

SECTION A. Project Title: Application of the Associated Particle Technique to Fieldable Prompt Gamma-Ray Neutron Activation Analysis Systems

SECTION B. Project Description and Purpose:
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Prompt Gamma-ray Neutron Activation Analysis (PGNAA) is an extremely useful tool for identifying the presence of various chemical or isotopic compounds concealed within thick-walled containers, made possible by the highly penetrating nature of the fast neutrons used to induce activity. Deuterium-Tritium (DT) neutron generators are particularly useful for this because of the energetic (14 MeV) neutrons they produce, which, in addition to being exceptionally penetrating, are also able to induce gamma-ray emission in elements requiring a high threshold for excitation, namely light elements such as carbon and oxygen. However, these same elements are also prevalent in the environment surrounding the field sample, creating a source of high background and, thus, low signal-to-noise ratios making the abundance fractions of these elements within the sample difficult to determine with sufficient precision to identify the compound inside, e.g., when looking for the presence of high explosives. One way this precision could be improved is by reducing the background signal from the environment through the Associated Particle (AP) technique, which requires a specialized DT generator having a built-in charged-particle detector that is used to identify the exact point in time and direction of emission that a neutron is produced by the generator. This technique has traditionally been applied to neutron imaging applications, but there is ample reason to believe it could also be used to improve fieldable PGNAA systems. This study would investigate the possible gains in precision that could be achieved by applying the AP technique to PGNAA assays, with particular focus on light elemental abundances and using a variety of different external gamma-ray detectors, namely High-Purity Germanium (HPGe), sodium iodide (NaI), and lanthanum bromide (LaBr₃) with each offering different tradeoffs in detection efficiency, energy resolution, and timing resolution.

PGNAA is a powerful tool for detecting the presence of specific chemical elements that may be contained within a dense material such as steel. It is sensitive to most elements and allows one to determine relative ratios of elements, or even isotopes in some instances, without requiring any physical alteration or manipulation of the container. The basic principle behind the method is the use of energetic neutrons, which are highly penetrating through matter, to irradiate a sample of unknown composition. The neutrons interact with the material inside through various nuclear scatter and capture reactions, many of which release energy as gamma-radiation. The emitted gamma radiation is characteristic of the interacting material and is then detected outside the sample container by an external spectrometer that is capable of identifying the chemical element and nuclear reaction responsible for it. This method has been employed, for example, at Idaho National Laboratory (INL) for nearly 30 years through the various installments of the Portable Isotopic Neutron Spectroscopy (PINS) systems, which has been utilized by the Army's Recovered Chemical Materiel Directorate (RCMD) to identify suspect chemical warfare material in recovered munitions.

Although fieldable PGNAA systems such as PINS are effective for detecting a large variety of key elements, even sensitive to variations in isotopic abundances, the method is sometimes limited by low neutron reaction probabilities, low gamma-ray detection efficiencies, and high background signals. These limitations tend to be most problematic for light elements such as carbon, nitrogen, and oxygen. Their low neutron-interaction probabilities mean that they produce less gamma-rays compared to other elements, and the gamma-rays they do produce tend to be highly energetic which results in a low likelihood of depositing their full energy within the detector outside the sample, making it difficult to identify which element produced them. Furthermore, these elements are ubiquitous in the surrounding pavement, soil, air, and moisture existing in the field environment. Thus a large background of these characteristic gamma-rays are emitted from sources outside the test samples making it difficult to ascertain what fraction, if any, arise from the sample in question.

One way to combat, at least, the low neutron reaction probabilities for some of these elements is to increase the energy of the irradiating neutrons. Whereas many fieldable PGNAA systems have utilized sealed sources of Californium-252, which produce a range of neutron energies up to 10 MeV or more, however the majority of emitted neutrons have only a few MeV or less. This is less than ideal, for example, when trying to detect the presence of carbon and oxygen which require neutron energies greater than 4 and 6 MeV, respectively, to induce emission of their characteristic gamma-rays. In order to improve sensitivity to these elements, DT neutron generators have been employed which produce very energetic neutrons of 14 MeV. Although these higher-energy neutrons are much better suited to excite the light elements such as carbon and oxygen in the test item, they unfortunately have an equally high chance for doing the same to the light elements in the surrounding environment, resulting in little to no improvement in the signal to background ratios of their detected characteristic gamma rays. This is particularly problematic for test samples of relatively small size where many more neutrons will tend to miss the sample and interact mainly in the surroundings. Thus, to see a real improvement in the light element detectability, one has to somehow distinguish between gamma-rays arising from the test item and the environment.

One potential way of addressing this problem is through the Associated Particle (AP) method. This method uses a specialized DT neutron generator which has a charged-particle detector built into its inner target where the neutrons are produced. In the case of a DT generator, a triton (H-3) is accelerated inside the vacuum tube within the generator and hits a deuterated (H-2) target. The triton undergoes a fusion reaction with a deuteron that produces an energetic neutron and alpha particle (helium-4 nucleus) emitted in opposite directions from each other. In the AP neutron generator (APNG) some of the alpha particles will hit the internal charged-particle detector, thus signaling to the user the precise moment that a neutron was emitted in the direction opposite the detector. This detection signal can then be used as a gate signal for an external gamma-ray detector to discriminate between gamma-rays that were detected in coincidence with a neutron that was emitted towards the test item and other gamma-rays produced from neutrons emitted into the surrounding materials such as the ground. In principle, this method should significantly improve the signal-to-background ratios of the light element characteristic gamma-rays, and generally all elements excitable by fast neutrons. Furthermore, since the velocities of the DT-produced neutrons and subsequently induced gamma-rays are known, one could even employ fast detectors such as LaBr₃ to impose strict time constraints on the coincident gamma-rays to potentially isolate certain depth ranges within the test item and/or suppress events where the neutron was scattered out of the test item and into the surrounding environment inducing a gamma-ray there. Use of the AP method up to this point in time has been mainly utilized in radiography and imaging applications; INL personnel are not aware of any instance of its use in fieldable PGNAA instruments.

Research Plan: INL does not own a specialized, AP-type neutron generator. The cost to purchase one would be prohibitively expensive for a seed LDRD project (~\$100k). The neutron radiography research group from Oak Ridge National Lab (ORNL) agreed to let INL borrow one of their AP-type generators for

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a period of 6 months to test its feasibility for PGNAA. The APNG has already been shipped to and received by INL and is currently located in the PINS lab (IF-675) which contains the necessary infrastructure (i.e., shielding, interlocks, vaults, dose meters, radiological work permits, detectors, and electronics) to facilitate experiments utilizing neutron generators. The neutron generator for this project is a model API-120; the only difference from an environmental point of view is that the neutron output is much less than other models currently used at PINS, thus the expected rad doses would scale down accordingly. As the API-120 is owned by ORNL, it will have to be shipped back to them later this year. The PINS lab also houses a large array of sample test items relevant for PGNAA experiments, in particular various high explosive simulants, which consist primarily of different ratios of the light elements carbon, nitrogen, and oxygen contained within steel casings. This project would also take advantage of a previously purchased data acquisition system that is specialized for performing fast coincidence spectroscopy, capable of sub-nanosecond timing resolution. The latter might allow for the depth range determination of a penetrating AP-tagged fast neutron down to 1 cm precision, when paired with a sufficiently fast gamma-ray detector such as LaBr₃. Such a detector is already owned by the PINS group along with other gamma-ray detectors that might prove useful for the present study like NaI and HPGe.

The primary goal of this project would be to leverage all these existing resources at INL and take full advantage of this limited-time opportunity of the borrowed APNG from ORNL. Note that the latter component is the single most critical item for this research and will only be available through the midpoint of the present fiscal year. The main objectives of this research would be to a) determine if an advantage can be gained in the speed and precision of PGNAA field measurements utilizing the AP-method, in particular with regards to the detection of light elements such as carbon and oxygen, and b) quantify any measured improvement of the AP-PGNAA system relative to the standard PGNAA measurement in terms of signal to background ratios and uncertainties in measured elemental ratios, c) compare these limits using various gamma-ray detectors like HPGe, NaI, and LaBr₃, and d) investigate the assay depth sensitivity by placing as tight of time constraints as possible on the coincidence events using the fastest detectors available.

To carry out these research objectives, the APNG will be operated in the south cave of the PINS lab using the already existing radiation safety protocol/interlock system in place. The first set of experiments will utilize a HPGe detector for recording the induced gamma-ray spectra for various samples irradiated by the APNG, followed by measurements with NaI and LaBr₃ scintillation detectors. A fast photomultiplier tube (PMT) will be attached to the viewport of the built-in charged particle scintillation detection medium of the APNG provided by ORNL. The signal from the PMT will be used to generate a coincidence time gate between the AP-tagged DT neutrons and induced gamma-rays recorded in the external detector. A multichannel Pixie-4e digital data acquisition system will be used to process the detector signals and analyze the coincidence events. The gamma-ray detector signals will be split so that both gated and ungated spectra can be recorded simultaneously. The two spectra can then be analyzed for each test sample to determine the effect of the AP-method.

Data will be collected for various test samples, focusing most on those containing various proportions of light key elements such as carbon, nitrogen, and oxygen. A number of high explosive simulants will be studied as well as inert samples such as sand, graphite, and water. INL will also examine the effects on some chemical warfare simulants such as VX nerve agent and HD (sulfur mustard) blistering agent, as well as an assortment of smokes and riot control agents and possibly simulated biological agents. These simulants are maintained as part of the normal inventory at the PINS facility. Although time for collecting experimental data is limited to the duration of our possession of the APNG from ORNL (about mid FY21), significant time would still be required to analyze the data in the following months. It is anticipated that the results, whether indicating favorability of the AP-method for PGNAA or not, will be prepared for submission to a scientific journal such as Nuclear Instruments and Methods in Physics Research. It is expected that this project to be fully completed by the end of FY21.

The project does not generate any air emissions or waste. The simulants are used over and over again.

SECTION C. Environmental Aspects or Potential Sources of Impact:

Air Emissions

N/A

Discharging to Surface-, Storm-, or Ground Water

N/A

Disturbing Cultural or Biological Resources

N/A

Generating and Managing Waste

The only waste that is anticipated might be paper. If paper is generated then it will be recycled.

Releasing Contaminants

N/A

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Using, Reusing, and Conserving Natural Resources

Any waste that is generated should be diverted from the landfill to the extent possible.

SECTION D. Determine Recommended Level of Environmental Review, Identify Reference(s), and State Justification: Identify the applicable categorical exclusion from 10 Code of Federal Regulation (CFR) 1021, Appendix B, give the appropriate justification, and the approval date.

For Categorical Exclusions (CXs), the proposed action must not: (1) threaten a violation of applicable statutory, regulatory, or permit requirements for environmental, safety, and health, or similar requirements of Department of Energy (DOE) or Executive Orders; (2) require siting and construction or major expansion of waste storage, disposal, recovery, or treatment or facilities; (3) disturb hazardous substances, pollutants, contaminants, or Comprehensive Environmental Response, Compensation, and Liability Act (CERCLA)-excluded petroleum and natural gas products that pre-exist in the environment such that there would be uncontrolled or unpermitted releases; (4) have the potential to cause significant impacts on environmentally sensitive resources (see 10 CFR 1021). In addition, no extraordinary circumstances related to the proposal exist that would affect the significance of the action. In addition, the action is not "connected" to other action actions (40 CFR 1508.25(a)(1) and is not related to other actions with individually insignificant but cumulatively significant impacts (40 CFR 1608.27(b)(7)).

References: 10 CFR 1021 Appendix B to subpart D, Item B3.6, "Small-scale research and development, laboratory operations, and pilot projects", B1.24 "Property Transfers" and B1.31 "Installation and relocation of machinery and equipment".

Justification: The proposed R&D activities are consistent with CX B3.6 "Siting, construction, modification, operation, and decommissioning of facilities for small-scale research and development projects; conventional laboratory operations (such as preparation of chemical standards and sample analysis); small-scale pilot projects (generally less than 2 years) frequently conducted to verify a concept before demonstration actions, provided that construction or modification would be within or contiguous to a previously disturbed area (where active utilities and currently used roads are readily accessible). Not included in this category are demonstration actions, meaning actions that are undertaken at a scale to show whether a technology would be viable on a larger scale and suitable for commercial deployment.",

B1.24, "Transfer, lease, disposition, or acquisition of interests in personal property (including, but not limited to, equipment and materials) or real property (including, but not limited to, permanent structures and land), provided that under reasonably foreseeable uses (1) there would be no potential for release of substances at a level, or in a form, that could pose a threat to public health or the environment and (2) the covered actions would not have the potential to cause a significant change in impacts from before the transfer, lease, disposition, or acquisition of interests."

B1.31, "Installation or relocation and operation of machinery and equipment (including, but not limited to, laboratory equipment, electronic hardware, manufacturing machinery, maintenance equipment, and health and safety equipment), provided that uses of the installed or relocated items are consistent with the general missions of the receiving structure. Covered actions include modifications to an existing building, within or contiguous to a previously disturbed or developed area, that are necessary for equipment installation and relocation. Such modifications would not appreciably increase the footprint or height of the existing building or have the potential to cause significant changes to the type and magnitude of environmental impacts." and,

Is the project funded by the American Recovery and Reinvestment Act of 2009 (Recovery Act) Yes No

Approved by Jason Sturm, DOE-ID NEPA Compliance Officer on:02/04/2021