor calculated for the specific samples and counting geometry. In doing this, the measured yields for all the individual shots and detectors are given in Table 2, with the measured yield for detectors with two different-sized samples counted being given as the average of the measured yield from each sample. The large gaps in the table are due to shots where samples were not counted in the detectors, there were errors in the MCS software and data was lost, or where the previous sample was being counted for an extended period of time to allow for an observation of the Cu-64 present.

Table 2: Calculated yields for all shots using dense plasma focus supplied by NSTec for data taken with copper samples counted in the NIF 6" NaI coincidence system, the NSTec 6" NaI coincidence system, or the 3" NaI coincidence system, along with yields calculated from lead probe, silver detector, and praseodymium sample. All values are given as (neutron yield) \times 10⁻¹². For shots where multiple copper samples were counted in the detectors, the yield is given as the **average of the yield from each sample. Data for specific sample sizes is given in Appendix A.**

To determine the quoted errors for the data samples, the errors from equipment malfunctions,

calculation of the *F* factor and counting statistics were propagated through to the final solution to determine the overall error in the value. For the copper yields, the bulk of the error comes from the calibration of the F factor method^[19], with the exception of shots 1-3. Sometime when these shots occurred, the tape slipped, so there was a 5 % uncertainty in the distance from the DPF source to the samples.

Analysis:

 From the plots of the radioactive decay of Cu-62 in the detectors, such as those in Figures 6 and 7, we can determine that the decay product was indeed Cu-62 and not dominated by another isotope or a false reading. This is the case because the 9.74 minute half-life of Cu-62 is significantly shorter than that of Cu-64 and approximately twice that of Cu-66. Also, because Cu-66 is not a positron emitter, the probability of it registering in the coincidence system is extremely low. This is due to the calibration of the time windows for the coincidence system to be on the order of 100 ns and the energy of the Cu-66 gamma ray being outside the SCA window for the 511 keV photon. The potential of these decay values being a false reading from the Ge-68 calibration source is small. If a portion of the Ge-68 source ended up being left in the system, either by fragmentation of the source or operator error, the half-life is 271 days, which would result in a significantly different decay curve than that of the Cu-62. Also, with this long halflife, the activity of the Ge-68 would change negligibly over the counting interval for the copper sample. Therefore, it would be appear to the detectors as a large source of background radiation, and would not noticeably affect the decay rate. As a result, we can look at the measured decay rate of the Cu-62 from the exponential fit for the data in Figures 6 and 7. For the two copper samples inserted in the detector, the measured decay constants were .0708 min⁻¹ and .0703 min⁻¹, which correspond to half-lives of 9.79 minutes and 9.86 minutes, respectively. Comparing the half-lives of these samples to the accepted value for Cu-62, they are both within 1.5 % of the accepted value, thereby falling well within the margin of error for this experiment. Also, the R^2 values of .9892 and .9999 imply that the exponential fit is extremely accurate for modeling the data, so we can be certain that we were detecting the decay of Cu-62.

 The praseodymium sample consistently gave a higher reading than the other materials used, with it being the largest calculated yield 75 % of the time. From this, it can be inferred that the praseodymium likely gives an upper bound on the measured yield for the shot, and that there is some effect in the counting system that results in the praseodymium producing more counts than would be expected. To count the praseodymium, the sum-peak method was used instead of the coincidence method as used for copper. This method allows in all the counting efficiencies being cancelled out by other factors, however, the use of thin samples for the praseodymium could result in positrons escaping the sample and annihilating in the NaI detector instead. Even if the measurement is high due to positrons escaping, the readings for the Pr-141 sample are still generally within their 18% error as calculated by Meehan *et. al.*[21] As the lead probe is a laboratory standard detector, and has been extensively calibrated, with the exception of the two shots where there was an error and no data was recorded, it can be assumed that the lead value would be the most consistent due to its fixed location and operating voltage. However, the low threshold can allow for neutrons to scatter into the detector, and the calibration for a lead probe is done at a distance of $12 - 18$ ", such that the validity of the calibration at large distances is uncertain. Another aspect with the lead is that its extremely short half-life for the lead-207 metastable state, coupled with its high detection efficiency and cross section, requires the detector to be at a larger distance to prevent saturation in the plastic scintillator. For the copper samples, the measured yields are generally closest to those of the silver samples, but usually within the error margin of at least one of the other materials, be it the praseodymium or lead.

 As one of the objectives for the experiment was to cross-calibrate the system relative to different size samples, we looked at the measured yield for the samples individually. In doing this, we were able to distinguish between the different sized samples and determine the relative

yields dependent on the sample sizes. It was especially beneficial to look at shots on the two 6" NaI systems where there were two different sized samples irradiated and counted in the same detector. This is the most beneficial data set because it allows for a determination of which sample size is generally the most accurate relative to the lead, praseodymium, and silver. This in turn helps determine which size samples will give the best results for experiments done at NIF. The data for the different samples in the 6" NaI detectors on the different shots can be seen in Figure 9.

Figure 9: Measured total neutron yields for copper samples in 6" NaI coincidence systems discriminated by system and sample size and average measured yield from lead, praseodymium and silver activation along with overall average from all detectors

From Figure 9, it is evident that the 3 inch samples generally give the closest results for the specific systems, as the only time where the 3" sample was further from the average value than

the smaller copper sample was for shot 8 in the NSTec system. This is reinforced further by the average ratio of measured yields between the copper samples and the lead, praseodymium and silver average, which can be seen in Table 3. From the data, it is even clearer that the 3 inch sample is the most accurate, as the average measured yields is 20% closer to the average yield for the lead, praseodymium and silver than the other samples. However, if we ignore the measured yield for the 3" sample counted in the NSTec system on shot 8, the ratio drops significantly, but ends up being 7.56×10^{-1} , which is still significantly closer than either of the other sample sizes.


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Table 3: Average ratios of measured yield for specific sizes of copper samples in 6" NaI detectors to the average yield of 
the lead, praseodymium, and silver measurements
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For the 1 cm \times 1 cm samples, the calculated *F* factor is very close to the measured *F* factor from Cooper, *et. al*. As this is the case, the measured yield would be expected to also be close due to all other factors in the yield calculations being the same. From the ratio of the yields, it was determined that the yield measurement using the calculated *F* factor was approximately 2% less than the yield measurement using the previously determined *F* factor, such that $\frac{Y_{Calc}}{Y_{Meas}}$ = .98. Because these yield measurements are both well within the error

margin of the calculations (~10 %), there is no reason to believe that either the measured *F* factor from the earlier experiment or the calculated *F* factors are incorrect.

It is also evident that, in general, the measured yields for the copper samples are significantly lower than the measured yields calculated for the lead, praseodymium, and silver activation. This difference is evident for nearly all the samples counted in the 6" systems, and all of the shots counted in the 3" system. A possible explanation for this is the DPF wall, which is .375" thick and made of stainless steel. This wall was calculated into the *F* factor as a .25" thick

wall, such that the additional .125" would increase the amount of attenuation. As the *F* factors for these data sets were calculated using the fundamental quantities for neutron reactions in the material and the detector counting efficiencies, the presence of a thin wall that was not modeled into the system would provide attenuation that would result in a low measurement of the total neutron yield. However, since the unaccounted for portion of the wall is thin, the total attenuation due to scattering in the wall would be small. This is because neutrons are one of the most penetrating forms of ionizing radiation, and will generally pass through the material without interacting enough for the scattering angle to be appreciable. In calculating the number of neutrons that would scatter out of the sample, it was found that for a stainless steel DPF wall of thickness .375" , the flux would decrease to 94.6 % of the value for a .25" thick wall. As such, the measured yield would increase by 5.7 %, which results in a value closer to, but still less than the other materials. However, this is possibly a reason for the lower yield measurement using the *F* factor method, as it is unknown whether the yield measurements using the praseodymium, lead, and silver correctly modeled the neutron attenuation due to the wall for their calculations. Also, because the trajectory of neutrons in air at STP is approximately linear, there would be approximately no attenuation over the short distances between the DPF source and the samples, and the decrease in counts due to scattering would be entirely due to the DPF wall.

 The second oddity in the calculated yields is the significant decrease in the yields for those calculated using the *F* factor and the 3" NaI coincidence system. The calculated yield for this system is not within the margin of error for any of the reference values (Pb, Pr, or Ag), and only once within a margin of error for the yield measurements using the 6" NaI coincidence systems. As this is the case, the most likely causes are the modeling and calculation of the

counting efficiency for the 3" detector, or some fault in the electronics. Because the calculations for counting efficiencies were done using the assumption of 19.1 % efficiency for counting a germanium-68 calibration source, the most likely reason that the values were consistently low is that the counting efficiency of the 3" NaI system with a Ge-68 source is less than 19.1 %. If we make this assumption, and further assume that the counting efficiencies modeled are half of what they actually are, then all of the yield calculations would double and be within a margin of error of at least one of the praseodymium, lead, and silver samples for all but the first shot. If we assume that the actual counting efficiency is only 1.5 times the modeled value, then, with the exception of shots 4 and 5, the calculated yield would fall in the middle of the Pr, Pb, and Ag measurements. A final cause of this decrease could be the geometry detector system itself due to the geometry dependence of the *F* factor calculation. Slight changes in the geometry due to a dent in the casing or a detector becoming loose and moving slightly in the system could affect the counting efficiency, but any such difference could change the measured yield by at most 1 percent, with a more reasonable estimate being <.1 %.

 Looking at the exponential decay curves for the data series counted on the 3 inch NaI coincidence system provided an interesting insight into the counting electronics used for the experiment. For the samples counted in the 3" detector for both the first and third day of the experiment, I noticed that the decay constant for these samples was noticeably less than the accepted value, as is demonstrated in Figure 10. However, for the samples counted on the second day of the experiment, the decay constants for the samples on the 3 inch system are within 1 percent of the accepted value for copper-62.

Figure 10: Exponential decay curve for 3 inch copper sample counted on 3" NaI coincidence system in Shot #4, demonstrating decay constant 4.2% less than accepted value

The fact that this occurred only for the 3 inch NaI system is interesting because the measurement of the yield for these values was also consistently low. However, to calculate the yield, the assumption was made that the half-life of copper-62 is known, and the yield was calculated using an activity determined with that value for the decay constant. In doing this, the initial activity of the sample that I used for the measurement of the yield is higher than the activity as determined by propagating the exponential decay curve backwards in time. This means that the yield that I calculated is greater than if calculated using the activity from the exponential fit of the decay. Having discovered this discrepancy in the half-life, it was hypothesized that the cause of the error is a malfunction in either the MCS board or the computer clock. The presence of an intermittent problem in either the MCS or computer clock could result in the measured half-life having large errors on some shots due to incorrect timing of the counting intervals by the computer. These errors would change the measured half-life by associating a certain number of counts with an incorrect interval. The cause of this problem is unknown, but may have been

related to the heat, as at least one of the computers overheated and shut down during the experiment. As these timing issues were first noticed recently, it would be beneficial to find the specific board and computer that were used with the 3" system to determine if there are problems with the internal clocks.

Sources of Error:

 The significant sources of error for this experiment are systematic in nature dependent on the geometry for the irradiation of the samples. As the samples were being irradiated using a dense plasma focus which had a maximum output of $>10^{12}$ neutrons in a single pulse, it was necessary to have shielding in place and operate the DPF remotely to minimize the radiation exposure. This was important because the samples had to be lowered into the target chamber on a train car attached to a tape measure to determine their distance away from the source. This contributed to the sources of error because for shots 1-3, the tape slipped at some point, but was not noticed until the fourth shot, so the distance from the sample to the DPF source was estimated to be $(d_0 - 2.54) \pm 2.54$ cm. Although this amounts to an error of approximately 5 % in the distance from the source, as the shots were done at a distance of approximately 51 cm instead of 53.3 cm, the yield is dependent on the square of the distance. As such, the error in the calculated yield for the copper samples on these shots could be up to 10.25 %.

The second error that comes from the use of the dense plasma focus is that the geometry for the irradiation of the samples cannot be entirely determined as the angle of the samples relative to the DPF field pinch is only approximate. This affects the results because the neutron energies are not uniform over 4π , and has a desired angle of 95° relative to the orientation of the plasma current in the pinch for 14.1 MeV neutrons^[19]. This angular dependence is due to the

conservation of momentum for ions in the plasma. At angles greater than 95°, the neutron energy is less than 14.1 MeV, whereas for angles less than 95°, the energy is greater than 14.1 MeV. Because the angle is partially unknown, but assumed to be close, the irradiation of the samples would differ slightly from the desired arrangement, which would result in the neutrons irradiating the samples having energy different than 14.1 MeV. This could have an effect on the measured yields by changing the cross sections for neutron interactions with the materials. However, none of the materials used have a spike or drop in the reaction cross section around 14 MeV, such that the difference in cross section should not be large enough to have a significant impact.

 The second source of error was the thickness of the DPF wall, as its actual thickness was .125 inches thicker than its modeled thickness. Because the wall adds attenuation to the neutron beam, the measured yield was ~6 % less than if the attenuation was accounted for in the *F* factor. The greatest effect of this additional thickness is in the comparison between the copper samples and the other materials. If the attenuation was accounted for correctly by the researchers working with the other detectors, then the results from the copper are artificially low. As such, adjusting the yield corresponding to this additional attenuation will give a measured yield that is not only more accurate, but also more agreeable with the other data.

 The third source of error is the design of the detectors and the use of the *F* factor for calculating the yield with different detectors. As the counting efficiencies were modeled for the 6" NaI coincidence system that is used for copper activation samples at the National Ignition Facility, and the data I worked with were for both 6" NaI systems, a possible difference in the coincidence system construction would have an effect on the yield. As both systems were constructed by the neutron activation group at Sandia Labs, the only difference between them

should be the NaI detectors as the NIF system uses newer detectors than the NSTec system. The effect of the different system on the yield calculation is unknown, as the exact interior dimensions and sizes of the NaI crystals could not be measured. The only known difference between the two is that the casings on the detectors in the NSTec system were iron, whereas the casings in the NIF system were aluminum due to a change in construction by the manufacturer. However, any differences should be small as both 6 inch coincidence systems were designed to be identical and the 3 inch system was designed to be similar to the Shiva copper activation system used by Lawrence Livermore National Laboratory for copper samples on $NOVA^{[20]}$.

Conclusions:

 At National Security Technologies, we used a dense plasma focus containing a mixture of deuterium and tritium fuels to irradiate copper samples for a cross-calibration of 6" thalliumdoped sodium-iodide and 3" NaI(Tl) detectors relative to a lead probe, silver detector and praseodymium samples. Using the (n,2n) reaction in copper to measure the 14.1 MeV neutrons produced by D-T fusion, we calibrated the *F* factor method for determining the neutron yield with these coincidence systems through the theoretical calculation of the *F* factor based off of fundamental quantities for the copper and the detector efficiency for the specific geometry. Having done so, we found that the measurement of the total neutron yield with the *F* factor method for copper activation is low compared to the measurement of the yield with lead, silver and praseodymium.

 By comparing the neutron yield for the 6" coincidence system to that of the 3" system and the lead, praseodymium, and silver, we found that the measured yield for the 6" system is significantly closer to the other materials than the 3" system. As such we can conclude that as a

diagnostic for a large-scale experiment, such as the Laser Inertial Fusion Engine and National Ignition Campaign experiments at Lawrence Livermore National Laboratory, the 6" coincidence system will be the most effective for measuring the total neutron yield of the individual shots. Furthermore, by knowing the exact geometry and capsule irradiation for ICF experiments, the theoretical method of calculating the *F* factor will give an accurate measurement of the total neutron yield.

 Finally, by comparing the ratios of the measured yields for different sizes of copper samples in the 6" NaI coincidence systems to the average yield from the lead, praseodymium, and silver, we found that the larger sample sizes resulted in ratios closer to one. For these samples, the most pronounced difference was going from the 1 inch sample to the 3 inch sample. The average measured yield for 3 inch samples was approximately 20 % closer to the average yield with the other materials than the average yield for the 1 inch. Also, we found that the yield calculated with the *F* factor method gives approximately the same answer for 1 cm diameter values as that using the measured *F* factor from G. W. Cooper, C. L. Ruiz *et. al.*^[19]. As such, we concluded that for the copper activation detectors on NIF, of the samples tested at NSTec, it would be most beneficial to use the 3 inch diameter by $\frac{3}{8}$ inch thick copper samples for measuring the total neutron yield. Furthermore, we concluded that for the 3 inch diameter samples, the measured total neutron yield would be 89 ± 10 % the average measured yield from praseodymium, lead, and silver samples.

Proposed Further Research:

 To improve the experimental results, it would be beneficial to irradiate the samples in a more controlled geometry. This irradiation can be done using either a DPF for producing the D- T neutrons, or with an ion beam, but should have the samples located at the desired angle relative to the source and at a more controlled length, rather than a variable length and uncertain angle. Doing this will allow for the measurement of a yield with more certainty due to the specific modeling of the system for this geometry, thereby giving a more accurate value for the theoretical *F* factor. Specifically, the ideal experiment would be to use an ion beam, such as the IBL at Sandia National Labs with a fixed target chamber. In doing this, the use of a velocity selector will result in a deuteron beam of nearly equal energy and associated particle counting in the beam will give a known beam current. Using these quantities, we can calculate an approximately absolute measurement of the number of neutrons that would be produced, allowing for a direct measurement of the *F* factor for any desired sample size, rather than having to calculate the factor for specific geometries and sample sizes. Furthermore, the use of an ion beam would allow for both pulsed neutrons and a steady-state 14.1 MeV neutron flux, which would allow for an exact calculation of the detector efficiency for copper samples.

 The second experiment that would be beneficial is to measure the detector efficiency with sources of known activity. To do this, the two methods that would be most efficient would be to insert a germanium-68 source into the coincidence system or to saturate a copper sample to insert into the system. For the germanium source, the objective would be to count the 511 keV photons that occur from the decay of gallium-68 and determine the number that were detected in the system. For a copper sample, the use of a nuclear reactor or steady-state fission source (such as californium-252) will be able to saturate the sample to a known activity to count the 511 keV photons from the decay of Cu-62 and Cu-64. These methods are effective for determining the total detector efficiency because they allow for an absolute calculation of the activity of the sample. Then by simply taking the ratio of the measured activity against the decay of the
saturation activity, the coincidence system efficiency can be determined for measuring the beta decay in the samples.

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are for a primarily qualitative observation of the yield relative to the samples. Only the 6" systems are shown because that size system
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Appendix B – Raw Data:

Shot 1 – NIF System:

Shot $1 - 3$ " NaI System:

Shot 2 – NIF System:

Shot 2 – NSTec System:

Shot 3 – NIF System:

Shot 3 – NSTec System:

Shot $3 - 3$ " NaI System:

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Shot 4 – NIF System:

Shot 4 – NSTec System:

Shot $4 - 3$ " NaI System:

Shot 5 – NIF System:

Samples Counted Sample Mass (g) Distance to DPF (cm)

Shot 5 – NSTec System:

Shot $5 - 3$ " NaI System:

Shot 6 – NIF System:

Shot $6 - 3$ " NaI System:

Shot 8 – NIF System:

Shot 8 – NSTec System:

Shot 9 – NIF System:

Shot 10 – NIF System:

Shot 10 – NSTec System:

The Rabbit Tube consists of six 1 inch diameter samples stacked back to back in a sheath with $\frac{1}{4}$ " sides for the casing

Shot 11 – NIF System:

Shot $11 - 3$ " NaI System: