

Irradiation Environment of the Materials Test Station

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Abstract. Conceptual design of the proposed Materials Test Station (MTS) at the Los Alamos Neutron Science Center (LANSCE) is now complete. The principal mission is the irradiation testing of advanced fuels and materials for fast-spectrum nuclear reactor applications. The neutron spectrum in the fuel irradiation region of MTS is sufficiently close to that of fast reactor that MTS can match the fast reactor fuel centerline temperature and temperature profile across a fuel pellet. This is an important characteristic since temperature and temperature gradients drive many phenomena related to fuel performance, such as phase stability, stoichiometry, and fission product transport. The MTS irradiation environment is also suitable in many respects for fusion materials testing. In particular, the rate of helium production relative to atomic displacements at the peak flux position in MTS matches well that of fusion reactor first wall. Nuclear transmutation of the elemental composition of the fusion alloy EUROFER97 in MTS is similar to that expected in the first wall of a fusion reactor.

1. Introduction

The Materials Test Station (MTS) is an irradiation facility under design at Los Alamos National Laboratory with the mission of providing a test bed for fuels and materials under development for the next generation of fast-spectrum fission reactors. As an irradiation facility for nuclear fuels and materials it is unique in that the source of neutrons is nuclear spallation rather than nuclear fission. The neutron source is driven by a 1-MW proton beam delivered by the Los Alamos Neutron Science Center (LANSCE) linear accelerator. Conceptual design of the facility is now complete. Preliminary and final design phases are estimated to take two years, followed by two years to construct the facility. The total project cost range to complete design, construction, and commissioning is \$75M to \$95M. The US Department of Energy's Office of Nuclear energy is the principal sponsor. The facility concept and essential features are described elsewhere [1,2].

Because of the unique features of its accelerator-driven liquid-metal-cooled spallation neutron source, MTS can safely provide an irradiation environment that is in many respects prototypic of proposed fast reactors, as well as provide an irradiation capability for other DOE missions such as fusion materials testing. Design, construction, and operation will conform to accelerator safety requirements.

The spallation source generates neutrons that in turn irradiate test fuel and materials specimens to assess their performance under prototypic fast reactor conditions of linear heat generation rate, operating temperature, and neutron spectrum. The MTS design can accommodate state-of-the-art in-situ diagnostics to measure evolution in test specimens during irradiation. The MTS could be designed to allow probe beams of photons, protons, or neutrons to measure micro- and macro-structural features

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in a manner that would be difficult to employ in a reactor. This opens up unprecedented opportunities to conduct separate effects testing in MTS, in addition to integral tests on fuel rodlets (i.e., short pins each containing a fuel stack of ~10 cm or less).

The design life of the MTS is twelve years. The MTS is scheduled to operate 4,500 hours per year at a beam current of 1.25 mA (historical data indicate that delivered beam time is about 85% of scheduled beam time). The beam spot on each of the two spallation targets is nominally 15 mm wide by 60 mm tall. A nearly uniform current density over the beam spot is achieved by rastering a small beam over the beam spot dimensions, which produces a nearly uniform proton beam current density on target of $70 \mu\text{A}/\text{cm}^2$. Thus, the annual proton dose on the target front face is 6.0×10^{21} protons/cm². The anticipated minimum life of the target is expected to be six months, but the expectation is that, with operating experience, it will not require more than annual replacement.

2. Irradiation Environment for Fast Reactor Fuels Testing

The critical parameters important for fast reactor fuels testing are:

- fast neutron flux exceeding 1×10^{15} n/cm²/s (“fast” includes all neutrons with energy greater than 0.1 MeV);
- peak annual fast neutron fluence of at least 1.5×10^{22} n/cm² (along with the fast neutron flux, this sets a minimum requirement on facility availability, or hours operated per year);
- irradiation volume sufficient to irradiate at least thirty rodlets (a rodlet consists of a 10-cm-high fuel column plus gas plenum, and cladding);
- uniform fission rate (within $\pm 5\%$) throughout a fuel pellet with nominal dimensions of 5 mm diameter by 5 mm high;
- ability to irradiate at a fuel cladding surface temperature within the range 350°C to 550°C, controllable to within 10°C.

The MTS design meets all of these parameters.

2.1. Fast Neutron Flux, Irradiation Volume, and Annual Fast Fluence

The spatial distribution of the fast neutron flux, at the horizontal mid-plane of the MTS target, is shown in figure 1. The fuel irradiation region is located between the two spallation target sections, and is designed to accommodate up to 40 rodlets. The peak fast neutron flux in the fuel irradiation region is 1.3×10^{15} n/cm²/s, with 18 rodlets whose mid-plane fast flux exceeds 1.0×10^{15} n/cm²/s. At 5 cm above and below target mid-plane the fast flux has dropped to half of the mid-plane value. With a scheduled beam delivery of 4,500 hours per year and an expected availability of 85% of scheduled hours, the annual peak fast fluence is 1.8×10^{22} n/cm².

2.2. Fission Rate Uniformity

The uniformity of the fission rate over the dimensions of a fuel pellet (nominally 5 mm diameter by 5 mm high) influences the radial and axial temperature distribution within the pellet, and the axial and radial dependence of the production and subsequent transport higher actinides and fission products. It is therefore important to replicate as closely as possible the near-uniform fission rate exhibited by fast reactors. Radially, the fission rate within a fuel pellet depends on the neutron transport mean free path. If the mean free path is greater than the pellet diameter, the fission rate shows no radial dependence. This is the case for both fast reactors and MTS, where the mean free path exceeds 1 cm. Axially, the fission rate is proportional to the fast neutron flux. In MTS, the axial gradient is zero at target mid-plane where the flux is greatest. The gradient is steepest 3 to 4 cm above and below target mid-plane. Here the gradient is 20%/cm. Over the 5-mm height of a nominal fuel pellet, the axial uniformity $\pm 5\%$.

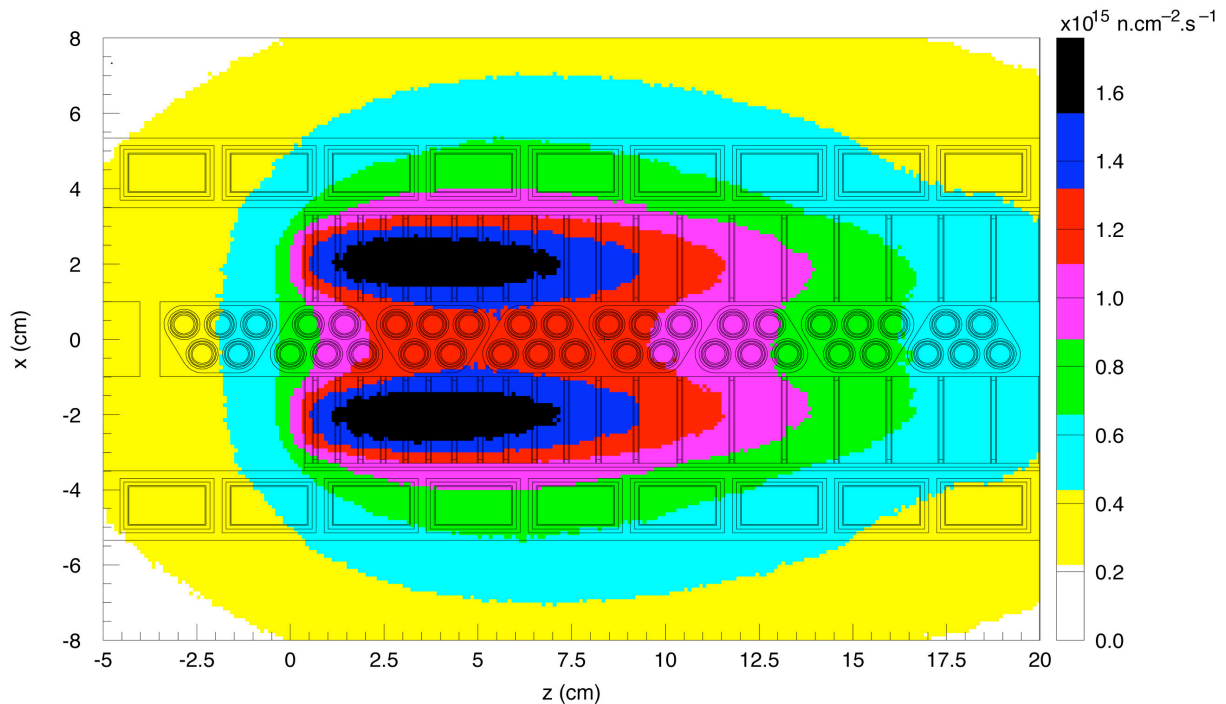


Figure 1. Spatial distribution of the fast neutron flux ($E > 0.1$ MeV) at target mid-plane.

2.3. Clad Irradiation Temperature

A critical parameter for obtaining meaningful irradiation data is the temperature at which the irradiations are performed. To achieve conditions similar to that of a fast reactor, the fuel cladding temperature must be controlled within a specified tolerance, and cladding temperatures up to 550°C must be attainable. These requirements are very difficult to achieve using water as the fuel coolant. After considering a number of coolant options, lead-bismuth was selected as the coolant for the test fuel pins. This coolant does not react exothermically with water or air (as is the case with sodium), has a high heat transfer coefficient, and is liquid over a large temperature range. It has the disadvantages of being a solid at room temperature (requiring trace heating on loop piping and components), requiring active oxygen control to reduce corrosion, and producing polonium-210 as an activation product. Polonium-210 is an alpha emitter with a 138-day half-life, and limiting its release during off-normal events will require special attention to the design of safety systems. The high-heat transfer coefficient of lead-bismuth yields a relatively low film drop ($\sim 30^{\circ}\text{C}$) that is predictable to within 30%. Thus, by measuring the inlet and outlet temperatures of the lead-bismuth, the temperature of the cladding encasing the fuel at any point along the fuel rodlet height should be known to within 10°C . The cladding temperature can be controlled by adjusting the lead-bismuth inlet temperature.

2.4. Neutron Energy Spectrum

Changes in reaction rates among nuclear reactions that arise from differences in neutron energy spectra will yield changes in isotopic concentrations as the fuel is burned. The consequences of differences in isotopic concentrations depend on several factors, including fuel initial composition, neutron spectrum, and how irradiation duration. One indicator that can be used to gauge the degree of differences between neutron spectra is spectrum-averaged cross sections. These are given in table 1 for three spectra: a sodium-cooled oxide-fueled fast reactor, the MTS, and the spectrum within a fresh cadmium basket inserted into a flux trap within Idaho National Laboratory's Advance Test Reactor (ATR) (taken from Chang and Ambrosek [3]). The cadmium basket absorbs the appreciable thermal neutron flux present in the ATR, and thereby shields test fuel loaded in the basket from these thermal

neutrons. The “shielded ATR” data are included here to compare MTS with this alternate method of simulating a fast reactor irradiation environment. The last two columns in table 1 show the ratio of the spectrum-averaged cross sections for the MTS and shielded ATR relative to a fast reactor. A ratio of unity indicates reaction rates for the test spectrum will be similar to that in a fast reactor. For fission reactions in fissile isotopes, we see that the MTS is fully prototypic of a fast reactor. The shielded ATR spectrum has fission cross sections that are 7 to 14 times greater, which allows the shielded ATR irradiations to achieve volumetric fission rates that are prototypic of fast reactors, even though the fast flux is well below prototypic fast reactor values. Capture rates for MTS are 70 to 80% of a fast reactor, which is very close but not fully prototypic. By contrast, the shielded ATR spectrum has capture rates that are 16 to 40 times greater than a fast reactor spectrum. This will result in differences in fuel composition for shielded ATR irradiations versus fast reactor irradiations. For example, fuel with significant ^{238}U can produce elevated concentrations of ^{239}Pu at the periphery, a phenomenon that can lead to the formation of a small-grained, high-porosity outer shell in high-burnup UO_2 fuel [4]. The situation is reversed for (n,2n) reactions: the shielded ATR spectrum has reaction rates that are similar (1 to 3 times greater) to those in a fast reactor, whereas the MTS has (n,2n) rates that are 10 to 60 times greater than a fast reactor. Note, however, that threshold reaction cross sections such as (n,2n) are 10 to 50 times smaller than fission and capture cross sections, which mitigates the extent of change in fuel composition. One example of how the larger (n,2n) cross sections for MTS irradiations can lead to non-prototypic fuel composition is enhanced production of ^{237}Np in UO_2 fuel.

Table 1. Spectrum-averaged cross sections for a fast reactor, MTS, and Cd-shielded ATR.

		Spectrum-Averaged Cross Section (Barns)			Ratio	
		Fast Reactor	MTS	ATR-shielded	$\frac{\text{MTS}}{\text{Fast Reactor}}$	$\frac{\text{ATR-shielded}}{\text{Fast Reactor}}$
Reaction	Isotope					
Fission	235U	1.3939	1.4052	10.31	1.01	7.40
	239Pu	1.6263	1.7527	11.02	1.08	6.78
	241Pu	1.8635	1.8128	25.18	0.97	13.51
Capture	235U	0.2784	0.2028	4.456	0.73	16.01
	238U	0.1593	0.1258	4.799	0.79	30.13
	237Np	0.7781	0.5451	26.58	0.70	34.16
	239Pu	0.1875	0.1379	5.545	0.74	29.57
	241Pu	0.2252	0.1698	7.835	0.75	34.80
	241Am	0.9829	0.6932	38.36	0.71	39.03
(n,2n)	235U	0.0019	0.0366	0.0030	19.06	1.54
	238U	0.0024	0.0531	0.0038	21.73	1.54
	237Np	0.0005	0.0203	0.0008	41.74	1.72
	239Pu	0.0006	0.0157	0.0016	28.54	2.83
	241Pu	0.0034	0.0323	0.0054	9.43	1.58
	241Am	0.0002	0.0093	0.0002	52.95	1.08

3. Irradiation Environment for Fusion Materials Testing

While designed for fast reactor fuels testing, the MTS irradiation environment is in many respects well suited for fusion materials testing. For this application, desirable attributes include helium and hydrogen production in steels, nuclear recoil spectrum, and elemental composition evolution with dose that are commensurate with that expected within the first wall of a fusion reactor. Other desirable

attributes include a high peak displacement rate in iron, high average facility availability, controlled sample irradiation temperature in the range of 400°C to 1000°C.

3.1. Hydrogen and Helium Production Rates in Steels

For fusion applications, the hydrogen and helium production rates are generally quoted as a ratio relative to the atomic displacement rate, which has units of displacements per atom (dpa) per unit time. For iron-based alloys, the conditions for a fusion reactor first wall are 10–15 appm He/dpa and 40–50 appm H/dpa [5]. For MTS, a broad range of He/dpa ratios are accessible in the irradiation regions, as shown in figure 2, where the He production rate is plotted as a function of the atomic displacement rate within 40-mm³ volume elements within the fuel irradiation (red data points) and materials irradiation (blue data points) regions. The black lines represent the fusion-relevant range of 10–15 appm He/dpa. Summing the data points within this fusion-relevant region produces the red line in figure 3, where minimum displacement rate is plotted as a function of irradiation volume. Also plotted is the minimum displacement rate versus irradiation volume for all He/dpa ratios (blue line). The broad range of He/dpa ratios in MTS will allow researchers to study the sensitivity of materials performance as a function of this important parameter.

In a fusion reactor first wall the hydrogen production rate in iron is about 4 times the He production rate, whereas in MTS this ratio is slightly larger, in the range of 5 to 7.

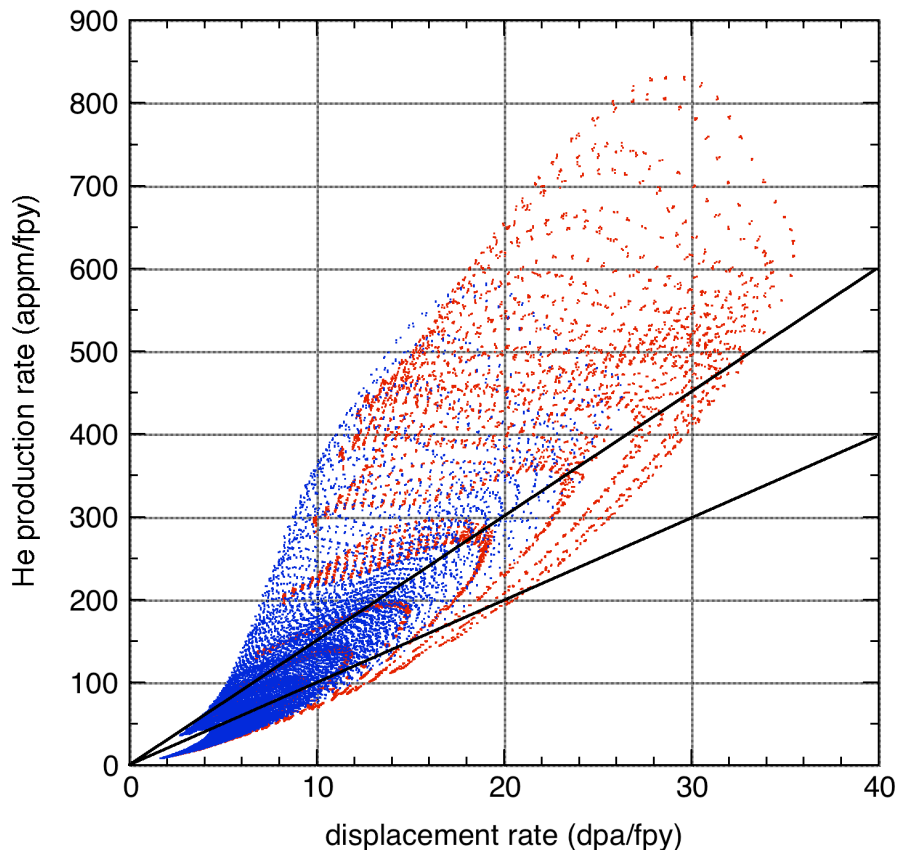


Figure 2. He production rate versus atomic displacement rate in the irradiation regions of MTS.

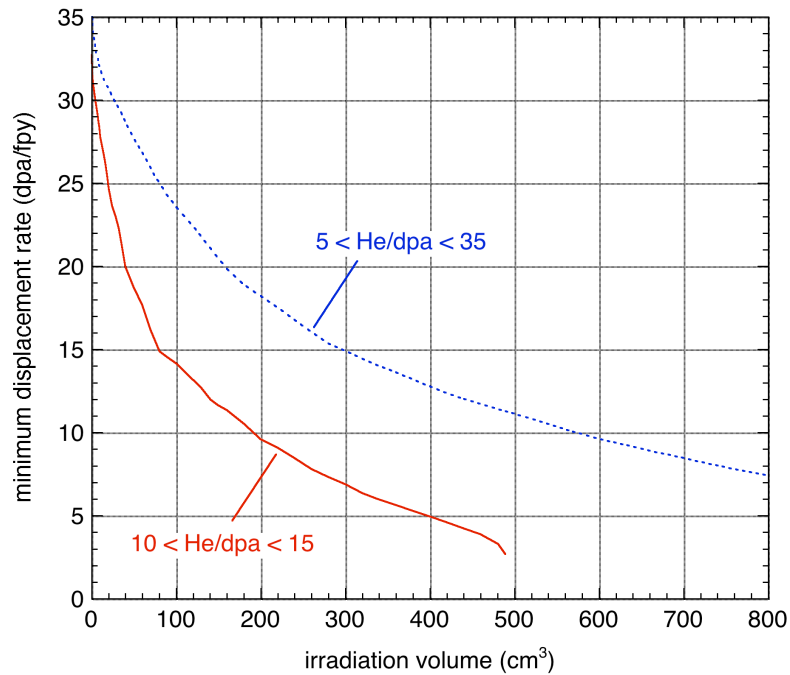


Figure 3. Minimum displacement rate versus irradiation volume for two ranges of He/dpa ratio.

3.2. Nuclear Recoil Spectrum

The recoil spectra in the materials irradiation region of MTS has been calculated and compared to that expected in the first wall of a fusion reactor [6]. Nuclear recoil spectra are typically compared using the damage production function W , which is the fraction of atomic displacements caused by recoils with kinetic energy T or less. The damage production function for MTS is compared to that of a fusion reactor first wall in figure 4. It shows the fraction of total displacements caused by nuclear recoils with energies less than 50 keV are nearly the same for both systems. This is an important result as Frenkel pairs produced by such low-energy recoils tend to be more isolated and so have lower probability of recombination as compared to the damage cascades produced by higher energy nuclear recoils. The functions for the two facilities diverge above 50 keV but this is likely not a serious concern as high-energy recoils tend to differ only in the number of sub-cascades they produce, but not in the morphology of the sub-cascades.

3.3. Elemental Composition Evolution

As materials are subjected to intense neutron radiation, neutron-induced nuclear reactions lead to changes in the elemental composition. For steels, the production of phosphorous and sulfur is a concern as studies have shown that low concentrations can embrittle ferritic-martensitic steels [7], which are leading candidates for fusion reactor applications. For the ferritic-martensitic steel EUROFER97 irradiated in MTS, the change in elemental composition of was calculated as a function of dose and compared to the change expected in the first wall of a fusion reactor [8]. Results are plotted in figure 5. It shows that at 200-dpa dose, the concentrations of phosphorous and sulfur are both below 10 appm, which are below the allowable concentrations for this alloy [9].

4. Conclusions

The irradiation environment in the MTS has been optimized to provide a high fast neutron flux over a volume sufficient for conducting fast reactor fuels testing at the rodlet scale. The neutron energy spectrum in the irradiation regions is similar to that of a fast reactor, with the addition of a high-energy tail. In terms of fuel irradiation, the high-energy tail leads to enhanced threshold reaction rates, such as

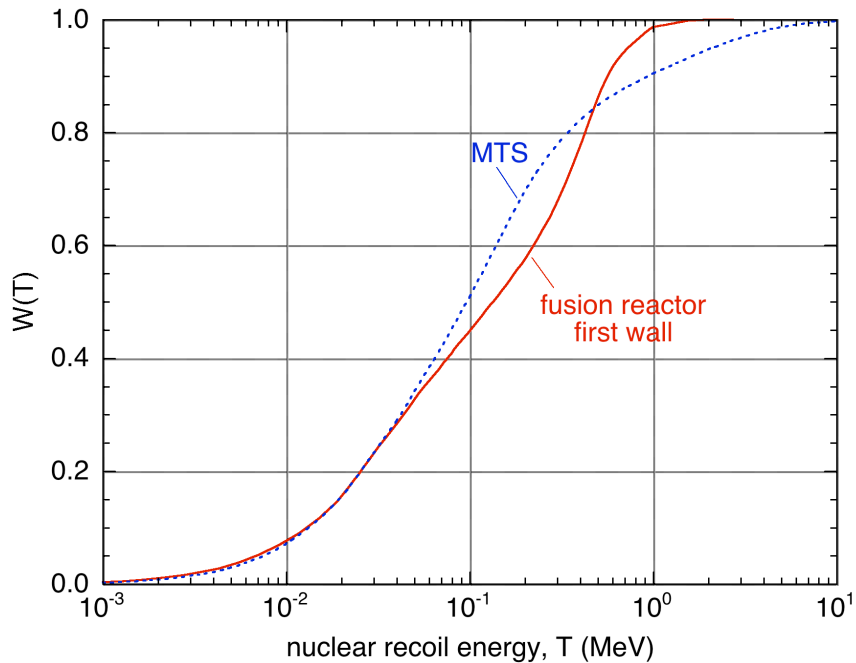


Figure 4. The damage production function W for MTS and a fusion reactor first wall.

(n,xn) and ($n,x\alpha$) whose effects are likely small, but ultimately depend on the fuel form and composition. The enhanced helium production arising from the ($n,x\alpha$) reactions gives MTS a helium/dpa ratio in the peak flux region similar to that expected in a fusion reactor first wall. The nuclear recoil spectrum below 50 keV is also similar to a fusion reactor, and the production of trace elements at high dose are sufficiently low so as to not be a concern in terms of enhanced embrittlement of ferritic-martensitic steels, which makes MTS well suited for fusion materials testing.

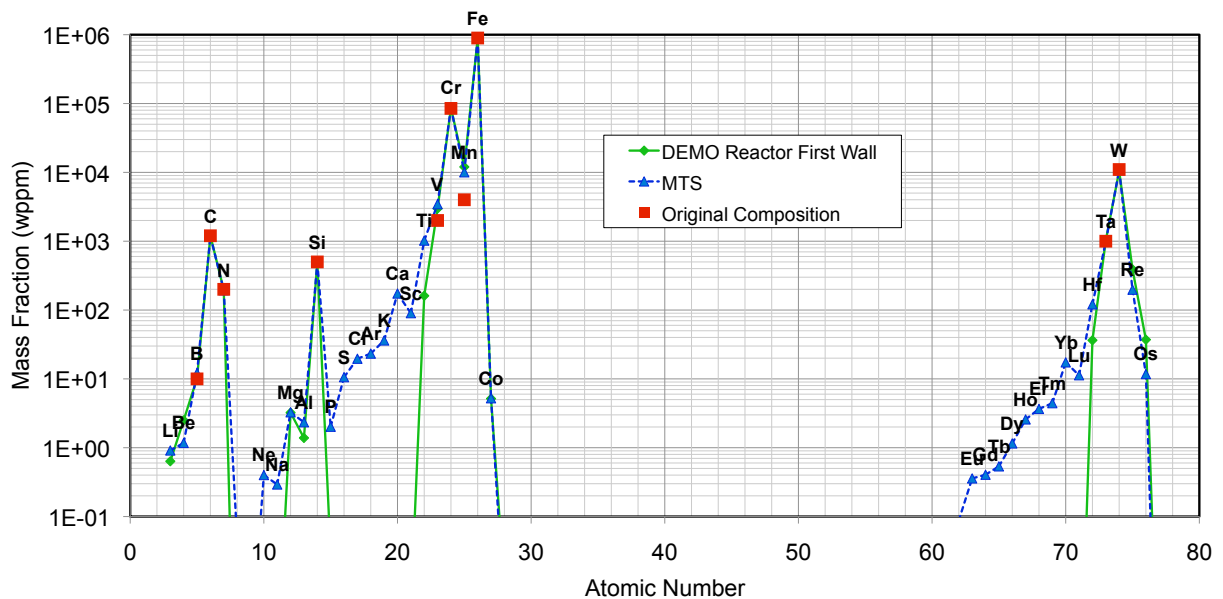


Figure 5. Elemental composition of EUROFER97 irradiated to a dose of 200 dpa in MTS and a fusion reactor.

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