Comparison of Composite and Water Moderator Performance at SNS SNS/TSR-202

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This document describes the neutronic performance characteristics predicted for a composite hydrogen-water moderator at the Spallation Neutron Source.

1 Background

High-power spallation neutron sources, such as the Spallation Neutron Source Project to be constructed at Oak Ridge National Laboratory, have large radiation damage rates on most of the neutronically significant components, including the moderators. This fact is especially significant for moderators composed of liquid methane. In the liquid state (around 100 K) methane suffers from severe problems with radiation-induced polymerization, to such a level that the highest-power existing spallation sources must significantly limit the lifetime of their liquid methane moderator vessels, imposing significant constraints on operational strategies for the facility.[1] At damage rates corresponding to the Spallation Neutron Source (2 MW of proton power on a mercury target), the use of liquid methane does not currently appear to be practical, even though it provides performance characteristics generally agreed to be highly desirable. In fact, most "new" spallation source projects, when they develop proposed instrument suites, assign approximately half of the initial instruments to liquid methane moderators when that choice is offered.[2] As no practicable means to use liquid methane at SNS-like damage rates is known, we have explored an alternative moderator concept, the hydrogen-water composite moderator, as a surrogate for liquid methane on high-power spallation neutron sources which would be tolerant of radiation, given that neither hydrogen nor water are significantly subject to radiation damage. Although the inspiration and initial goal of our explorations was to investigate the composite moderator as a surrogate for liquid methane, we also consider it as a new type of moderator, one which has its own unique performance characteristics, and may be desirable in its own right.

2 Composite Moderator Desiderata

The characteristics that make liquid methane (or any moderator) useful and desirable include:

- the pulse shape; certain features are more desirable at different wavelengths, for different reasons, in different applications:

[1] G. M. Allen et al., "The ISIS Cold Moderators," *Proceedings of the International Workshop on Cold Moderators for Pulsed Neutron Sources*, OECD (1999).

[2] G. S. Bauer, "Pulse Neutron Source Cold Moderators—Concepts, Design and Engineering," *Proceedings of the International Workshop on Cold Moderators for Pulsed Neutron Sources*, OECD (1999).

  - narrow or precise features show up best with sharp pulse shapes,
  - broad features can be characterized by broader pulse shapes, which nominally have more neutrons,
  - quickly decaying "tails" increase signal-to-noise ratios,

- the spectral distribution; if resolution needs of the instrument are satisfied, more neutrons at the right energies and the right times are almost always a good thing.

Because the hydrogen density in liquid methane is very comparable to that of water, the slowing-down behavior of the two materials is nearly identical, and at high energies (e.g., 10 eV) the pulse shapes are essentially indistinguishable. Both liquid methane and water are thermalizing moderators; moderators which develop a "storage term" (i.e., the Maxwellian spectrum) that persists for relatively long times as the neutrons maintain a thermal equilibrium with the hydrogen in the moderator and gradually leak out. Because the temperature of liquid methane is lower than that of water, however, the characteristic slowing-down pulse shape persists to lower energies. This transition energy is approximately 160 meV (0.7 Å) in water, and around 30 meV (1.5 Å) for methane. Thus liquid methane gives sharper, narrower (slowing-down) pulses, when compared to water, at wavelengths corresponding to, for example, much of powder diffraction (0.5–2 Å), making it ideal for that task. Furthermore, since the spectral temperature for liquid methane is reduced by a factor of three from water, the low-energy neutron flux goes up a factor of five to six, resulting in significantly more neutrons in the increasingly popular low energy-long wavelength ranges. Although the above-mentioned Maxwellian storage term gives significantly higher intensities at the corresponding neutron energies, it also results in a characteristically long exponentially decaying pulse shape, typically with a time constant $\tau \approx$ 20–40 μs for poisoned water or poisoned liquid methane. This can be a disadvantage when compared to a so-called non-thermalizing moderator, in which there is no storage term, and thus no long-lived decay (as well as significantly reduced time-averaged intensity). Liquid parahydrogen is an example of such a moderator.

In general, we can then list a number of characteristics we would like to obtain from a composite moderator intended as a surrogate for liquid methane:

- narrow pulse shapes in the slowing down region ($\delta t \cdot v \approx$ 28 mm for both water and liquid methane),
- narrow pulse shapes, relatively invariant with energy, in the thermalized region, for which the width is controllable with appropriate poisoning ($\delta t \approx$ 30–60 μs for water and liquid methane, depending on poison depth),
- slowing-down-to-thermal transition energy similar to that of liquid methane (30 meV),
- pulse shapes with sharp rising edges,
- exponentially decaying tails no worse than water or liquid methane ($\tau \approx$ 20–40 μs for water and liquid methane, depending on poison depth), and
- more low-energy neutrons.

3 Composite Moderator Description

Our concept for a hydrogen-water composite moderator is one in which the viewed face of the moderator is a thin layer of hydrogen at 20 K, closely backed by a fairly thick layer of water at 300 K. As this water layer is optically thick, and would quite likely be poisoned for pulse width control, the moderator can be arranged symmetrically, as shown in Figure 1, where hydrogen layers form each viewed surface. In the composite moderator configuration shown in Figure 1 each viewed face is a 12.5 mm-thick layer of supercritical hydrogen. Each hydrogen layer is

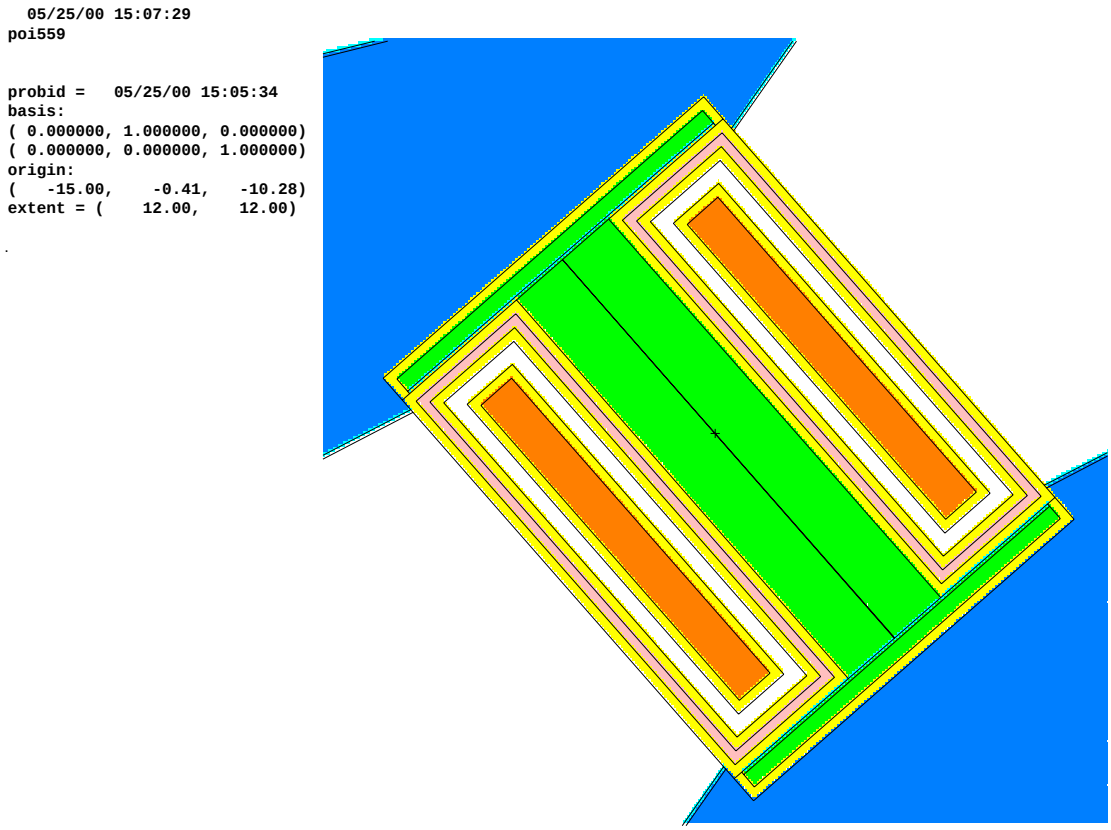

Figure 1: Composite moderator configuration. The viewed layers are supercritical hydrogen, the central region is liquid water. The central line shows the base configuration poison location.

closely backed by a central layer of water 37.5 mm thick at 300 K. The moderator is decoupled with cadmium from the reflector. The studies reported here examine two poisoning configurations, one without poison, and one with poison in the center of the water region, some 18 mm beneath the surface of the water layer, and thus some 31 mm within the "moderator material region" of the assembly. Other poison configurations have been studied; comparisons between different poison scenarios for the composite moderator appear elsewhere. This geometry of hydrogen and water was developed as part of an optimization process in which the spectrum of the composite moderator was made, as much as possible, similar to that of a nominal liquid methane moderator.[3]

[3] L. A. Charlton et al., "Neutronic Moderator Design for the Spallation Neutron Source," *Proceedings of the 2nd International Topical Meeting on Nuclear Applications of Accelerator Technology (AccApp'98)*, American Nuclear Society (1998).

4 Overall Model Description

We used a realistic neutronic model to study the performance of the composite moderator, one which includes four moderators in total. Two of these moderators are viewed from both sides, one of which (the bottom upstream) is the composite. All moderators have nominal viewed faces of 100 mm (horizontal) by 120 mm (vertical). The inner reflector (out to a radius of ≈320 mm) is beryllium and is cooled with heavy water. This inner reflector is surrounded by heavy water-cooled lead. Table 1 summarizes relevant characteristics of the target station configuration used for this set of calculations, and Table 2 summarizes the base moderator configuration.

Proton Energy	1 GeV
Pulse Rate	60 Hz
Average Power	2 MW
Energy per pulse	34 kJ
Proton Beam Shape	rectangular
Proton Beam Size	200x70 mm^2
Proton Pulse	$\delta(t)$
Target	Hg
Inner Reflector	Be
Inner Reflector Coolant	D_2O
Outer Reflector	Pb
Outer Reflector Coolant	D_2O

Table 1: Target station parameters used in calculations. All normalizations are performed per 34 kJ-pulse.

Beam-line	Moderator Location	Moderator Material	Temperature (K)	Decoupling Material	Poison Material	Poison Depth (mm)
2	TU	H_2	20	Cd	Gd	27
5	TD	H_2	20	–	–	–
8	BU	Composite	–	Cd	Gd	31
11	TU	H_2	20	Cd	Gd	27
14	BD	H_2O	300	Cd	Gd	27
17	BU	Composite	–	Cd	Gd	31

Table 2: The moderators in the base design configuration. Hydrogen is 20–27 K supercritical, modeled as 20 K liquid. Water is 300 K liquid.

The top upstream moderator is cadmium-decoupled hydrogen at 20 K and has curved viewed surfaces. The moderator material has a maximum thickness of 65 mm, and an average thickness of about 55 mm. The moderator is poisoned with gadolinium at the centerline and is viewed from both sides. The top downstream moderator is fully coupled unpoisoned hydrogen at 20 K, and is viewed from one side only. This moderator also has a curved viewed surface, with maximum thickness of 65 mm, and an average thickness of about 55 mm. This moderator also has approximately 20 mm of light water surrounding it as premoderator. As described above, the bottom upstream moderator is a hydrogen-water composite, viewed from both sides, in which a 12.5 mm layer of hydrogen at 20 K forms each viewed surface, as shown in Figure 1. Each hydrogen layer is closely backed by the central layer of

water 37.5 mm thick at 300 K. The moderator is decoupled with cadmium. In the base configuration, the moderator is poisoned with gadolinium in the center of the water layer, while in the variant configuration, the moderator is un-poisoned. The bottom downstream moderator is cadmium-decoupled, gadolinium-poisoned water at 300 K, and has a curved viewed surface. The moderator material has a maximum thickness of 65 mm, and an average thickness of about 55 mm. The moderator is poisoned with gadolinium at the centerline and is viewed from both sides.

Although each of the four moderators above was included in the model used to calculate the composite performance, the performance of those moderators is not what is described below. The moderator performances described below are those for different moderators in the bottom upstream position; each case should nominally be considered mutually exclusive, although any given moderator might nominally appear elsewhere in the system. The characteristics of the moderators compared appear in Table 3.

ID Code	Moderator Material	Temperature (K)	Decoupling Material	Poison Material	Poison Depth (mm)
source_poi503_bu_cmpsPO	Composite	–	Cd	Gd	31
source_poi203_bu_cmpsit	Composite	–	Cd	–	–
source_poi264_bu_decH2O	Water	300	Cd	Gd	27
source_poi203_bu_decLqM	Liquid Methane	100	Cd	Gd	25

Table 3: Moderator options studied for the bottom upstream position, and ID codes for corresponding data files.

Liquid methane as a moderator material is included in this comparison; this should not be taken as an indication that liquid methane is under active consideration for this moderator, but rather as a reference, since the original intent of the composite moderator optimization was to emulate liquid methane's performance. Note that comparisons with other moderators are appropriate as well; one in particular is decoupled poisoned hydrogen, which will be present elsewhere in the system. That particular comparison is available elsewhere.[4]

5 Results

Figure 2 shows the pulse shapes calculated for 10 meV neutrons, well within the thermalized region for both water and methane moderators. It is immediately apparent that the composite moderator has a significantly broader pulse shape than either water or methane, although the degree of that increase depends upon the poisoning. Although the unpoisoned composite moderator intensity at 10 meV is identical to that of the methane moderator, the increased width also implies a decrease in peak intensity, by definition an undesirable effect. Furthermore, the nature of the peak intensity is somewhat different for the composite, in that there is a broad plateau region, not very useful for precise position location.

Figure 3 shows the pulse shapes calculated for 100 meV neutrons, within the thermalized region for a water moderator but not for a methane moderator. The composite moderator, whether poisoned or not, seems to clearly display the undesirable characteristic signs of thermalization—a relatively broad, slowly decaying pulse, along with significantly less intensity, both peak and averaged, than the also-thermalized water moderator. Even though the composite has 2–3 times the averaged intensity as the methane moderator, the peak intensity for methane is still higher. Finally, we see that the plateau in the peak intensity region observed at lower energies has now resolved itself into a "double-peak" structure, representing an extremely difficult pulse shape to parametrically describe.

[4] E. B. Iverson, "Detailed SNS Neutronics Calculations for Scattering Instrument Design," SNS/TSR-203, 2000

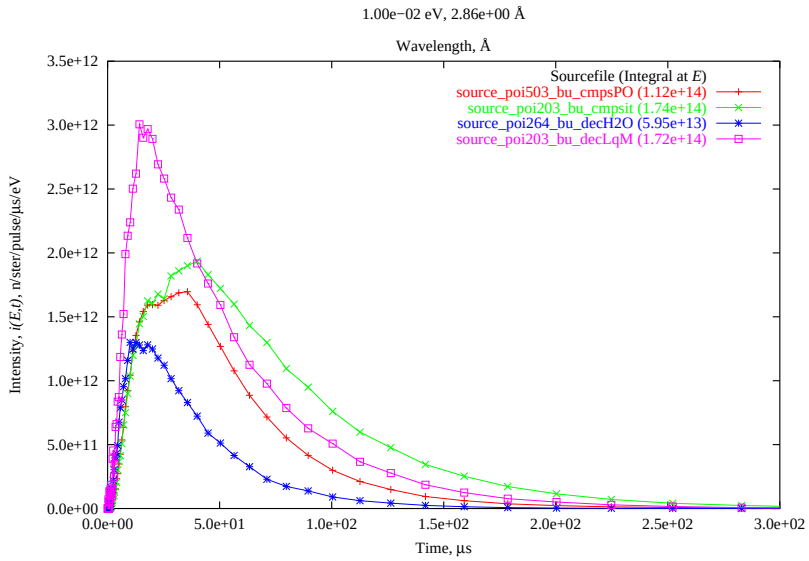

Figure 2: E =10 meV emission time distribution (pulse shape). All moderators in thermal region. The poisoned composite moderator is denoted source_poi503_bu_cmpsPO, the unpoisoned composite source_poi203_bu_cmpsit, the poisoned water source_poi264_bu_decH2O, and the poisoned liquid methane source_poi203_bu_decLqM. $\Delta E/E \approx 23$ %.

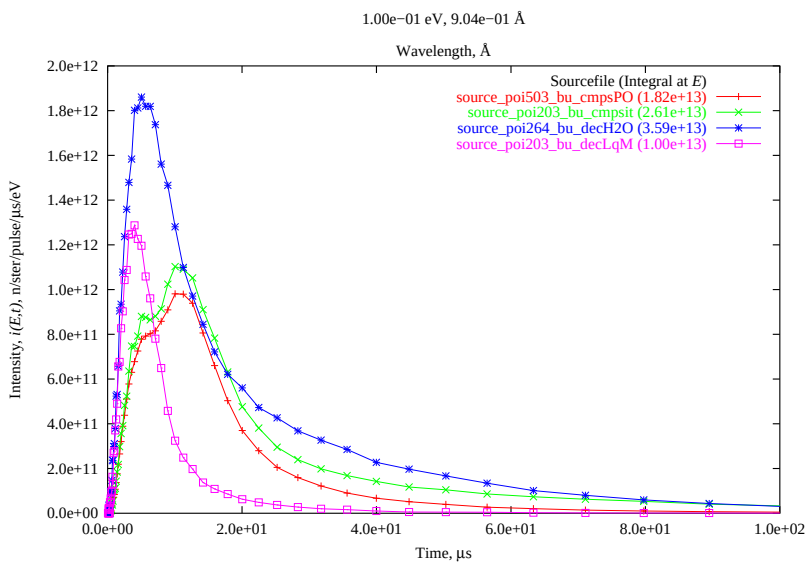

Figure 3: E =100 meV emission time distribution (pulse shape). Composite and water moderators in thermal region, liquid methane in slowing-down region. The poisoned composite moderator is denoted source_poi503_bu_cmpsPO, the unpoisoned composite source_poi203_bu_cmpsit, the poisoned water source_poi264_bu_decH2O, and the poisoned liquid methane source_poi203_bu_decLqM. $\Delta E/E \approx 23$ %.

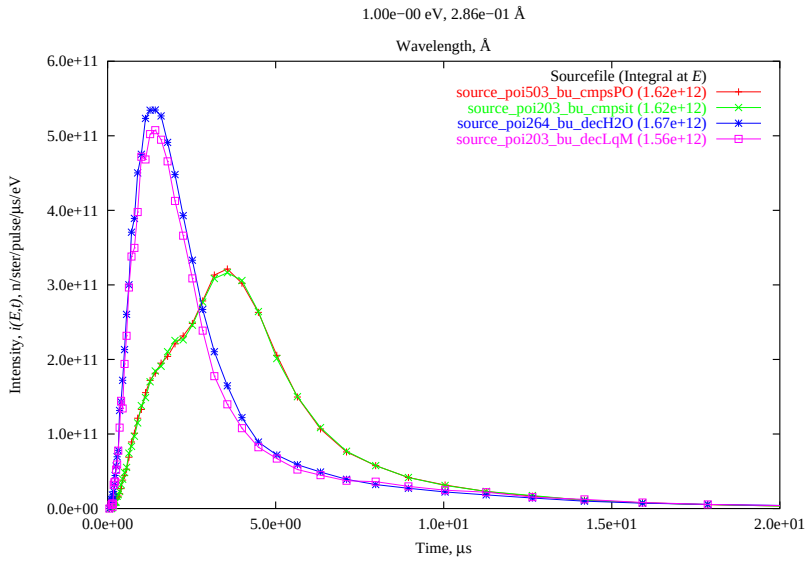

Figure 4: $E = 1$ eV emission time distribution (pulse shape). All moderators in slowing-down region. The poisoned composite moderator is denoted source_poi503_bu_cmpsPO, the unpoisoned composite source_poi203_bu_cmpsit, the poisoned water source_poi264_bu_decH2O, and the poisoned liquid methane source_poi203_bu_decLqM. $\Delta E/E \approx 23$ %.

Figure 4 shows the pulse shapes calculated for 1 eV neutrons, within the slowing-down region for all moderators. The water and methane pulse shapes appear to be functionally identical, as expected. The composite pulse shape on the other hand is extremely unpleasant. At 1 eV the (gadolinium) poison is of course no longer relevant. Although the time-averaged intensity is equivalent for all moderators, the pulse shape for the composite is significantly broader than for water or methane, leading to a significant reduction in peak intensity. This is somewhat counter-intuitive, as one might think of a composite moderator in a homogenized sense, having a hydrogen density somewhat between that of water and liquid hydrogen, and thus a slowing-down time behavior somewhat between that of water and hydrogen. However, the oddly-shaped pulse shape observed above is still present. Furthermore, we can now easily note that the rising edge of the pulse shape is significantly slower than that of water or methane; in fact, it is nearly identical to that of liquid hydrogen.

Figure 5, showing the pulse width (FWHM) as a function of energy, further demonstrates the onset of thermalization in the composite moderator, as indicated by the sharp increase in pulse width, at energies very similar to that for water rather than methane. Additionally, the composite moderator in general gives significantly broader pulse shapes overall, even in the typically narrow slowing-down region. Finally, we can see that the low-energy-asymptotic behavior of the pulse width provides additional indication that the composite moderator is a “thermalizing” moderator.

Figure 6, however, shows that the composite moderator, while having quite good time-averaged intensity (always superior to methane while competing very well with water), does not have a characteristic Maxwellian shape, and thus might not be considered a thermalizing moderator by most definitions.

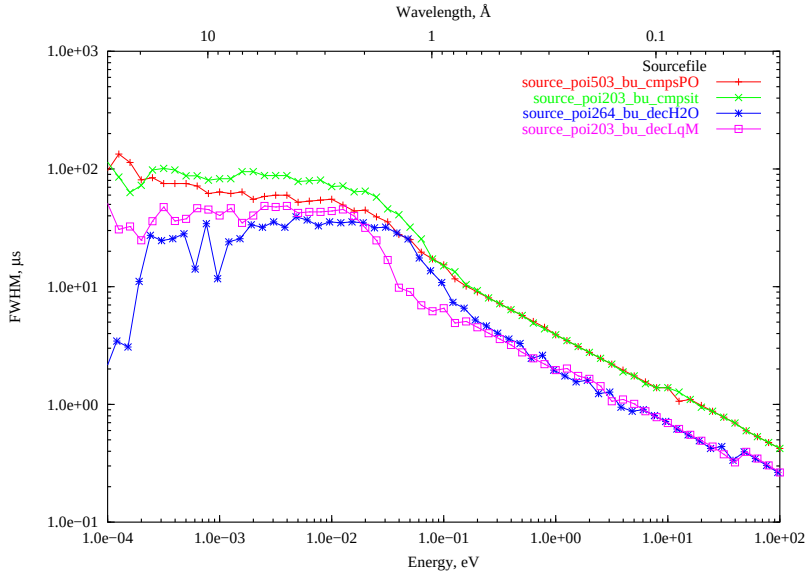

Figure 5: Pulse width for compared moderators. The poisoned composite moderator is denoted source_poi503_bu_cmpsPO, the unpoisoned composite source_poi203_bu_cmpsit, the poisoned water source_poi264_bu_decH2O, and the poisoned liquid methane source_poi203_bu_decLqM. Statistics are poor at low energies.

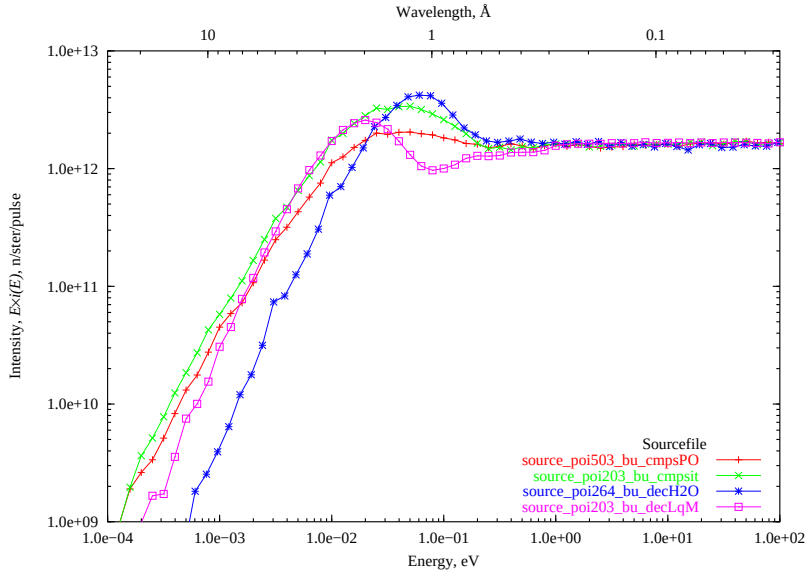

Figure 6: Intensity per unit lethargy for compared moderators. The poisoned composite moderator is denoted source_poi503_bu_cmpsPO, the unpoisoned composite source_poi203_bu_cmpsit, the poisoned water source_poi264_bu_decH2O, and the poisoned liquid methane source_poi203_bu_decLqM.

6 Discussion

If we consider again the desiderata mentioned earlier, we find that the composite moderator performance is not particularly satisfactory. Although it does match the low-energy intensity of liquid methane, and even further provides (at least in the design phase) some capability for tuning of the spectral intensity as a function of energy, the characteristic performance of the moderator is much more like that of water, with some of the undesirable characteristics of liquid hydrogen, as well as some unique undesirable tendencies.

The composite moderator:

- displays broader pulse shapes at all energies than either water or methane,
- transitions to a thermalization regime like water,
- has slow rise times like liquid hydrogen, and
- displays a moderately bimodal pulse shape at medium-to-high energies.

As a result, higher resolution information (in scattering experiments using a composite moderator) will not be present, and the pulse shapes at shorter wavelengths will be very difficult to fit.

The origin of the composite moderator behavior is not particularly well-known. The (para-) hydrogen layer overlaying the water backing is rather transparent, transmitting approximately 30% at 1 eV (uncollided) and 97% below 15 meV. This might imply that the "water peak" is visible, but overlaps the "hydrogen" peak by just enough that the effect is a broader, rather than bimodal, pulse. Thus if we think of the composite moderator pulse shape as some sort of superposition of a thin hydrogen pulse shape and a typical water pulse shape, we might expect to see a broadened, oddly-shaped pulse shape, as indeed we do. Another potential explanation is that the very tightly coupled hydrogen and water layers, when treated as a two-region heterogenous relaxing system after the methods of Carpenter et al.[5] will, on the basis of interactions between the two regions, display a ringing-like phenomenon. This sort of behavior would also lead to significantly broadened slowing-down pulse shapes, as well as oddly-shaped pulse shapes, both characteristics which we observe.

Although a detailed analysis of the serviceability of the composite moderator is beyond the scope of this document, there are some conclusions that seem inescapable. First of all, the composite moderator does not mimic liquid methane. The slowing-down properties, thermalization properties, and general pulse shape characteristics are fundamentally different. Furthermore, the composite moderator performance displays very few canonically desirable features, and even then only the somewhat less important ones. It seems likely that any scattering instrument design effort seeking a moderate-to-high resolution moderator to use would choose water or liquid hydrogen, already available elsewhere in the SNS facility, rather than choosing the composite moderator.

On the other hand, the composite moderator does give a significantly different spectral shape than either conventional thermalizing or non-thermalizing moderators. This spectrum provides significant improvements in time-averaged intensity across a large range in energy, *if instruments can exploit it.* Some neutron scattering instruments do not rely on peak shape analysis, are not limited in their resolution by the moderator performance, and might be very well served by the simultaneous increase in intensity at low and medium energies (i.e., 1 and 100 meV).

7 Calculational Techniques

The simulations reported are the results of calculations using the MCNPX code from LANL (version 2.1.5). The spectral intensities shown result from calculations using point detector tallies located 5 m from the viewed

[5] J. M. Carpenter, Y. F. Takeda, and D. F. R. Mildner, "Spatial Eigenfunctions, Time Eigenvalues, and Leakage Currents of Two-Region (Grooved) Neutron Moderators," *Nuclear Science and Engineering* 98 (1988), p. 326.

surface of the moderator. The emission time distributions (pulse shapes) come from current tallies on the viewed surface of the moderator material, and are averaged over 2π steradians. Weight windows used to accelerate the pulse shape calculations were generated by separate iterative runs using MCNP 4B in parallel mode on a large cluster of machines with a neutron-only source term. MCNPX runs using these weight windows produced the reported results, which have further been scaled (from the point detector calculations) to correspond to the peak intensity coming off of the moderator face in the normal direction, rather than the average over 2π steradians. Each moderator nominally requires a unique set of weight windows, and thus a unique set of runs. In some runs, however, the results for moderators other than the one for which the weight windows were optimized are adequately sampled.

8 Extrapolation

The results of the various simulations are reported, both here and in the source files, over a broad range of energies. In the event that results for energies outside this range are desired, certain extrapolations are reasonable. Spectra can be extrapolated to higher energies by using a simple power law, as they are very nearly, but not exactly, $1/E$ up to energies of approximately 100 keV. Emission time distributions for higher energies (in the slowing-down region) can in general be assumed to be invariant as a function of vt (velocity multiplied by time), equivalent to t/λ. However, the proton pulse at SNS is not actually a delta function in time, but rather has a width of a few hundred nanoseconds. Neutron pulse shapes for energies above 3 eV or so will be influenced by this proton pulse shape, while neutron pulse shapes for energies above 40 eV or so will be completely dominated by the proton pulse shape, and thus will be invariant as a function of time. At low energies, the spectral intensity from water or methane moderators can be assumed to follow a Maxwellian distribution. The low-energy intensity from the composite moderators can be extrapolated with a power law relationship from the data shown, and will likely not correspond to a Maxwellian distribution.

9 Data Availability

These results are available electronically as “source files;” ASCII files containing the spectra and emission time distributions, with comments showing the file format. Each moderator is represented by a single source file, the names of which are based on the ID codes indicated in Table 3. These source files can be downloaded from http://www.sns.anl.gov under “R&D Projects.”

10 Detailed Pulse Shapes

The detailed pulse shapes as produced by the simulation appear below. These data are completely un-processed, and appear exactly as they result from the calculations. The results are for different moderators, as described in Table 3, each in the bottom upstream position.

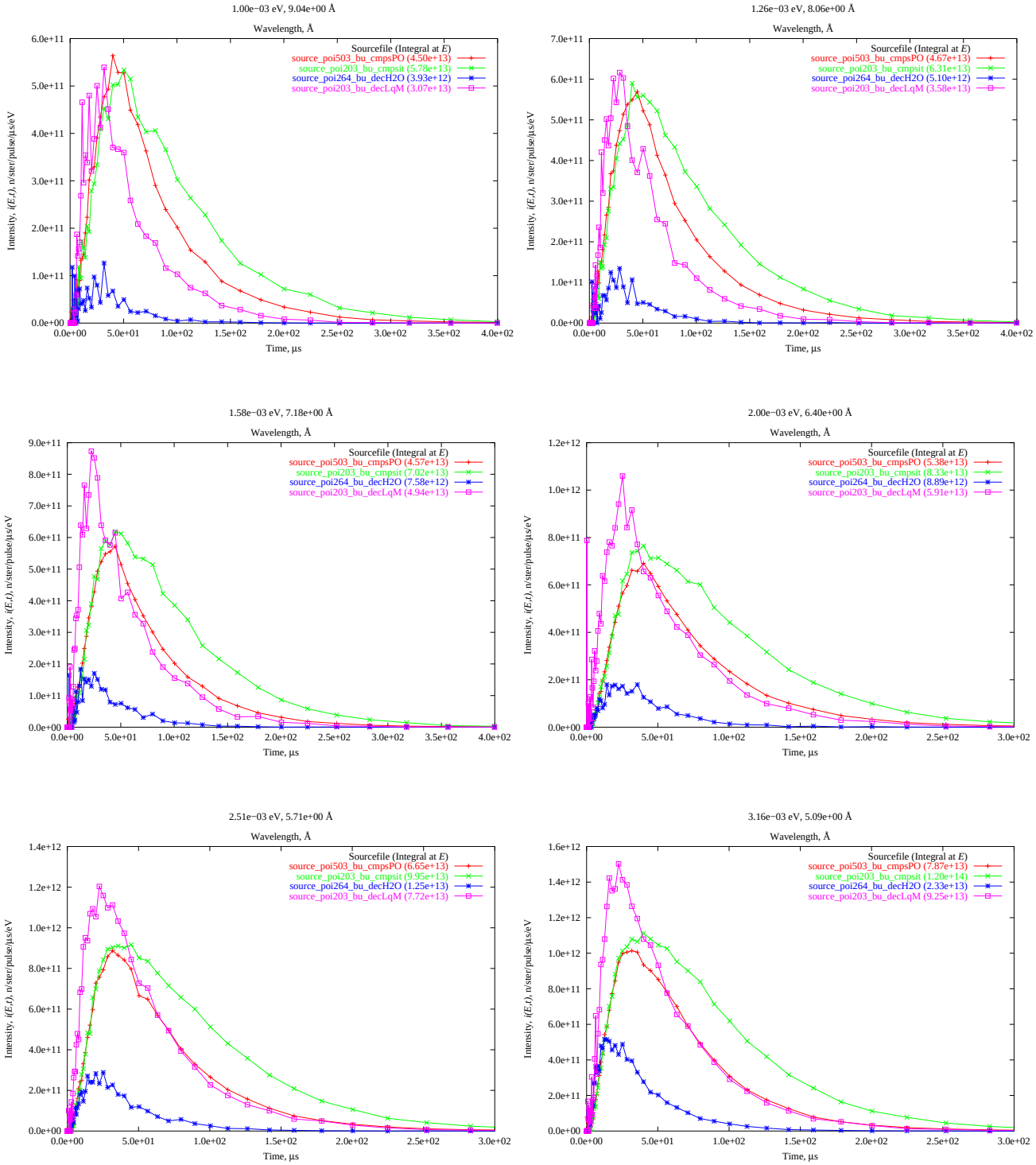

Figure 7: Emission time distributions. The poisoned composite moderator is denoted source_poi503_bu_cmpsPO, the unpoisoned composite source_poi203_bu_cmpsit, the poisoned water source_poi264_bu_decH2O, and the poisoned liquid methane source_poi203_bu_decLqM. $\Delta E/E \approx 23$ %.

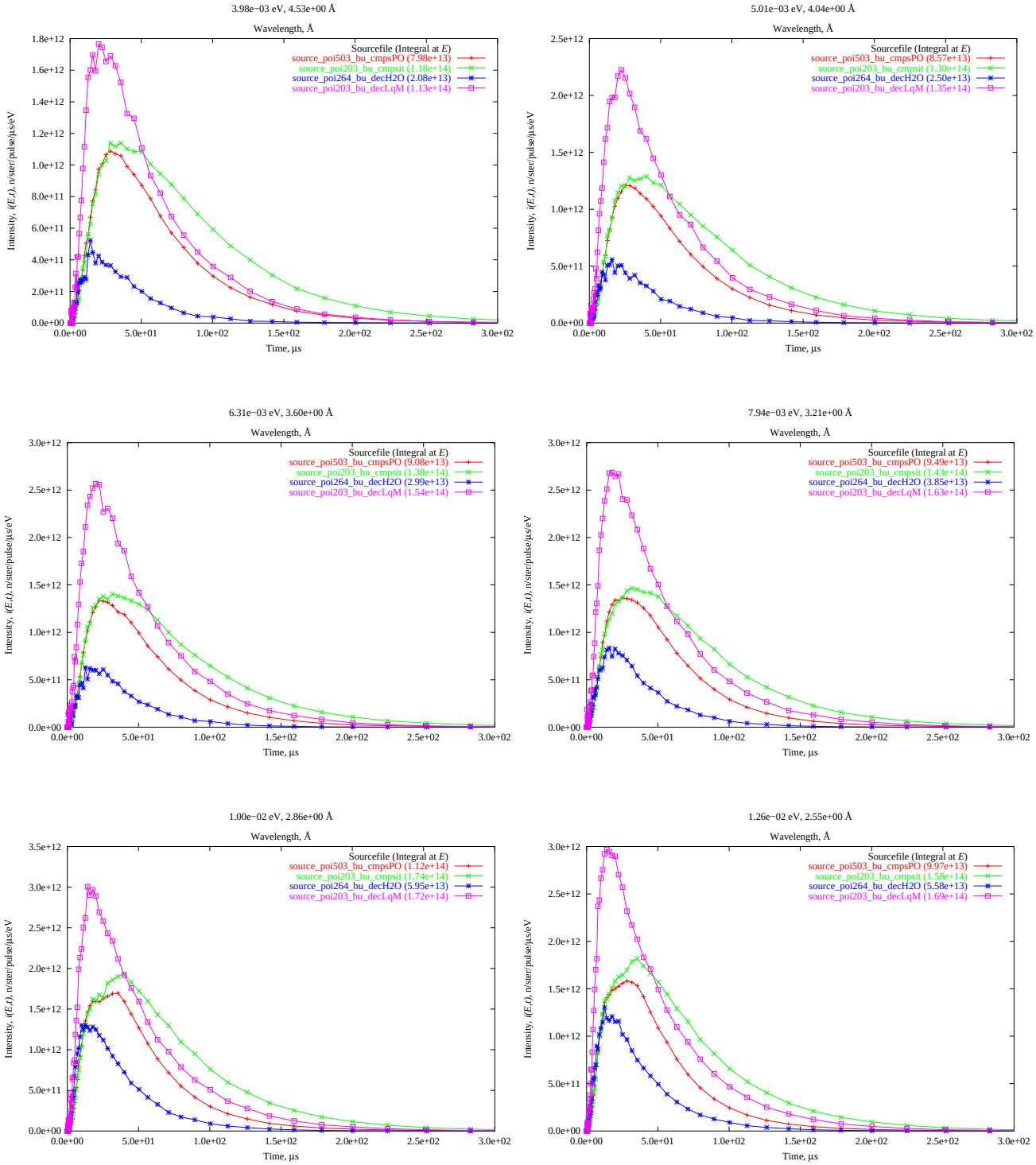

Figure 8: Emission time distributions. The poisoned composite moderator is denoted source_poi503_bu_cmpsPO, the unpoisoned composite source_poi203_bu_cmpsit, the poisoned water source_poi264_bu_decH2O, and the poisoned liquid methane source_poi203_bu_decLqM. $\Delta E/E \approx 23\,\%$.

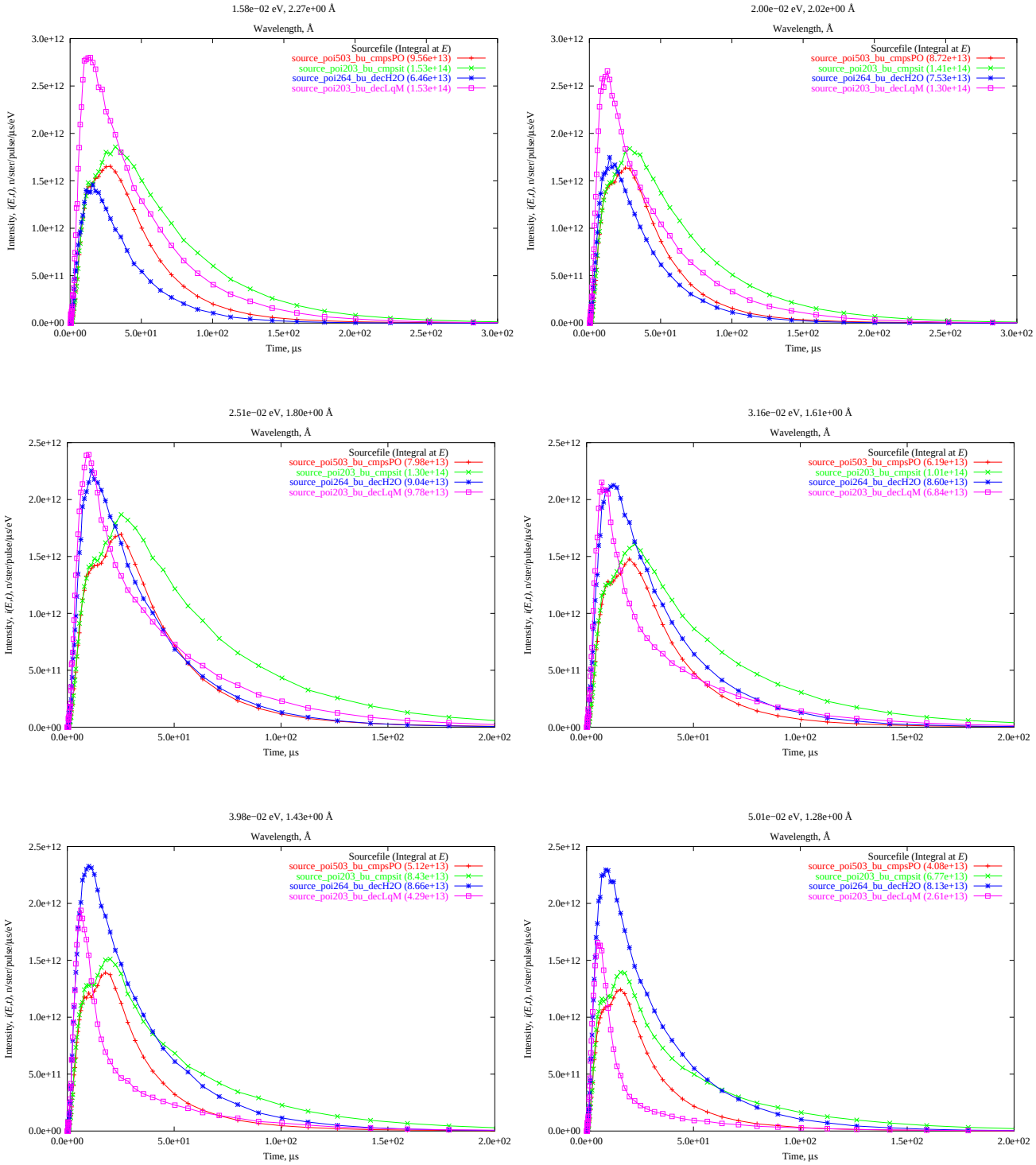

Figure 9: Emission time distributions. The poisoned composite moderator is denoted source_poi503_bu_cmpsPO, the unpoisoned composite source_poi203_bu_cmpsit, the poisoned water source_poi264_bu_decH2O, and the poisoned liquid methane source_poi203_bu_decLqM. $\Delta E/E \approx 23$ %.

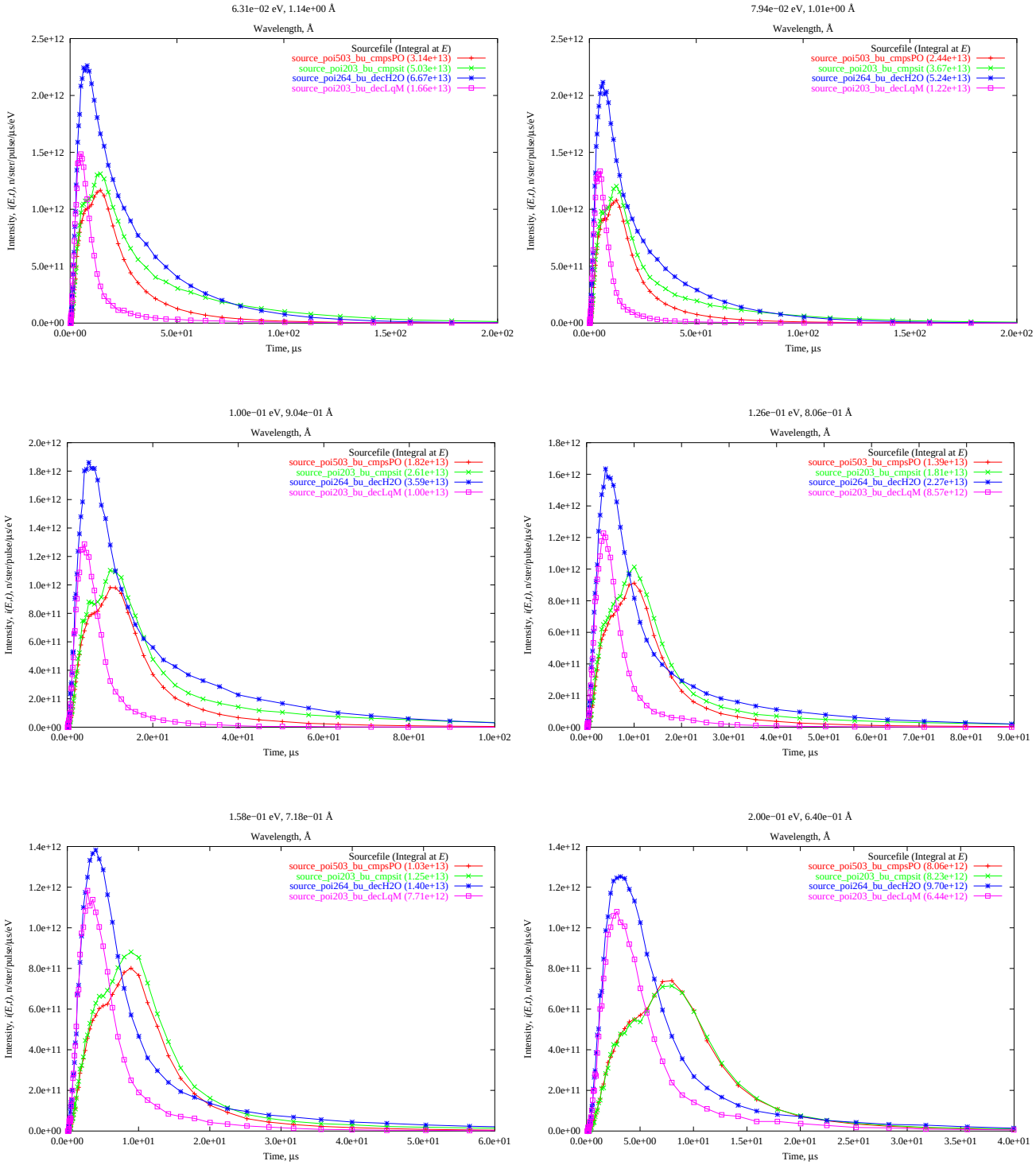

Figure 10: Emission time distributions. The poisoned composite moderator is denoted source_poi503_bu_cmpsPO, the unpoisoned composite source_poi203_bu_cmpsit, the poisoned water source_poi264_bu_decH2O, and the poisoned liquid methane source_poi203_bu_decLqM. $\Delta E/E \approx 23$ %.

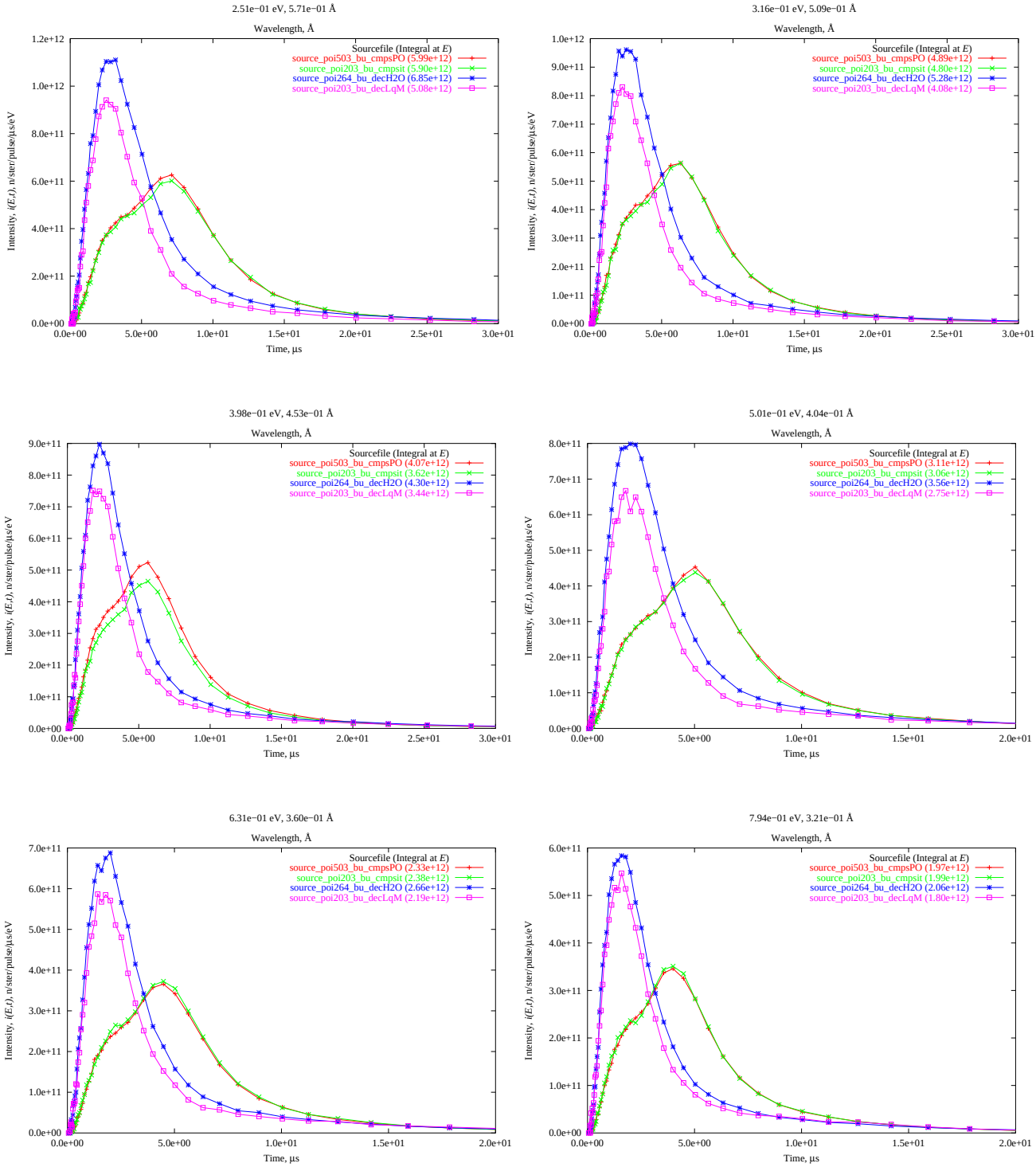

Figure 11: Emission time distributions. The poisoned composite moderator is denoted source_poi503_bu_cmpsPO, the unpoisoned composite source_poi203_bu_cmpsit, the poisoned water source_poi264_bu_decH2O, and the poisoned liquid methane source_poi203_bu_decLqM. $\Delta E/E \approx 23$ %.

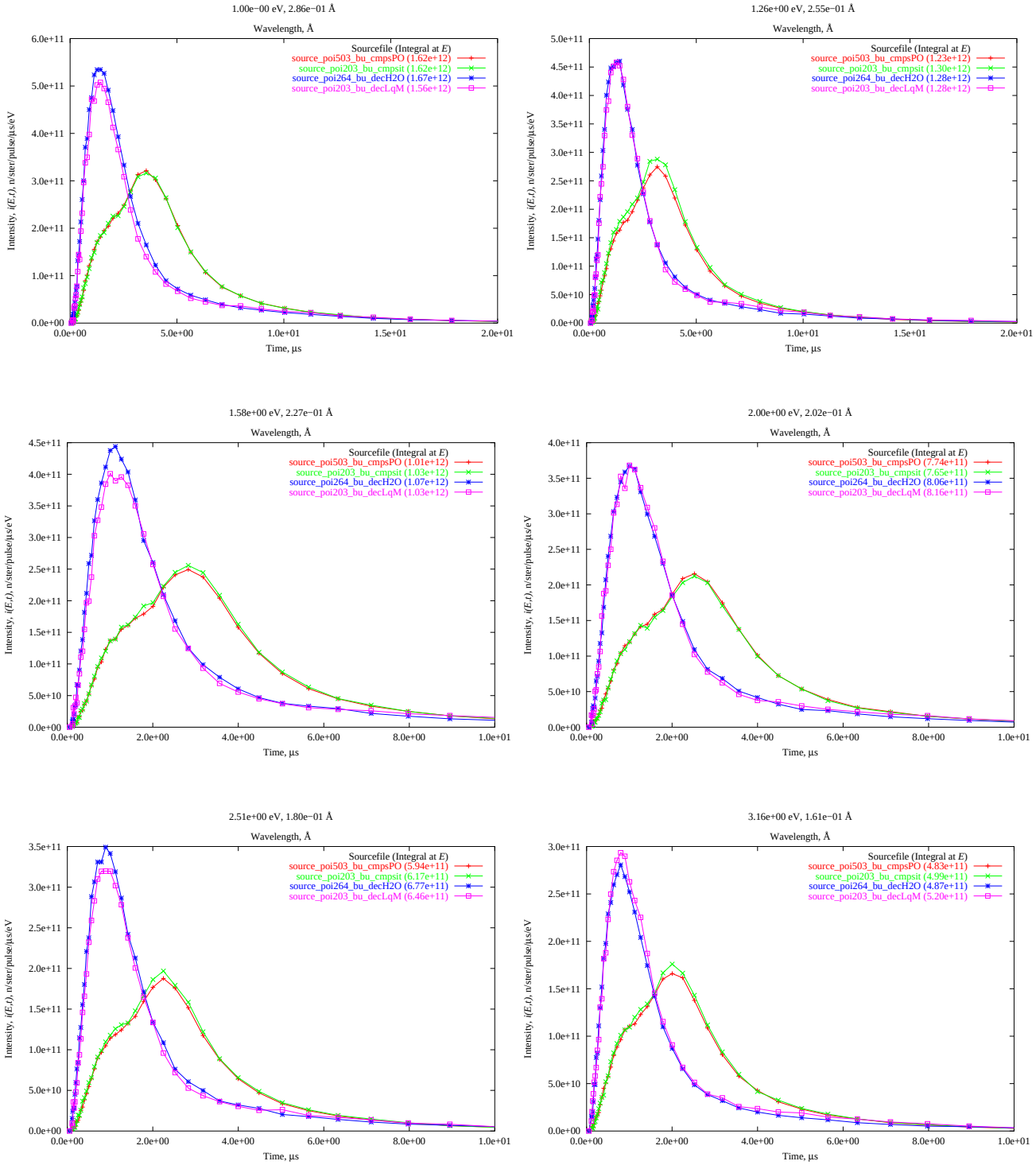

Figure 12: Emission time distributions. The poisoned composite moderator is denoted source_poi503_bu_cmpsPO, the unpoisoned composite source_poi203_bu_cmpsit, the poisoned water source_poi264_bu_decH2O, and the poisoned liquid methane source_poi203_bu_decLqM. $\Delta E/E \approx 23$ %.

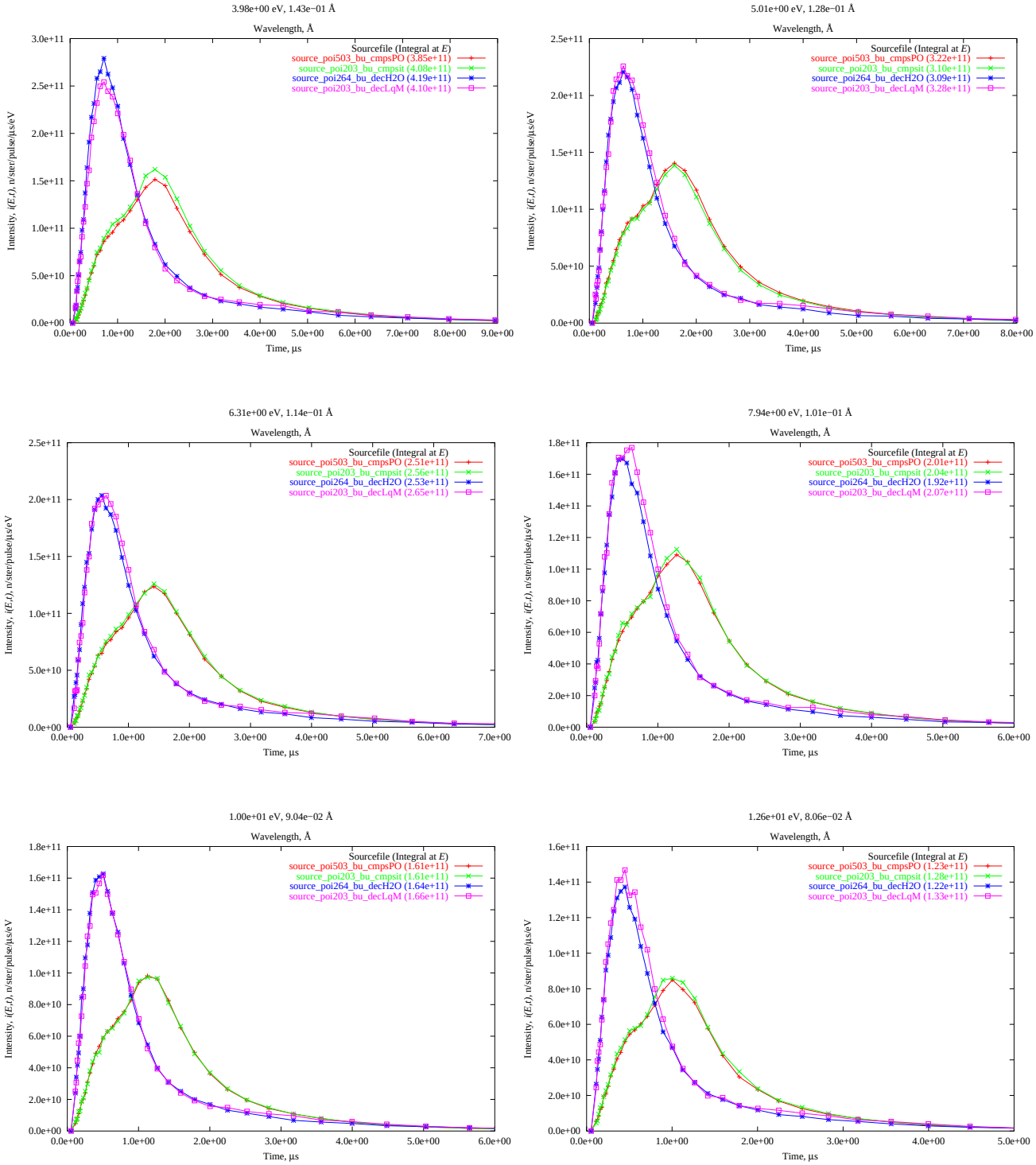

Figure 13: Emission time distributions. The poisoned composite moderator is denoted source_poi503_bu_cmpsPO, the unpoisoned composite source_poi203_bu_cmpsit, the poisoned water source_poi264_bu_decH2O, and the poisoned liquid methane source_poi203_bu_decLqM. $\Delta E/E \approx 23$ %.

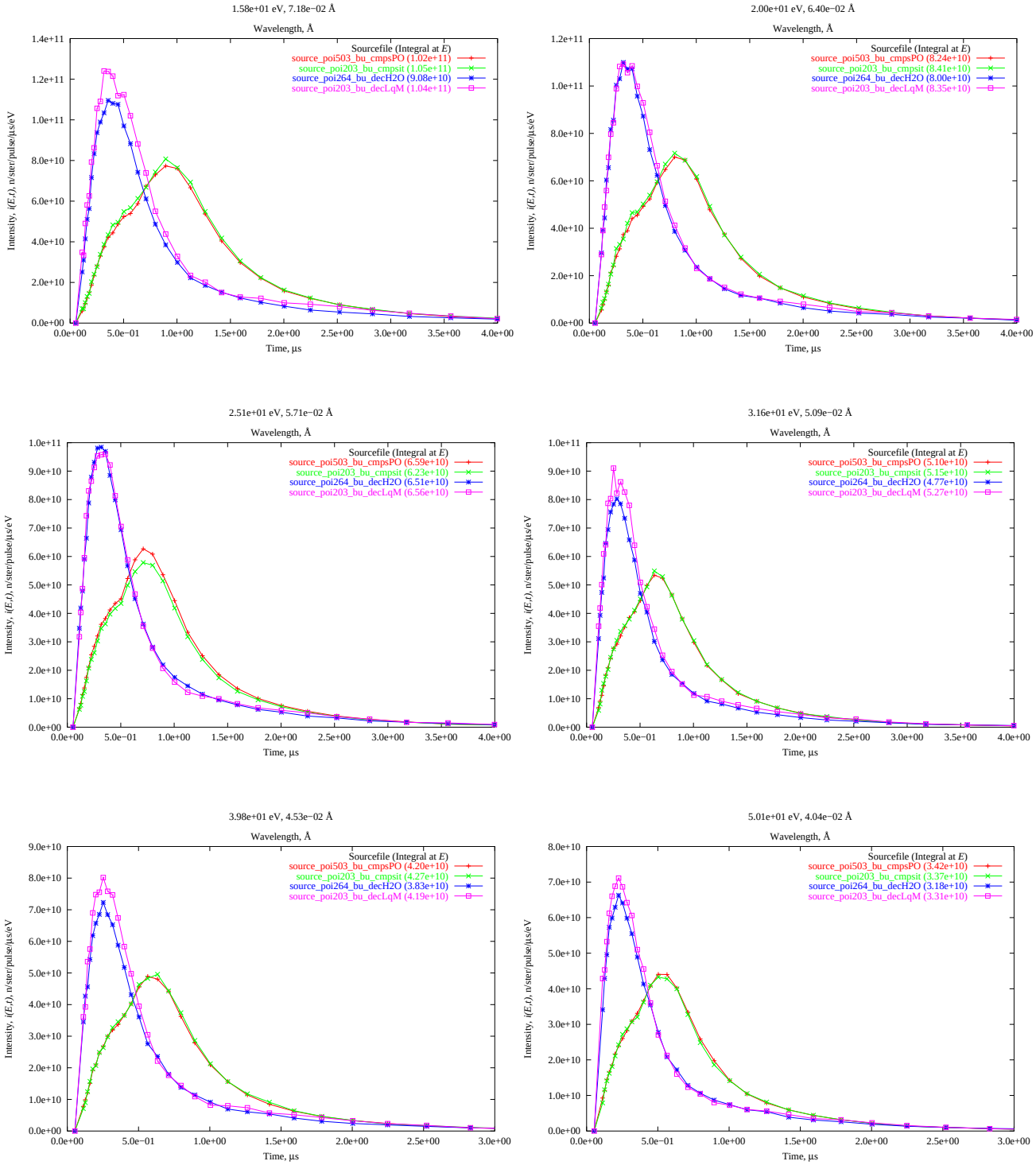

Figure 14: Emission time distributions. The poisoned composite moderator is denoted source_poi503_bu_cmpsPO, the unpoisoned composite source_poi203_bu_cmpsit, the poisoned water source_poi264_bu_decH2O, and the poisoned liquid methane source_poi203_bu_decLqM. $\Delta E/E \approx 23$ %.