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**NON-DESTRUCTIVE EVALUATION TECHNIQUES FOR CHEMICAL
WEAPONS DESTRUCTION***

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1. Introduction

The safe and verifiable disposition, either by incineration or chemical neutralization of chemical warfare (CW) agents requires correct *a priori* identification of each munition or container to be processed. A variety of NDE techniques have been used or tested for the examination and characterization of munitions.[1,2] In the U.S., three widely used techniques are X-ray radiography, acoustic resonance spectroscopy (ARS), and prompt gamma ray neutron activation analysis (PINS). The technical bases, instrumental implementations, and applications of the U.S. versions of these methods are briefly discussed.

2. Non-Destructive Evaluation (NDE) Techniques

2.1 X-RAY RADIOGRAPHY

X-ray radiographs are routinely used to locate items in overpack containers, to image the internal structure of munitions to aid in their classification, to assess the presence, absence, and/or status of energetics such as the burster charge and fuze, and to differentiate between solid- and liquid-filled items. The U.S. Army Non-Stockpile Chemical Material program is responsible for the proper disposition of recovered munitions and items that may contain CW agents.[3] These munitions are often rusted, pitted, and have lost their markings. An x-ray image is usually the first piece of NDE data acquired on these munitions.

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X-ray radiographic systems can be classified according to the method used to obtain the x-ray image -- either real-time video systems or film-based systems. The relative advantages and disadvantages of these two approaches are compared in Table 1.

TABLE 1. A relative comparison of X-ray radiography techniques	
Film-based	Real-time video
<ul style="list-style-type: none"> • Cheaper to buy, but more expensive to use • Image optimization is "trial and error" • Gives the best resolution when properly optimized 	<ul style="list-style-type: none"> • More expensive to buy but cheaper to use • Image optimization is done in real time • Adequate resolution for all but the most demanding tasks

Either film-based or real-time video readout systems can be mated with a selection of x-ray generating tubes or "heads". The available x-ray heads differ primarily in the energy of x-ray radiation that they produce. In general, the higher energy heads are somewhat heavier, less portable, and have higher power demands than the lower energy devices, but have higher output and greater penetration ability.

Very portable units weighing less than 8 kg and capable of operating on a battery pack are available with x-ray energies of up to 150 keV (accelerating voltage up to 150 kV). Experience in the U.S. indicates that these units are capable of imaging some details of the burster well and liquid fill level in thin-walled containers such as small to medium-sized mortar projectiles and canisters, but they lack the penetrating power to image the internals of heavier-walled munitions such as 155 mm projectiles.

To penetrate the walls of these heavier artillery projectiles requires x-ray tubes with outputs of about 300 kV. Portable (but not battery powered) 300 kV units usually incorporate a separate x-ray generating head and a control unit. The combined weight of these pieces is nominally 40 to 50 kg. A 300 kV unit, coupled with either film or video is capable of adequately imaging the burster well and liquid level in, for example, 155 mm and 175 mm artillery projectiles. In addition, some of the details of the projectile fuzes can be determined. These units are generally incapable of properly imaging fine details of the fusing system in 155 mm projectiles (such as the position of certain of the fuze pins) and are limited in their ability to image larger munitions such as bombs.

To adequately image the fine details of the fusing system and to penetrate the walls of heavier munitions requires x-ray potentials of 420 kV and greater. These larger units, although highly capable, are more usually installed systems with total weights of 500 kg or more.

2.2 ACOUSTIC RESONANCE SPECTROSCOPY (ARS)

ARS [4] is a technique that can classify CW munitions by analyzing the effect that the fill has on the container's mechanical resonances. ARS accomplishes this classification by comparing a measured acoustic signature to a known standard or template. ARS does not measure physical characteristics of an object directly, but rather infers them by comparison to known items.

Any physical object, whether solid, hollow, or fluid filled, possesses many modes of vibration. These modes of vibration, commonly referred to as the natural resonant modes or resonant frequencies, are determined by the object's shape, size, and physical properties. In the case of munitions, such as an artillery projectile, the observed resonances are a function not only of the projectile case, but are also dependent on the physical characteristics of the munition fill.

The resonance spectrum, obtained by continuously exciting the object over a wide frequency range (sine-wave frequency sweep) and measuring its response, provides an acoustic signature of the object. The measurements can be made with direct contact transducers, such as piezoelectric crystals, or with a non-contact setup using a speaker for excitation and a laser vibrometer for response measurement. Typically, the frequency sweep range used for chemical munitions lies between 1 KHz and 30 KHz and the entire frequency sweep can be carried out in less than 60 seconds.

The acoustic resonance spectrum thus contains the information necessary to determine the fill material by a comparative process. The ARS instrument measures the acoustic spectrum of the object and extracts the acoustic signature. These frequencies are compared to a library of signature templates. Each template represents a different potential fill material, for example, the chemical or high explosive fills that are found in artillery shells.

A highly portable ARS system, the ARS300, has been developed by the U.S. Los Alamos National Laboratory (LANL).[4] The instrument, powered by removable, rechargeable batteries and weighing less than ten pounds, consists of a stand-alone electronics package coupled to a notepad computer and to a transducer assembly consisting of two magnetically coupled piezoelectric transducers. The electronics package produces a swept frequency sine wave signal and measures the resulting vibrations. Measurement acquisition time is approximately one minute. The acquired acoustic spectrum is transmitted to the computer, where decision algorithms perform the classification. The computer also provides a pen-driven user interface for the system. This system is pictured in Figure 1. A predecessor of this system, the ARS100, was used during United Nations Special Commission (UNSCOM) inspections of the Iraqi stockpile following the Persian Gulf War.

ARS measurements are rapid, generally requiring less than 60 seconds per measurement, making it a desirable technique for rapid screening of large numbers of munitions. ARS requires a database of spectra from some 20 to 30 representative munitions in order to establish comparison templates. Thus, ARS measurements are primarily useful for confirmation and classification assessments on stockpile munitions. Heavily rusted, pitted, or damaged munitions cannot be reliably classified.

Early ARS prototypes were field tested in two exercises at the U.S. Tooele Army Depot (TEAD) in 1991 and 1992.[1] The ARS system and other NDE systems were tested on a variety of CW munitions and bulk storage containers. ARS proved to be most useful on munitions rather than bulk containers, since it is easier to excite the

Figure 1. The LANL-developed ARS300

body resonances on the smaller items. In 1994, the ARS300 system was tested against surrogate 155 mm artillery projectiles.[2] The classification tests were blind, and the system demonstrated 90% reliability at the 90% confidence level.[4]

An ARS home page is available on the world wide web. This information is located at <http://pia.csa.lanl.gov/ars/>.

2.3 PROMPT GAMMA RAY NEUTRON ACTIVATION ANALYSIS

The U.S. implementation of a prompt gamma ray neutron activation instrument for NDE on munitions and containers is the Portable Isotopic Neutron Spectrometry (PINS) chemical monitoring system.[5] PINS was originally developed at the U.S. Idaho National Engineering Laboratory (INEL) for non-destructive monitoring of CW munitions in the U.S. and Russian stockpiles in support of chemical warfare treaty monitoring. After proof of concept tests were carried out with actual CW agents and high explosives in April 1991 at Tooele Army Depot, it quickly became apparent to the U.S. Army that PINS could also elucidate the content of *non-stockpile* munitions and containers that had lost their identifying marks or fill records. Often these munitions have been buried for years, and rust has obliterated their markings. In addition to the buried munitions, old "dud" munitions, i.e. bombs or projectiles that failed to explode on impact, are often recovered from firing ranges. Duds are also rusted, difficult to identify, and can be extremely dangerous; their fuzes may have armed and their casings may be fragile from corrosion and the shock of landing. For safety, these munitions are frequently overpacked in air-tight metal containers. The PINS system can assay these items through the outer container walls.

PINS employs neutrons from a radioisotopic californium-252 source to interrogate the contents of a munition. The neutron passes through the munition's steel casing, scatters, slows down, and is captured by the nucleus of one of the chemical elements within the munition. The capturing nucleus then emits one or more gamma-rays. Additional gamma rays can be produced by inelastic scattering reactions. The resultant

energetic gamma rays easily penetrate steel, and they escape the munition and are recorded by a gamma-ray detector.

Nearly all chemical elements have a unique and well-known neutron-induced gamma-ray spectral signature. Because of their intimate connection with basic and applied nuclear physics, detailed tables of nuclear energy levels and gamma-ray spectral catalogs and tables have been compiled. [see for example 6,7] From this library of data the gamma rays detected during a PINS assay can be correlated to the elemental composition of the munition fill.

Since PINS does not directly sense molecular structure, the fill of a munition or container is inferred from the elemental information provided by the neutron-induced gamma rays. The various nerve agents are rich in phosphorus; the blister agents mustard gas (agent HD) and lewisite (agent L) are rich in chlorine. Sulfur is present in mustard gas but absent in lewisite; arsenic is present in lewisite but absent in mustard gas; the elements arsenic and sulfur distinguish these agents. High explosives are rich in nitrogen, but contain no arsenic, phosphorus, sulfur or chlorine. Hence, given the elemental constituents, a simple decision tree algorithm can classify a munition as explosive or CW, and if the latter, identify the CW agent family.

Prompt gamma ray neutron activation methods such as PINS exhibit varying sensitivities for the detection of specific chemical elements, an important limitation. The detection sensitivity of the PINS system can be qualitatively expressed in terms of the normal assay times required to detect the elements key to fill classification in various munitions. The PINS technique is, for example, quite sensitive to chlorine and hydrogen, far less sensitive to nitrogen (the key element of high explosive), and has intermediate sensitivity for sulfur, arsenic, and phosphorus. Certain chemical elements, such as oxygen and carbon are not detected. (This renders PINS incapable of discriminating between the various formulations of high explosive.) In munitions of moderate size, such as 4.2 inch mortar and 155 mm artillery projectiles, a mustard agent (HD) fill can be reliably classified in 500 to 1000 seconds of assay time. Identification of a GB or high explosive fill in these munitions generally requires 2000 seconds. [5] As could be expected, PINS assay times must be increased to properly classify the fill of small munitions such as 75 mm projectiles.

The PINS equipment consists of a californium-252 neutron source, a high-purity germanium (HPGe) gamma-ray detector, a portable multichannel analyzer (MCA), a notebook computer, a detector stand and shielding, and certain support equipment. The equipment is contained for shipment and storage in six equipment boxes.

The ^{252}Cf source is nominally a 5 microgram source emitting about 10^7 neutrons per second. The source is doubly encapsulated, and is certified as "special form" in accordance with the regulations of the International Atomic Energy Agency, permitting its routine shipment world-wide by common air carrier. When not in use the source is stored in a shielded 18-liter "bucket" that also serves as the shipping container. A PINS operator, handling the source by its attached lanyard, generally receives *no* measurable radiation dose during a full week of field assays.

During operation, the HPGe detector must be cooled to liquid nitrogen (LN_2) temperature. A 40-liter "mother" dewar sized to store enough LN_2 for weeks of

operation is provided as part of the support equipment. The 1.2-liter LN₂ dewar that is an integral part of the HPGe detector holds enough LN₂ for about 18 hours of operation. After a measurement day is completed the detector is attached to the mother dewar and refilled for the next day's operation. Liquid nitrogen is also routinely shipped by common air carrier.

The PINS electronics and multichannel analyzer (MCA) are contained in a briefcase-sized aluminum case. The MCA unit provides power for the radiation detector, and can operate for eight hours on its internal battery. The MCA and detector are connected by a 25 meter cable set. The long cable permits the operator to be far away from the detector and radioactive source, to further minimize the operator's exposure both to radiation and to potential chemical hazards.

A notebook personal computer (PC) provides a flexible, user-friendly control panel for the PINS system and implements data analysis software. The data analysis software examines the neutron-induced gamma-ray spectrum peak by peak, performs an energy calibration, looks up the detected gamma-ray peaks in a custom data base, and identifies the key chemical elements present inside the munition or container. Next, an identification algorithm using decision tree logic infers the chemical compound or mixture within the item under test, and finally, the assay results are presented to the operator. Analysis of a spectrum requires about 30 seconds. A "blind" PINS version that performs these functions while not permitting the operator to view the acquired spectrum is presently under development for use in situations where there is a concern about information security. The PINS equipment is pictured in Figure 2.

Figure 2. A PINS system assaying a laboratory surrogate.

PINS has been extensively tested against known and surrogate munitions.[1,2] In blind testing at the U.S. Army's Dugway Proving Ground (DPG), civilian PINS operators from DPG were presented with 22 surrogate-filled 155 mm projectiles and 22 surrogate-filled canisters. (Safety requirement obviously precluded using actual agents in unlabeled containers.) PINS correctly identified all of the blind test items.[8]

As a part of the U.S. Army's Non-Stockpile Chemical Material Program, x-ray radiography and PINS assays have been widely employed as a part of the munitions assessment effort.[3] The x-rays are used to determine if the munition is explosively configured, and provides information on fill material phase and fill level. The item is then assayed by PINS to elucidate the suspected fill. These data are then available to a Munitions Assessment Review Board that makes a recommendation on the final disposition of the item. Munitions that are deemed safe to access and are suspected of containing a chemical fill are drilled, drained and the fill materials analyzed and processed in a facility at the U. S. Army's Aberdeen Proving Ground that is properly equipped for these operations.[9] Munitions that contain no CWM are disposed of by other approved means.

The post NDE analyses of munition fill from the disposal activities provide valuable data for the assessment of the PINS system accuracy. Of 34 munitions verified to contain some type of CWM, the PINS classification has been correct on 31 (91%). There have been no false negative PINS results. The three classification errors were VX-filled 175 mm projectiles identified as GB-filled.[10]

PINS has been less successful in the identification of HE (and other nitrogen-containing compounds such as dinitrobenzene) in small items. For example, 18 of 19 75 mm projectiles that were recovered from the King's Creek area of APG were found during sampling to contain either TNT or nitrobenzene.[9] The PINS assay on these munitions had failed to detect the nitrogen in these munitions. In response to these results, INEL researchers have developed and are testing a PINS configuration that will improve the assay sensitivity for nitrogen in these small munitions.

3.0 Summary

A convenient suite of NDE techniques is available for the characterization of munition fill. Large numbers of similar stockpile munitions can be reliably and quickly classified using Acoustic Resonance Spectroscopy (ARS). Measurement times are about one minute per item. However, ARS measurements require a comparison template and are less useful for assay of old, rusted, and one of a kind munitions. For munitions of this type, x-ray radiography and assay by prompt gamma ray neutron activation analysis can reliably determine the munition fill, and ensure that the items are properly scheduled for destruction.

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