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With the advent of the National Ignition Facility (NIF) increasing interest has focused on plasma physics processes that might be observed for the first time, in particular those mediated by electrons that could alter the populations of low-lying nuclear excited states. Inclusion of such processes into our simulation codes could impact our interpretation of radiochemical ratios measured during the underground test program if the nuclear cross sections proceeding on a ground state vs. a low lying nuclear excited state differ. (The radiochemical ratio ^{170}Tm/^{168}Tm is formed when (n,2n) and (n,γ) reactions that proceed on ^{169}Tm produce the radioactive species ^{168}Tm and ^{170}Tm. Such radiochemical ratios are used to infer the overall neutron fluence in a plasma environment that has experienced nuclear interactions). ^{169}Tm is an important nucleus for stockpile stewardship radiochemistry as well as being an *s*-process branch point in stellar nucleosynthesis theory.

There are several nuclear decay and excitation processes mediated by electromagnetic coupling with atomic electrons. In cold, neutral atoms, since there are no valence electrons and no inner shell vacancies, the only process that can occur is for the nuclear gamma decay energy to be transferred to a bound electron and typically ejected into the continuum (i.e. internal conversion, or IC). This process is well understood theoretically and experimentally. However, in a high temperature plasma environment, there are both free electrons and many inner-shell and outer-shell vacancies that make other nuclear processes possible. For example, nuclear decay can excite a bound electron to an unoccupied bound state (bound internal conversion, or BIC). Conversely, the inverse of each process can also occur in a plasma environment. The inverse of IC occurs when a free electron is captured into the atomic bound state and transfers the released energy to an excited state in the nucleus (i.e. nuclear excitation by electron capture, or NEEC [1]). The inverse of BIC occurs when the electron goes from a higher level to a lower level and resonantly transfers the decay energy to excite the nucleus (known as nuclear excitation via electron transition, or NEET [2]).

We recently explored for the first time inclusion of these electron-induced nuclear excited state population effects in a simulation of the NIF Rev5 ignition capsule [3] that incorporates ^{169}Tm as a radiochemical tracer. In our simulation we loaded $7.4\text{x}10^{14}$ atoms of ^{169}Tm into the innermost 5 μm ablator layer. With a 50-50 DT fuel mix the Rev5 capsule provides a yield of 15.7 MJ of energy and a neutron yield of $5\text{x}10^{18}$ n's.

Rates vs. Cross Sections & Flux

Figure 1 shows the electron-induced nuclear excitation rates (NEET + NEEC + γ-absorption) for the 8 keV ground to first excited state transition in ^{169}Tm tabulated over a range of electron temperatures ($2 < T_e < 20$ keV) and densities ($0.1 < \rho < 490$ g cm^{-3}). A similar plot showing electron-induced *de*-excitation rates (IC + BIC + γ–decay) is given in [4]. Overlaid on the rates is the thermodynamic evolution of the Rev5 capsule simulation. At peak burn (T_e=19.5 keV, ρ=319 g cm^{-3}) the excitation rate is $7.7\text{x}10^{-3}$ ns^{-1}, while the *de*-excitation rate is $6.0\text{x}10^{-3}$ ns^{-1}.

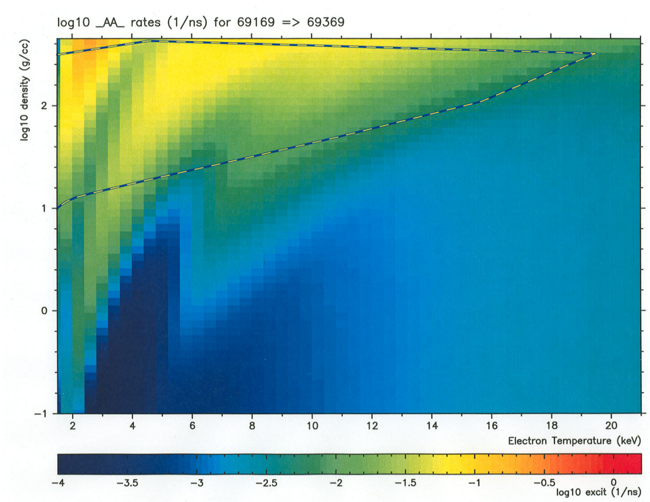

Figure 1. Electron-induced nuclear excitation rate as a function of electron temperature and density for the 8 keV ground to first excited state transition in ^{169}Tm.

Figure 2 shows the most important neutron-induced cross sections in this work as well as the instantaneous neutron flux sampled in the DT burn region at peak burn. These are but a small fraction of the full reaction network used for this survey [4], but they illustrate the main points. For each channel we show the cross section proceeding on the ground state (GS: solid line) and on the 8 keV first excited state (M1: dashed line) of ^{169}Tm. Note that the (n,n') cross section proceeding on the (loaded) ground state will preferentially populate the excited state for neutron energies > 1 MeV. Population by either the electrons or neutrons would lead to an enhancement of ^{170}Tm due to the 30% larger (n,γ) cross section that proceeds on the ^{169}Tm first excited state, thus providing a potential radiochemical signal. By contrast, the same amount of ^{168}Tm would be made no matter the distribution of ^{169}Tm ground or first excited state population due to the identical (n,2n) cross sections. We will explore the difference in resulting radiochemical ratios with the electron excitation rates turned on and turned off.

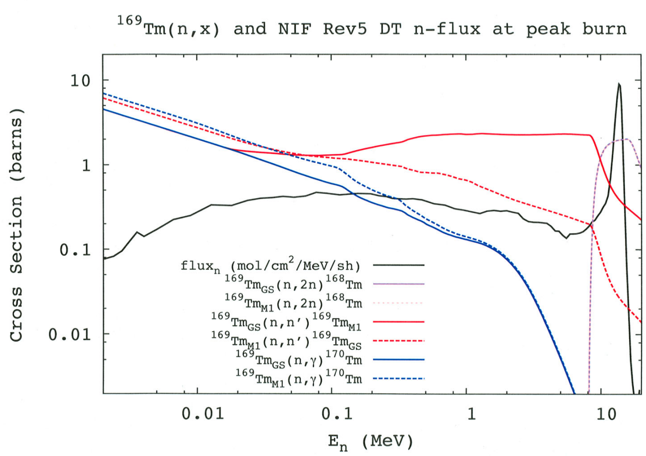

Figure 2. Neutron-induced cross sections on ground and first excited state targets of ^{169}Tm and the neutron flux at peak burn in the NIF Rev5 ignition capsule.

Results

Figure 3 shows the temporal evolution in our Rev5 DT burning capsule simulation (offset at peak burn time = 0 ns) of the scaled quantities: electron temperature (T_e, red), density (ρ, black), and the population of the 8 keV first excited state of ^{169}Tm (X_{M1}, in green). We have normalized the initial population of the ground state by the loaded amount to make X_{GS} = 1, and present all populations in terms of mass fraction. At peak burn X_{M1} = 0.017, or 1.7%. Also shown are three *nuclear flows* (the product of a target state population and a "reaction rate") with which we contrast the contribution to X_{M1} due to the neutrons and electrons.

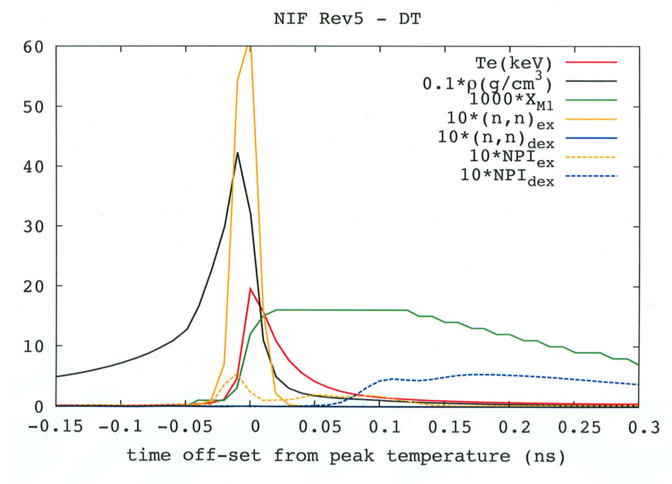

Figure 3. Quantities affecting the population of the 8 keV first excited state in ^{169}Tm.

The solid gold curve is the neutron excitation flow $(n,n)_{ex}$ (X_{GS} times the integral over all neutron energies of the product of the flux and the (n,n') cross section in Figure 2). The dashed gold curve shows the electron-induced excitation flow NPI_{ex} (X_{GS} times the excitation rate in Figure 1). Over the duration of the burn the neutrons clearly dominate the contribution to X_{M1}. The dashed blue curve is the electron *de*-excitation flow NPI_{dex} that *de*-populates X_{M1} after the capsule comes apart. The *de*-excitaion flow for the neutrons is very small (due to a small X_{M1}).

The populations of ^{170}Tm and ^{168}Tm at the end of our NIF Rev5 DT simulation with the electron rates turned on were 2.4×10^{-4} and 3.64×10^{-2}, respectively, making for a radiochemical ratio of ^{170}Tm/^{168}Tm = 6.6×10^{-3}. *These quantities were identical with the electron-induced rates turned off.* This was not because the capsule didn't get hot (at ~20 keV it did), or that the neutron flux was small (it wasn't), but rather there was no *time* for either excitation flow to populate X_{M1} enough to allow for substantial neutron capture to take place on the excited state (T_e was above 1 keV for only 0.14 ns). However, if one could maintain peak conditions long enough for the electron excitation and *de*-excitation rates to come into equilibrium (73 ns) X_{M1} would reach

36%, more than enough to distinguish a difference in the radiochemical ratio due to enhanced neutron capture when including the electron rates (vs. not).

In conclusion, the Rev5 capsule design on NIF, with its short DT burn timescale, is not a viable platform to observe NEEC or NEET using the ^{169}Tm first excited state transition.

References

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