# IPNDV Working Group 3: Technical Challenges and Solutions
# Nuclear Material (1)—Technology Data Sheet

*September 14, 2016*

**Nuclear Material (NM) Technology Name:** Calorimetry

**Physical Principle/Methodology of Technology:**

Calorimetry measures the thermal power output of heat-producing NM. The heat results from the radioactive decay of isotopes by alpha particle emission (for most Pu isotopes and $^{241}$Am) and by beta decay (for $^{241}$Pu and Tritium). $^{239}$Pu for instance decays to $^{235}$U with the emission of an alpha particle and releasing 5.15 MeV in energy. The energy loss through the emission of spontaneous fission neutrons on the other hand is many orders of magnitude smaller than the total disintegration of energy and loss due to gamma ray emission, representing only a small percent of the total disintegration energy of Pu isotopes.

Calorimetry is most commonly used for Pu measurements due to the high heat output of most Pu isotopes. In addition, the build-up of $^{241}$Am as Pu ages significantly increases the power output. It is also possible to measure highly enriched uranium (HEU) when there is a high enough $^{234}$U content. For reference, isotopes that emit relatively large amounts of heat include $^{238}$Pu (560 W/kg), $^{239}$Pu (1.9 W/kg), $^{240}$Pu (6.8 W/kg), $^{241}$Pu (4.2 W/kg), $^{241}$Am (114 W/kg), and $^{234}$U (0.2 W/kg). Thermal powers ranging from 0.1 mW to 1000 W can be measured with calorimetry.

Used independently, calorimetry confirms that an item emits heat and determines how much heat is being emitted. The heat measurement is very accurate and precise. In typical nuclear material measurements, the calorimetry heat measurement is combined with the isotopic composition obtained from gamma spectroscopy to determine the mass of nuclear material. Calorimetry is unbiased and unaffected by geometry and material matrix effects and does not require any representative standards. The accuracy in mass mainly depends on how accurately the isotopic composition is known. To be able to determine mass, as required by the IPNDV simple scenario, calorimetry will however rely on knowledge of the isotopic composition.

Within given boundary conditions to be ensured by strong information barriers, the verification of attributes in nuclear disarmament for a Pu based device would ultimately seek to:

(1) confirm the presence of Pu,
(2) measure the $^{240}$Pu to $^{239}$Pu ratio (typically ≤ 0.1 for NEDs)
(3) measure the mass of $^{240}$Pu and
(4) consequently extract the mass of Pu from (2) and (3) I order to verify whether the mass exceeds an agreed threshold.

When the isotopic composition of the NM is determined (by either gamma spectrometry or mass spectrometry) and knowing the specific heat power (W/Kg) of each isotope making the item, the total mass of Pu is extracted, simply equal to the total power measured by the calorimeter divided by the effective specific power of the item. Because uranium isotopes emit many orders of magnitude smaller amounts of heat per gram of material ** compared to $^{241}$Am and Pu, calorimetry is mostly suitable for measuring Pu mass. However, HEU calorimetry is feasible for matrices with high uranium content such as metals, U-alloys, oxides and high-grade scrap.

**Potential Monitoring Use Cases** (pre-dismantlement, dismantlement, post-dismantlement, storage stage):

Some examples of how calorimetry could be used to facilitate monitoring include:

- *Warhead Confirmation Attribute Based on Pu Mass*: calorimetry could be used together with gamma-ray spectroscopy to measure the mass of Pu and determine if the Pu mass is greater than a threshold.
- *Warhead Confirmation Attribute Based on Heat Output*: calorimetry could be used to measure the heat output of an item and determine if the heat output is greater than a threshold. Note this is a less rigorous warhead attribute than one based on Pu mass.
- *Confirm Absence of Special Nuclear Material (SNM)*: calorimetry can effectively be used to confirm the lack of SNM by the lack of a thermal signature in the dismantled components that are declared to contain no SNM.
- *Heat-Based Template*: calorimetry can be used to confirm that two different items output the same amount of heat, or that the SNM outputs the same amount of heat before and after dismantlement.

Item configuration has a large effect on the size of the calorimeter and the measurement time. As a result, calorimetry may be plausible for some applications in the regime but not others. In most cases, calorimetry measurements on warheads are more difficult and more time intensive then calorimetry measurements on components due to their larger size. Therefore, warhead confirmation measurements might be better applied to the dismantled components rather than on the warhead.

**Used to measure U, Pu, or U and Pu:**

Calorimetry can be used to quantify Pu with a range of isotopic composition or HEU with adequate $^{234}U$ content.

**For detection technologies, what does the method determine/measure** (e.g., presence of nuclear material, isotopics, mass, etc.)?

Calorimetry confirms that an item is emitting heat and measures the rate of heat output.

The heat measurement can be used together with isotopic composition information to determine the mass of Pu, HEU and/or other heat-emitting nuclear materials. In an arms control application it may be most plausible to obtain isotopic composition from gamma spectroscopy.

**Physical Description of Technology** (e.g., approximate size, weight):

A calorimeter consists of a thermally insulated enclosure with associated electronics. A computer is usually attached to the calorimeter to read raw data and calculate the item power. Once an item is placed inside the thermal enclosure the calorimeter begins to heat up due to the heat emitted from the item. In conventional applications the temperature of the calorimeter is allowed to reach equilibrium, or steady-state. In a water-bath or air-bath calorimeter, the temperature increase in the water/air bath is measured. In a solid-state calorimeter, the heat-flow through the solid-state sensors can be measured directly. The relationship between the measured value and the item power is linear in well-designed calorimeters and determined by 2–3 calibration measurements of heat standards.

A calorimetry system can thus be built with a cavity size able to enclose the item to be measured and can thus vary from a small sample colorimeter to one that can measure 250L drums, including the following examples:

Relatively small calorimeters (sample volume approximately 3 cm diameter by 8 cm height) can typically be fully contained on a 1 m x 0.70 m x 1.2 m transportable trolley including electronics, thermal elements, and PC and weigh about 250 kg. A mid-sized calorimeter with a 25 L sample volume may be 1 cubic m, and weight 600 kg. Large calorimeters that are able to measure, for example, 55 gallon (250 L) tritiated waste drums, may measure about 3 m in height, by 2.5 m in depth and 4 m in width and may weigh 1,000 kg.

In general, small calorimeters have a more limited power range but offer better precision and faster measurement times. Larger calorimeters support physically larger samples with higher power outputs, but offer lower precision and require longer measurement times.

For a 25 L sample cell volume, a typical colorimeter may be 1 $m^3$, weigh 600 kg, and be able to measure a Pu sample in the range of 5 to 30,000 mW (1 g Pu emits about 3 mW of heat), in about 3 to 4 hours measurement time with an accuracy and precision of about 0.5 percent.

**Time Constraints** (e.g., measurement times including distance from object, time to install the equipment):

The measurement time depends highly on the amount of thermal shielding (as part of either the item matrix or item container), the size of the calorimeter, and the severity of temperature fluctuations in the room during the measurement. Small calorimeters (as described in the discussion above) with minimal thermal shielding in the item container may have measurement times of 1–2 hours, whereas mid-size calorimeters may have 2–5 hour measurement times. On the opposite extreme, large calorimeters with heavily shielded items may have measurement times that are several hours or possibly 1–2 days.

It may be possible to decrease the measurement time by as much as 50 percent using predictive algorithms. A large part of the measurement time is spent within 10 to 15 percent of the final equilibrium and, as high accuracy and precision are not sought here, predictive algorithms can be used to significantly decrease measurement times. In other words calorimetry can be developed to run in a fast mode.

Measurement time can also be decreased by keeping the calorimeter pre-heated between item measurements, and/or maintaining stricter controls on the environmental temperature, but these approaches add to the complexity of the use procedures.

**Measurement time to measure 500 g of Pu (0.1 $^{239}$Pu/$^{240}$Pu) or 500 g of $^{235}$U at 1 m from the surface of the container (order of magnitude: seconds, minutes, hours, days):**

The item is completely enclosed within the calorimeter. It may not be necessary for the edges of the item to touch the calorimeter. However, the air gap must be kept as minimal as possible to ensure a good calorimetry measurement. Therefore the standoff distance of 1 m does not apply to a calorimetry measurement.

The power output of 500 g of Pu (10 percent $^{239}$Pu and 90 percent $^{240}$Pu) is approximately 3.4 W, which is measurable by most calorimeters. The volume of the Pu item depends on the geometry and that affects the size of the calorimeter required. A solid sphere of 500 g of Pu has a radius of about 1.8 cm, which would fit in a small to mid-size calorimeter and could be measured in 2–4 hours. On the opposite extreme, the same amount of Pu in a container would require a larger calorimeter and may require

several hours to measure. In both cases, thermal shielding such as plastic or foam in the sample container would increase the measurement time.

**Will this method work in the presence of shielding? If so, what is the maximum amount of shielding that will still allow the method to work?**

Thermal shielding using plastics or foams in the sample container would increase the measurement time. In a well-designed calorimeter with no heat leaks, thermal shielding does not affect the accuracy of the heat measurement. However, thermal shielding can significantly increase the measurement time because it slows the flow of heat through the calorimeter.

**Technology Complexity** (e.g., hardware, software, and ease of use by personnel):

*Hardware*: The underlying physical concepts of calorimetry are simple. Calorimeters use simple materials, but may be difficult to inspect because the system's materials are usually large, interlocking pieces to avoid heat leaks.

*Software*: Software records heat output as a function of time and calculates the item power. The analysis based on calorimeter equilibrium involves simple math. The complexity of predictive algorithms varies greatly, but relatively straightforward prediction algorithms exist.

*Ease of Use*: Calorimeter operation and use is conceptually straightforward. However, deployment concerns are discussed below.

**Infrastructure Requirements** (e.g., electrical, liquid nitrogen, etc.):

- Voltage power supply
- Heat standards (electrical or radioactive) for calibration purposes
- Lifting equipment for large items/calorimeters
- Assuming a water bath design, the facility may need to accommodate on the order of 4 m^3 of water

**Technology Limitations/Variations** (e.g., detection limits for nuclear material, operational temperature range, differences in technology detector materials):

*Technology Limitations*:

- Correct identification of the heat source requires separate measurement methods
- Batteries or heat-emitting isotopes with no gamma emissions would make the Pu mass appear larger than it is, and would not be detected in gamma spectroscopy
- Large operational temperature range, but measurement time is affected by temperature variations in the room

*Technology Variations*:

- *Passive-Mode Versus Servo-Control Mode*: In passive mode the calorimeter heats to an equilibrium temperature. In servo-control mode, the calorimeter is maintained at an equilibrium temperature with a heating blanket or other distributed heat source. Servo-control mode is often significantly faster, but requires an additional heat source.

- *With or Without a Predictive Algorithm*: The use of a predictive algorithm decreases measurement time but increases the complexity of the data analysis.
- *Air or Water Bath Calorimeter Versus Solid State Calorimeters*: Water bath calorimeters are typically very heavy. Air bath calorimeters may be less precise. Solid state calorimeters may require additional electronics.

The size and design of a calorimeter depends on the size and heat output of the items to be measured and the accuracy sought. Neither highest accuracy nor highest precision are sought for most treaty monitoring applications. The primary determining factor for the size and weight of the calorimeter is the size of the items to be measured.

**Information Collected by the Technology** (used to help determine if an information barrier is required for use):

Information collected by calorimetry on its own includes the heat output as a function of time (raw data). With significant research and sophisticated analysis, it may be possible to extract information regarding the item geometry or materials from the exponential components in the heat-up rate. An information barrier may be desired to protect the details of the heat-up rate. Qualitative information, such as visual observation of the heat-up rate, should be fairly innocuous. The measurement result is total heat output of the item that may be a non-sensitive quantity especially if no isotopic information is available.

To obtain Pu mass it is necessary to combine the total heat output with the isotopic composition. The isotopic composition may be sensitive and warrant an information barrier. It may be possible, for example, to combine a gamma-spectroscopy system with a calorimetry measurement behind an information barrier to output a mass attribute result.

**Safety, Security, Deployment Concerns**:

*Safety*:

- There may be safety concerns with enclosing warheads or high explosives inside the calorimeter, during which time it would be difficult/impossible to visually supervise or monitor the warhead.
- The item will heat while it is inside of the calorimeter. The extent of heat-up depends on the design of the calorimeter. Heating certain items or components may pose safety concerns.
- Calorimeters that use a water bath or plastics may moderate the item's neutrons, which may pose safety concerns for certain items or components.

*Security*: Security concerns with collected information are discussed above.

*Deployment*:

- For measurements of large items, special equipment may be required to lift and place the item in the calorimeter
- Calorimeters often need to be transported as a single large piece of equipment due to their design to avoid heat leaks
- Strong desire for stable environmental conditions
- Potential procedures to pre-heat the calorimeter to decrease measurement time

- Need for 2–3 trusted calibration heat standards; heat standards can be electrical or radioactive, each with a different set of deployment considerations

**Technology Development Stage** (Technology Readiness Level, TRL):

TLR 7+; Calorimeters are commercially available and can be ordered to user requirements. However:

Although small- to mid-sized calorimeters are common and well-researched, large calorimeters, such as those used for warheads or pit containers, are less well established. Research may be needed to determine if it is possible to obtain the desired accuracy within an appropriate measurement time for such large items.

Calorimeters designed to facilitate portability and inspections by each treaty partner do not exist. Research would be needed to develop a design approach that is portable, and inspectable while maintaining functional robustness (i.e., minimizes heat leaks).

**Additional System Functionality** (e.g., outside the monitoring use case):

Calorimetry is used in a wide range of applications as described below. Due to its accuracy and precision, calorimetry can be used as an alternative to costly destructive analysis.

**Where/How the Technology Is Currently Used** (e.g., international safeguards, border protection):

Calorimetry is currently used in safeguards for waste characterization and for the quantification of alpha emitters (Pu, $^{241}$Am) and beta emitters such as tritium (and $^{241}$Pu). Due to the high accuracy and precision offered by calorimetry, it is often used as a standards measurement to determine uncertainties and biases in other NDA techniques such as neutron counting. Calorimetry has been routinely used since the 1970s in U.S. and European laboratories for Pu process measurements and for nuclear material accountancy.

**Examples of Equipment:**

http://www.kep-nuclear.com/calorimetry/

http://www.antech-inc.com/product-categories/calorimetry-waste-assay/

Plus see references below.

**References:**

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