Carbon Activation Diagnostic for Tertiary Neutron Measurements

Introduction

In inertial confinement fusion $(ICF)^1$ implosions, nuclear reactions in the fusion fuel produce energetic neutrons and a variety of charged particles. The primary reaction for DT fuel is

$$D + T \rightarrow \alpha + n (14.1 \text{ MeV}). \tag{1}$$

In a secondary reaction, a small percentage of 14.1-MeV neutrons will scatter elastically from D or T ions in the fuel (prime notation indicates a scattered particle):

$$n + D \rightarrow n' + D' (0 - 12.5 \text{ MeV}),$$
 (2)

$$n + T \rightarrow n' + T' (0 - 10.6 \text{ MeV}).$$
 (3)

As these scattered ions pass through the fuel, some will undergo tertiary, in-flight fusion reactions:

D' (0-12.5 MeV) + T
$$\rightarrow \alpha$$
 + n'' (12.0-30.1 MeV), (4)

$$T'(0-10.6 \text{ MeV}) + D \rightarrow \alpha + n''(9.2-28.2 \text{ MeV}).$$
 (5)

The yield of these high-energy tertiary neutrons is proportional to $(\rho R)^2$ for small values of ρR , where ρR is the areal density of the DT fuel, and to ρR for large values of ρR (Ref. 2) and is about 10⁻⁶ of the primary 14.1-MeV neutron yield.

The fuel areal density ρR is a fundamental parameter that characterizes the performance of an ICF implosion. For high areal densities ($\rho R > 0.3$ g/cm²), which will be realized in implosion experiments at the National Ignition Facility (NIF)

and Laser Megajoule Facility (LMJ), the target areal density exceeds the stopping range of charged particles, and ρR measurements with charged-particle spectroscopy will be difficult. In this region, an areal-density measurement using tertiary neutrons is one of the alternative methods. The use of tertiary neutrons for measurements of high areal densities in ICF implosions has been proposed by several authors^{2–4} in the past. This article describes the experimental development of a tertiary neutron diagnostic using carbon activation and the 30-kJ, 60-beam OMEGA laser system.⁵

Carbon as an Activation Material

The use of carbon as a threshold activation material was proposed many years ago.³ There are three main reasons why carbon is a good activation material for tertiary neutron measurement. First, the ¹²C(n,2n)¹¹C reaction has a *Q* value of 18.7 MeV, well above the 14.1-MeV primary DT neutron energy. Thus, the reaction ¹²C(n,2n)¹¹C will occur only from interactions with the high-energy tertiary neutrons. The experimental cross section of the ¹²C(n,2n)¹¹C reaction shown in Fig. 92.12 was measured in several experiments and can be used to calculate a tertiary neutron signal in a carbon sample.

The second attractive feature of carbon is the properties of its decay. The isotope ¹¹C decays to ¹¹B with a half-life of 20.39 min and emits a positron, resulting in the production of two back-to-back, 511-keV gamma rays upon annihilation. The ¹¹C half-life is advantageous since it is compatible with the experimental conditions on OMEGA. The OMEGA laser fires at approximately one-hour intervals, and it takes a few minutes to remove the disk from the target chamber and carry it to the gamma-detection system after the laser has been fired. Thus, if the half-life were much shorter, a significant amount of information would be lost during transfer. If it were considerably longer, there would not be enough time to record all of the decays between shots. The positron decay of ¹¹C is nearly identical to the ⁶²Cu decay used in the copper activation measurements of 14.1-MeV primary DT yields; therefore, the well-developed copper activation gamma-counting system^{6,7} can be used.



Figure 92.12

Experimentally measured cross section σ for the reaction ${}^{12}C(n,2n){}^{11}C$. The solid line is the fit used for calculations in the **Carbon Activation for OMEGA and the NIF** section.

The third reason carbon was chosen is the availability of high-purity samples. The purity of the activation sample is very important for tertiary activation diagnostic, as will be discussed in the **Contamination Signal in Carbon Activation** section below. Carbon is also a nontoxic, nonflammable, inexpensive, and safe material. These properties give carbon a big advantage over sodium, which has also been proposed.³

OMEGA Carbon Activation System

The activation samples with a 7.6-cm diameter and a different thickness can be inserted with a pneumatic retractor into a reentrant tube installed on the OMEGA target chamber. Before a shot, the activation samples are manually installed in the retractor holder and inserted into the target chamber at 42 cm from the target. Immediately after a laser shot, the activated sample is automatically extracted from the chamber and dropped through a plastic tube to the pickup basket in a room under the OMEGA Target Bay. The operator delivers the activated sample to the gamma-detection system in the counting room. Generally, this process takes 1.5 to 3 min. Since the space in the OMEGA target chamber is very limited, the same pneumatic retractor is used for both copper and carbon activation diagnostics.

The OMEGA gamma-detection system consists of two 7.6-cm-diam, 7.6-cm-thick NaI(TI) scintillation detectors separated by 0.89 cm or 1.78 cm and associated electronics. Each detector has an ORTEC 460 delay linear amplifier and an ORTEC 551 single-channel analyzer (SCA), which perform pulse-height discrimination. The time coincidence between two detectors is established by an ORTEC 418A universal coincidence module followed by an ORTEC 974 quad counter/ timer module that counts time, single counts from each detector and two detectors coincidence. The 974 counter is read by a PC-based data acquisition program every 5 s and recorded for future analysis. The window of each SCA is set between 426 keV and 596 keV in order to detect 511-keV gammas from positron annihilation. The energy scale of each SCA is calibrated with a ²²Na radioactive source before each set of measurements. The NaI detectors are shielded from all sides with 7.6 cm of lead to reduce cosmic ray background. As a result of the shielding, the background coincidence count rate is about 18 counts/hour. To isolate the NaI detectors from direct activation by 14.1-MeV neutrons produced during high-yield OMEGA shots, the gamma-detection system is placed at a distance of 120 m from the target. Dedicated experiments have shown that the gamma-detection system records no coincidence for yields up to 7×10^{13} .

Contamination Signal in Carbon Activation

Any material that produces a positron emitter by interaction with 14.1-MeV neutrons will add a contamination signal in the carbon activation diagnostic. The most-dangerous contaminants for carbon activation are copper and nitrogen, which produce positron emitters in reactions $^{63}Cu(n,2n)^{62}Cu$, $^{65}Cu(n,2n)^{64}Cu$, and $^{14}N(n,2n)^{13}N$. Each of these reactions has a threshold below 14 MeV. The nitrogen cross section for a 14.1-MeV neutron is 6.5 mb and is comparable to the carbon cross section for tertiary neutrons. The ^{63}Cu cross section for 14.1-MeV neutrons is about 100 times larger than the carbon cross section for tertiary neutrons. Since the primary 14.1-MeV neutron yield is about 10^6 times larger than the tertiary yield, the contamination in the carbon sample must be less than one part per million (ppm) for nitrogen and 0.01 ppm for copper.

In the development of the carbon activation diagnostic, graphite disks from Bay Carbon⁸ were used. Bay Carbon carefully selects their graphite for its physical and chemical compositions and performs chemical vapor purification (CVP) of the graphite. Bay Carbon purity is defined as follows: No more than three elements may be present (other than carbon), no single element may exceed one part per million

(ppm), and the total impurity level may not exceed two ppm in the graphite.

In addition to the purity of the sample material itself, proper packaging and handling procedures are very important. To keep the graphite disks clean before a shot, the disks are shrinkwrapped in plastic before being placed into the retractor and delivered to the gamma-detection system. The plastic is removed and discarded before counting. This procedure prevents surface contamination of the graphite disks. This is especially important on OMEGA because the same retractor is used for both copper and carbon activation. The graphite disks without shrink wrap showed a contamination signal of about 1000 coincidences per hour of counting. The shrink-wrapping procedure protects the graphite disks from surface contamination but leads to other forms of contamination.

The shrink-wrap plastic contains hydrogen. When bombarded by 14.1-MeV neutrons, the hydrogen atoms can produce elastically scattered protons with energies up to 14 MeV. These protons interact with carbon in the graphite disks and produce positron-emitting nitrogen via reaction ${}^{12}C(p,\gamma){}^{13}N$. This source of contamination was eliminated through the use of graphite foils (thinner pieces of graphite) placed on both sides of the disk between the disk and the shrink wrap. The graphite foils act as protective barriers for the protons from the plastic. This combination of the disk and two foils is called a "sandwich." The foils are made from the same graphite as the disks in order to keep the disks clean. The foils are 2.5 mm thick, enough to completely stop the protons originating in the plastic and prevent their penetrating the graphite disk. All contamination from such protons is restricted to the graphite foils, which are removed and discarded before counting.

Because the graphite porosity is about 20%, the graphite disks can absorb and store ¹⁴N nitrogen from the air. The 14.1-MeV neutrons can produce positron-emitting nitrogen ¹³N via the reaction ¹⁴N(n,2n)¹³N. To remove air from the graphite disks, a special purification facility was developed at the SUNY Geneseo.

The first step in the purification process is to remove the nitrogen and other contaminant gases from within the graphite disks and replace them with an inert gas such as argon that cannot be activated by 14.1-MeV neutrons. To do this, a large three-zone tube furnace is employed. This furnace is composed of a 7-ft-long, 7-in.-diam quartz tube. The graphite disks are inserted into the oven using a 5-ft-long rod. The oven is then brought to vacuum, heated to 1000°C, and maintained at that

temperature for several hours. After cooling to room temperature, the quartz tube is flooded with high-purity (0.99995) argon. The disks stay in this environment for several hours. They are extracted from the quartz tube by placing a large glove bag over one end of the tube, filling it to positive pressure with argon gas, and then using a rod to remove the disks from the quartz tube into the glove bag. Graphite foils are fit to both sides of the disk, and the disks are then placed into small vacuum bags, which are immediately vacuum sealed. The disks can remain in the vacuum-sealed bag for at least six months without contamination.

In earlier experiments, the disks were taken out of the vacuum bags immediately before the shot and quickly sealed in shrink wrap, which was left on during the shot. In later experiments, the disks were shot in the vacuum bags, never being exposed to air contaminants.

Experimental Results

The main goal of the diagnostic development was to minimize the contamination signal in carbon samples. Several iterations of the purification, packaging, and gamma-detection system were made until the system described above was developed. In this section, results obtained in 2001–2002, after major improvements to the system, are presented.

Tests of the carbon activation system were carried out on the 30-kJ, 60-beam OMEGA laser system⁵ in direct-drive implosions. A 1-ns square laser pulse shape with 28-kJ to 31-kJ energy was used to implode glass microballoons with shell thicknesses from 2.5 to 4 μ m filled with 20 atm of DT. These targets have a very low ρR and should produce no measurable tertiary neutrons. Thus, any signal in these experiments was a contamination signal. The primary DT neutron yield was measured by an absolutely calibrated time-of-flight scintillating counter located 20 m from the target. The neutron yields in these experiments ranged from 4×10^{13} to 9.6×10^{13} .

In the experiments on OMEGA, the cosmic ray background for the empty gamma-detection system was measured, fitted to a linear function, and subtracted from the coincident counts for each carbon disk. The results from the graphite disks were then normalized to a yield of 7.4×10^{13} , which was the average neutron yield for this series of measurements. The carbon disks without shrink wrap produced a contamination signal of about 1000 counts; disks in shrink wrap produce about 80 to 100 counts; and disks packaged as a sandwich in shrink wrap produce a contamination signal of only about 30 to 50 counts. The results for the carbon disks irradiated in sealed vacuum bags are shown in Fig. 92.13. The two shots with carbon disks in vacuum bags without foils show a higher contamination signal than from a sandwich, demonstrating the importance of shielding from the protons originating in the plastic. The six sandwich results include four shots from June 2001 and two "consistency check" shots from June 2002. All of them show a similar contamination signal of 30 to 50 coincidence counts for 7.4×10^{13} primary neutron yield.



Figure 92.13

Coincidence counts in the gamma-detection system as a function of time after a shot for 0.89-cm-thick disks and "sandwiches."

The 0.89-cm carbon disk thickness originates from a copper disk thickness optimized for 511-keV gamma absorption in copper. Since the absorption length of 511-keV gammas in carbon is about five times larger than in copper, the carbon disk thickness can be increased. In June 2002 we tested on OMEGA carbon disks with a thickness of 1.78 cm. The results of these tests normalized to the same 7.4×10^{13} primary neutron yield are shown in Fig. 92.14. The contamination signals from the thicker disks are similar to the thinner disks, suggesting that contamination is mostly a surface-related effect. In the thicker disks the tertiary signal will increase by a factor of 2, and the efficiency of the gamma-detection system decreases for the thicker disk by approximately 20%; the thicker disk has a 1.8 gain in sensitivity. Monte Carlo calculations for the optimal carbon disk thickness are in progress.

Consistent, repeatable results in two sets of measurements one year apart, which include the manufacture of new carbon disks, show stability and reproducibility of the carbon activation diagnostic chain—from manufacturing the disk to the purification and handling system.





Coincidence counts in the gamma-detection system as a function of time after a shot for 1.78-cm-thick sandwiches.

Carbon Activation for OMEGA and the NIF

Direct-drive spherical DD cryogenic target implosions⁹ are routinely carried out on the 60-beam OMEGA laser system, and implosions with cryogenic DT fuel are planned for the near future. The OMEGA cryogenic DT design and expected laser pulse shape with an intensity picket are shown in Fig. 92.15. The intensity picket shapes the adiabat of the main fuel and ablator, reducing both the seeds and the growth rate of Rayleigh-Taylor instability.¹⁰ A one-dimensional LILAC¹¹ simulation calculates a neutron yield of $Y_n = 6.0 \times 10^{14}$ and a neutron-averaged fuel areal density $\langle \rho R \rangle = 245 \text{ mg/cm}^2$ for the design shown in Fig. 92.15. The IRIS¹² postprocessor to LILAC was used to calculate the spectrum of all neutrons emerging from an OMEGA cryogenic DT target. Figure 92.16 shows the calculated tertiary/primary neutron spectrum and the same spectrum multiplied by the cross section of the ${}^{12}C(n,2n){}^{11}C$ reaction approximated by the solid line in Fig. 92.12.

The target design from Fig. 92.15 and tertiary spectrum from Fig. 92.16 were used to estimate the expected signal from a carbon activation sample in OMEGA cryogenic DT experiments. A 7.6-cm-diam, 0.89-cm-thick carbon disk at 40 cm from the target subtends solid angle 2.25×10^{-3} . The integral of the tertiary/primary neutron spectrum multiplied by the carbon cross section in Fig. 92.16 is about 3×10^{-5} mb/ primary neutron. The 80-g carbon disk will receive 1.2×10^{-7} activations for each incident primary neutron or 2.7×10^{-10} activations for each primary neutron produced in the

target. The gamma-detection system has an efficiency of 20%, thus the measured coincidence counts per produced primary neutron will be 5.4×10^{-11} . At a primary DT yield $Y_n = 6.0 \times 10^{14}$, we expect 3.2×10^4 coincidence counts from tertiary neutrons in the gamma-detection system. A similar calculation



Figure 92.15

The OMEGA cryogenic capsule design (a) and pulse shape with an intensity picket (b).



Figure 92.16

Ratio of tertiary N_t to the primary N_p neutrons per energy interval (MeV) as a function of tertiary neutron energy and the same ratio multiplied by the carbon cross section σ in mb.

for the implosion quenched at a $\langle \rho R \rangle = 150 \text{ mg/cm}^2$ and neutron yield $Y_n = 9.2 \times 10^{13}$ gives about 400 coincidence counts in the gamma-detection system. With the achieved level of contamination signal, the present carbon activation system can be used for areal-density measurements of OMEGA cryogenic DT targets.

Tertiary neutron measurements by carbon activation are an inexpensive, well-developed diagnostic that can be easily deployed on the NIF for areal-density measurements. All major diagnostic development problems have already been solved on OMEGA. Purified carbon packages are good for at least six months; consequently, they can be prepared in the already-existing purification facility at SUNY Geneseo and shipped to the NIF as needed. The carbon activation diagnostic will require a rapid transport system similar or identical to the yield activation system on the NIF because of the relatively short half-life of carbon.

The only problem that must be solved on the NIF for carbon activation is background in the gamma-detection system itself generated by the neutrons from a NIF implosion. Such a background-induced problem was observed on OMEGA when the gamma-detection system was located 40 m from the target. In Fig. 92.17 the background coincidence counts of an empty gamma-detection system are shown as a function of time. After a shot with a neutron yield of 7.35×10^{13} , a jump of 42 coincidence counts was recorded. Additionally the background stays high for about 1 h after the shot and then returns to the normal cosmic ray level. On OMEGA this background prob-



Figure 92.17

Background coincidence counts of an empty gamma-detection system at 40 m from the target during an OMEGA shot with a neutron yield of 7.35×10^{13} .

lem was solved by moving the gamma-detection system much farther from the target. Since neutron yield on the NIF will be about 10^5 times higher than on OMEGA, a judicious choice of neutron shielding and location of the gamma-detection system will be required. Background measurements during the shot will also be needed. Two identical gamma-detection systems could be implemented—one for signal and another for the background measurement.

Conclusions

This article has described the experimental development of the carbon activation diagnostic for tertiary neutron measurements performed on the 60-beam OMEGA laser system. We have created a purification facility and have developed packaging and handling procedures that significantly reduce the contamination signal in the carbon samples. Experiments on OMEGA in 2001–2002 have shown very good reproducibility of the contamination signal from the carbon samples.

The present carbon activation system is ready for arealdensity measurements of DT cryogenic targets on OMEGA. The same carbon activation diagnostic can be implemented on the NIF, although the neutron-induced background issues in the gamma-detection system need to be solved for the NIF.

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