Nuclear forensics via the electronic properties of particulate and samples

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Abstract:

Recent research has shown how solid state dosimetry techniques can be used as a powerful tool in nuclear treaty verification. Using thermoluminescence, it has been shown that common bricks can serve as gamma ray spectrometers with 10% energy resolution for ^{241}Am [1]. Using optically stimulated luminescence (OSL), it has been shown that ^{137}Cs and ^{60}Co can be identified and imaged again using common bricks [2]. Likewise, surface mount resistors (such as in portable memory flash drives, credit cards or other electronics) are capable of recording dose approaching common background levels using these techniques [3]. Even imaging of weapons grade plutonium has now been accomplished using OSL; the extent to which this can be accomplished using the mineral particulate from smears and air filters is yet to be explored. Now that the theory has been worked out to use this science to carry out retrospective assay of uranium enrichment, the various applications for treaty verification are almost unbounded including retrospective assay of historical uranium enrichment [4]. Applications using electron paramagnetic resonance (EPR) have shown great promise [5] but new options will be explored here. This technology effectively puts low resolution imaging gamma ray spectrometers in every inhabited location on the planet throughout all time as insulator samples are ubiquitous in building materials, personal items and electrical circuit peripherals.

Keywords: Solid state dosimetry; Nuclear Forensics; Nuclear Safeguards; Luminescence; Magnetic resonance

1. Introduction

The main goal in nuclear forensics is to characterize radiological material with regards to its isotopic composition, provenance, age and/or history. The conventional tools applied within this field of study generally require direct access to the source material of interest, or remnants of that material. However, the ability to perform forensic analysis at a distance, whether spatially or temporally, would provide an attractive toolset for both nuclear nonproliferation and safeguards applications. The purpose of this work is to illustrate how solid state dosimetry (SSD) of small samples may soon allow for retrospective nuclear forensics analysis using commonly disregarded mineral material.

The principle mechanism in SSD is charge trapping by lattice defects in crystalline insulator materials. During irradiation, electrons liberated from atomic bonds may become trapped at lattice defect sites such that the population of trapped charges is proportional to the radiation dose received. Once trapped and in the absence of additional stimulation, the populations of trapped charges will be stable over very long periods of time. To the extent that the populations of trapped charges can be quantified and the dose response for the crystalline material can be characterized, then the dose to the material can be reconstructed. The ability to determine the doses received by crystalline materials allows for the possibility to characterize historical radiation environments. The two main methods used, in this work, to quantify material dose are luminescence dosimetry and electron paramagnetic resonance (EPR).

Luminescence dosimetry uses the light emitted by materials upon optical or thermal stimulation to determine the population of trapped charges in a previously irradiated material. Additionally, these methods are best suited to determining the doses to inorganic insulator materials, such as silicate or aluminum oxide ceramics. The utility of luminescence dosimetry for assaying radiological sources using ubiquitous materials has been well documented in the literature. [1-3] For example, using thermoluminescence (TL), it has been shown that common bricks can serve as gamma ray spectrometers with 10% energy discrimination for ^{241}Am at the 1 standard deviation level [1]. This is based determining the empirical mass energy absorption coefficient $\mu_{en}(E)$ from the exposure based on the measured dose depth profile in the materials. Knowing the material composition then allows determining what energy the exposure had to have to give the measured value(s) of $\mu_{en}(E)$. Using optically stimulated luminescence (OSL), it has been shown again with these methods that ^{137}Cs and ^{60}Co can be identified and imaged using common bricks [2]. Likewise, surface mount resistors (such as in portable memory flash drives, credit cards or other electronics) are capable of recording dose approaching common background levels using these techniques [3].

Electron paramagnetic resonance dosimetry uses resonant absorption of precisely tuned microwaves under an applied magnetic field to quantify the number of radiation-induced trapped charges. Unlike luminescence dosimetry methods, EPR is generally best suited for organic

insulating materials. EPR is also recognized as the gold standard for epidemiological dose reconstruction for populations exposed to anthropogenic external radiological sources [4]. Previous work with sucrose crystals has demonstrated detection limits ranging from 500 mGy [5,6] up to 1.5 Gy [7,8] to list a short sample from multiple studies.

By utilizing a combination of EPR and thermoluminescence dosimetry, it should be possible to characterize historic radiation environments in nearly every corner of the developed world. Additionally, extending these techniques to small sample sizes would allow for forensics analysis using particulate material fortuitously collected during routine air and smear sampling. For example, plume modelling can be used to reconstruct historical releases by distant nuclear facilities [10]. If the aerosolized particulates in such a release are composed of nonconducting materials, then these particles can in principle be used as dosimeters with solid state techniques such as luminescence and EPR. While current techniques focus only on chemical and isotopic signatures in such samples with the added potential to conduct morphological analysis, dosimetric analyses could be performed on the particulate matter that is generally discarded. Recent work has explored this potential to use ubiquitous dust particulate from smears and air filters as common dosimeters via luminescent techniques [11].

In the case of luminescence dosimetry, it has been shown in the literature that using specialized instrumentation and measurement protocols accurate dosimetric reconstructions can be performed on particulate matter the size of a single grain of sand. [12] Such sample sizes are what one might expect to collect on an air filter or environmental smear. Similarly, using sophisticated sample measurements protocols our technique allows precision of a few percent with tooth enamel [13] or alanine [14] which has been shown to improve detection limits by an order of magnitude over traditional techniques [15].

1.1 Complementary Techniques

As with any detector modality, the detection limit depends on background signal amplitudes. When using EPR and luminescent techniques, the background typically depends on sample age and its inherent solid state chemistry. This latter component will affect sensitivity, signal stability and even native interfering signals, which can include sample preparation effects.

While luminescence dosimetry has proven useful for dosimetric measurements on particulate matter, the ability to perform single grain EPR measurements remains to be demonstrated. For EPR, the decision limit for any dose is dependent on the minimum detectable spin density of the spectrometer system [16]. Under ideal conditions, the minimum detectable number of spins for a commercial X-band spectrometer is approximately 10^{12} [16]. Since the spin density in the sample is proportional to the radiation dose received, it is likely that EPR dosimetry on small particulate samples would be relegated to only those samples with large doses. However, since many organic insulating materials tend to have very large saturation doses small aliquot EPR dosimetry may still be a viable option [17].

Most luminescence materials have a drastic light attenuation property such that exposure to ambient sources will cause the signal to decay drastically (having half lives on the order of hours) [18]. Consequently, for luminescence dosimetry analysis, environmental samples would ideally be collected from locations shielded from ambient light sources, such as underneath painted surfaces. Organic insulators often are not subject to the same light sensitivity and so may offer a more robust approach to scavenging dosimetric information from air samples and smears.

Despite the potential challenges inherent to luminescence dosimetry or EPR dosimetry, it is proposed here that having access to both will allow for robust characterization of historical radiation environments using scavenged materials.

2. Example Results

Given the intractable number of materials which are insulators to any extent, the literature reviews on common examples should be consulted for specific types if they have been evaluated [13,14]. A few new examples of potential interest in nuclear forensics carried out in our lab will be offered here but the full range of possibilities is expectedly yet to be explored.

The typical approach in solid state dosimetry techniques is to measure the initial dosimetric signal and then via subsequent irradiation to determine the sample sensitivity to radiation and so back extrapolate to the initial dose obtained by the sample prior to laboratory measurement. If the initial signal is measured as *Si* and then the sensitivity is signal intensity per dose given by the symbol *SIpD*, then the sample dose estimate is easily obtained by the ratio *Si / SIpD*. More generally this is done by additive irradiations where the subsequent measurements follow a linear trend, which is least squares fit, to back extrapolate to the initial dose.

2.1 Combined TL/OSL and EPR

Typical experimental configurations have been given elsewhere for luminescent and EPR parameters [18,19]. An example of the luminescent and EPR results from diatomaceous earth are shown in Figures 1 and 2 respectively [20]. The fit parameters from Figure 1 are given in Table 1. Note that when dose values become large (which is materially dependent), then the dose response is no longer linear but follows a saturating exponential profile as seen in figure 3. Here, the results were shown to be fairly insensitive to luminescence methods but offered the hope of new EPR properties very different from geological quartz samples.

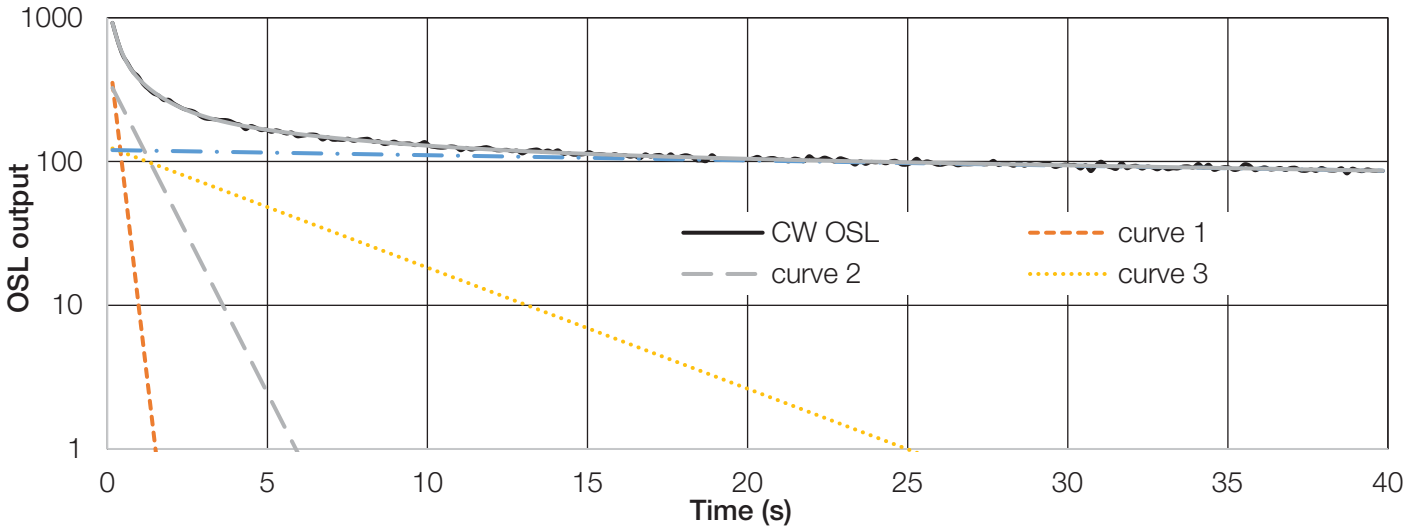

Figure 1: Deconvolved OSL spectrum from diatomaceous earth [20] where the measured signal is the continuous wave OSL (CW OSL) with fit parameters provided in Table 1. Note that the ordinate axis is logarithmic so all the fits are simple exponential decay curves.

Curve #	*A* amplitude parameter	*A* amplitude uncertainty	λ decay parameter	λ decay uncertainty
1	709	29	4.36	0.23
2	382	20	1.01	0.071
3	127.1	8.5	0.194	0.016
4	120.2	2.7	0.00834	0.00068

Table 1: Curve fit parameters from Figure 1 using the exponential model for each fit parameter being given by the functional form of $f(t) = Ae^{-\lambda t}$. where A is the amplitude parameter and λ is the decay parameter.

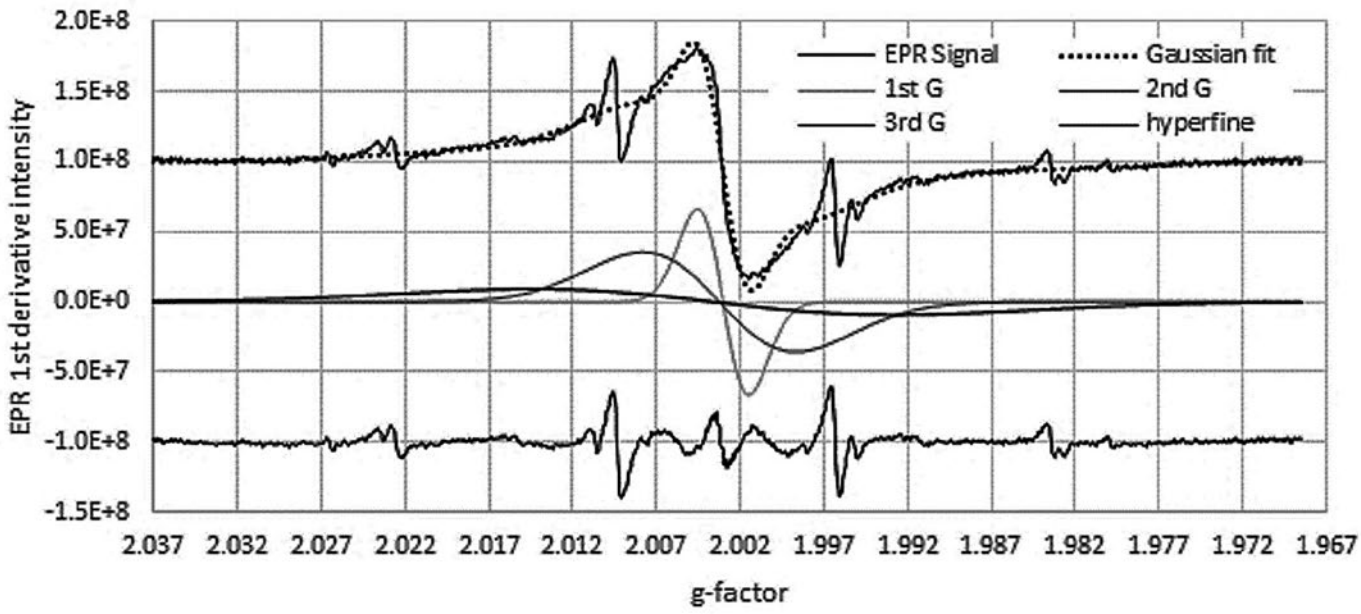

Figure 2: Deconvolved EPR spectrum from diatomaceous earth [20]. Various Gaussian components are deconvolved and shown in the central set being listed as 1st G, 2nd G and 3rd G, respectively. The residual unresolved spectral components are shown at the bottom as the hyperfine portion of the signal.

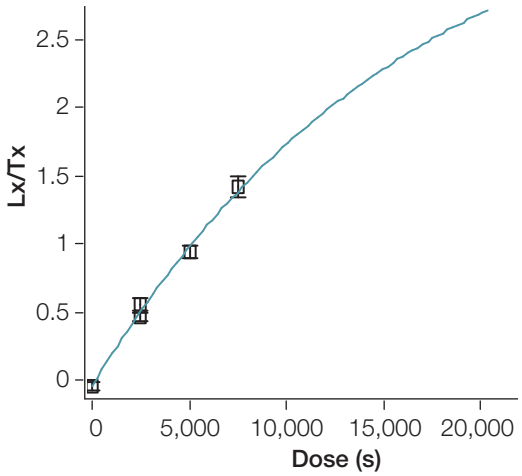

Figure 3: Example saturating exponential dose response shown as a function of additional exposure time in seconds. The functional form when trap centers are approaching maximum population results in a dose response following the functional $f(s) = k\left(1 - e^{-(s-E)/D}\right)$ where E is the resultant dose estimate, k is the maximum signal amplitude at infinite dose and D is the saturation dose.

2.2 EPR

Sucrose is known to be an excellent EPR dosimeter having high sensitivity, low detection limits and long signal stability [8]. Using the same techniques previously shown to have drastically reduced the detection limits for alanine dosimetry using EPR [15], the results from Figure 4 were obtained using 0.5 g aliquots. Here, although a goniometer was not utilized the results demonstrate an 80 mGy standard deviation of the residuals. The rest of the curve fit parameters are shown given in the inset of the figure.

A measured dose response, such as that shown in Figure 4, is still in the linear range; therefore, the sample sensitivity can be taken to be the slope of the least squares fit. Coupling such a dose response function with an initial signal intensity allows the equivalent initial dose to be determined. Although these results were not obtained using single sucrose granules, the linearity of the dose response out to 10 Gy is promising.

The intercept in Figure 4 represents the combined native organic signal and accumulated background radiation. If the signal were purely anthropogenic, the *x*-intercept of 150 mGy would be the resultant dose estimate for this sample. The standard error of the fit value 0.19 EPR counts per mGy represents the 1 σ level for a 0.5 g sample based on a residual analysis. Taking a single grain of sugar to be approximately 0.6 mg, then the EPR response for a single grain would be expected to be near 0.19x0.6/500 = 0.23×10^{-3} EPR mGy^{-1}. To then just approach the variation seen in Figure 4 would require around a full Gy of radiation dose. Taking into account all combined errors otherwise ignored in this estimate and going to the 95% CL, the detection limit should be many Gy for a single grain. Even if difficulties prevent detection below 10's of Gy, this would still be within the needed to linear range of the dose response to accommodate single granule dosimetry by EPR as the linear range extends to the kGy range [21].

2.3 TL/OSL

A recent development of forensic interest is the potential to use common electronic components as dosimeters at the natural background level [3] similar to that done with sugar in section 2.2. The ability to discriminate doses at the level of environmental exposures has the potential to radically implement ubiquitous sensing. As an example, it was shown in [3] that simply using multiple electrical elements in a cell phone is sufficient to carry out energy spectroscopy sufficient to discriminate low energy gamma emitters from industrial or medical source typically having higher energies.

The concept for using differential attenuation in ubiquitous materials is effectively equivalent to that already used in radiation worker dosimetry. In both TL and OSL dosimetry, 4 separate luminescent elements are used with each being able to individually measure dose. These are placed

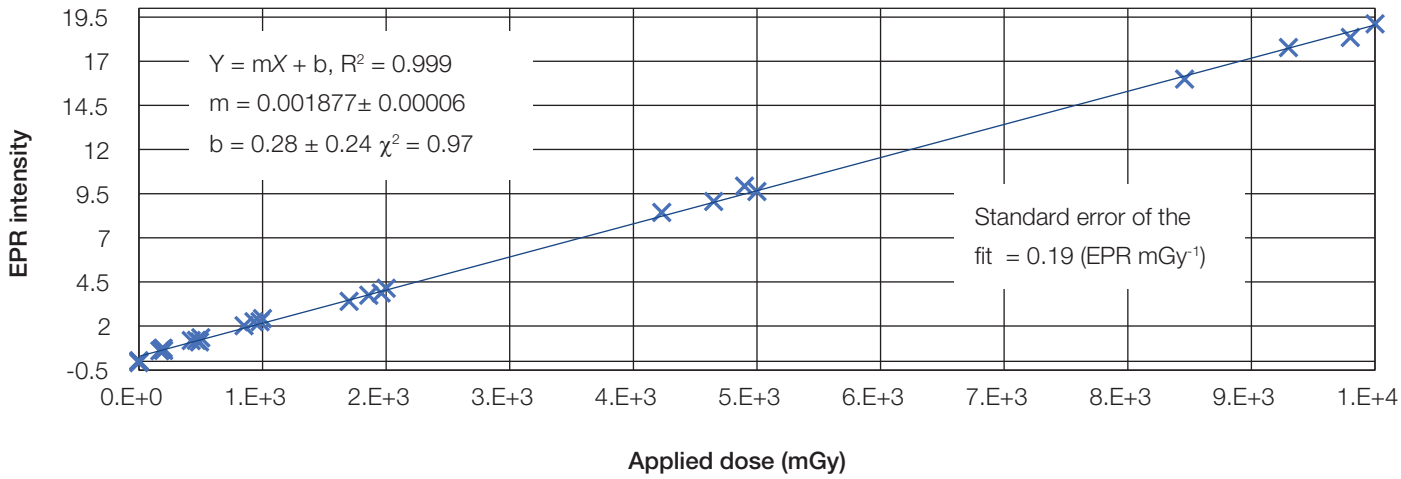

Figure 4: Dose response of commercial sugar granules using techniques initially developed for alanine EPR [13]. EPR values are shown in terms of peak to peak counts.

behind distinct attenuating materials such as tin, Mylar, copper and polyethylene (with the element behind the polyethylene being enriched in a neutron absorbing isotope). Using the resulting differential response of the 4 elements along with functional relationships among the same, algorithms can then discriminate between high energy gamma, x-ray, beta and neutron exposures allowing accurate worker dosimetry to be ascribed.

3. Analysis

The utility for developing technologies to determine historical doses to ubiquitous materials is exemplified in Figure 5. This generic bunker configuration would expectedly be under international treaty control requiring that the warheads remain in-place between successive inspections. If the entry controls are in any way spoofed, then the owner of these items could have deployed them intermediate to any treaty verification inspections. Utilizing these solid state dosimetry techniques, the dust particulate on the very walls of the bunker could determine if the ambient dose rate was consistent with the long-term placement of the warheads between successive inspections. Likewise, the bricks can provide age estimates as to whether any were new or replaced in any kind of spoofing attempt.

The application in forensics as proposed here is not a panacea of detection solutions. The measurement is only a single long-term acquisition. This means that a large dose rate source in motion (such as a spent fuel bundle) would appear as a distributed source along the path of the movement. If any imaging approach were attempted [2], then multiple samples would be required from well-defined locations.

In general, the method only provides some form of the product between dose rate and integration time (hence dose). If the time is known, then a dose rate or activity could be estimated. Conversely, if the source activity were known, then an integration time could be obtained. This in turn assumes intervening materials were known to account for shielding and scatter. Depending on operational knowledge, an upper bound may only be possible on a time window; this would then offer a lower bound on the source activity or dose rate giving rise to the resultant dose. All this of course assumes a single known, or assumed, source.

3.1 Caveats, limitations and considerations

As with any detector, SSD approaches have limitations although these vary greatly from those of electrically powered detector systems. Modern radiation detection systems have integration times ranging from seconds to days, where the integration time directly corresponds to the temporal resolution of that system. An SSD material would offer only a single measurement with an integration window typically being the lifetime of that material. The quartz in brick would provide the accumulated dose to that material all the way back to the time it was baked. Ambient quartz or feldspar from smears or air samples would give an OSL dose estimate back to the time it was zeroed based on exposure to sunlight or any other sufficiently intense optical photon source.

Depending on the material, non-radiogenic, native signals may exist and would need to be subtracted from the resultant measurements. Sample preparation can likewise induce interfering signals, which require identification and mitigation. To further complicate the approach, a number of factors can work to decrease the signal to noise ratios of these dosimetric materials. For example, internal

Figure 5: Nuclear weapons storage (creative commons license image).

radioactivity contributes to background signals not associated with the anthropogenic exposure of interest; time-dependent signal decay decreases the signal with increasing time from the event of interest; and variable sensitivity of sample materials complicates with determination of the minimum detectable limit of a given sampled material. All of these variables have to be properly understood and accounted for in order to provide precise dosimetric estimates from opportunistically scavenged sample material.

Perhaps the largest drawback to using SSD as a forensics technique is the overall throughput for obtaining results. Typical dose estimates from ubiquitous materials such as brick, wallboard (sheetrock or plasterboard) and confectionary range require days to weeks to produce. In this sense, the time commitment to obtaining quality results is comparable to modern radiochemistry and so does not allow rapid analysis unless quality is commensurately poor.

Other groups have proposed portable OSL systems [22] but without TL, they are of limited value in determining sensitivity, which severely limits their overall utility. Similarly, portable EPR systems have likewise been proposed but have also been shown to have very low sensitivity [23]. Currently, laboratory based SSD systems are required to obtain meaningful results which are now able to start approaching background levels.

3.2 Spoofing

As with any detector, surrogate sources could be envisaged, designed and constructed. Similarly, this detector could be annealed or made impotent. If an evasive actor were to heat all the walls around a source up to 500° C, then the TL/OSL/EPR signals would all be zeroed back as though it had just been created in the kiln. Alternatively, an actor could construct their walls out of conducting metal and keep the environment meticulously clean (dust-free) so as not to leave ambient dosimeters about. Even then, the lighting would require some form of insulation to enable workers to be productive without requiring miner's lamps to conduct operations.

Even an annealed wall will retain tell-tale SSD signatures if they are not uniformly heated to 500° C. Assuming this can be done, it would have to occur in such a way that heating the room itself did not leave evidence such as requiring all new wood, plastic and rubber peripherals as these would all combust or melt under such a kiln treatment. Still, removing a full brick may be reasonably rejected due to building integrity if reparative measures are not properly implemented.

Given the requirements inherent to spoof an SSD forensics approach, it would be prohibitively difficult to fully erase all evidence of noncompliant actions on the part of any nuclear state. While spoofing activities would add additional uncertainty to the forensic analyses presented above, it should be noted that more advanced analysis methods exist that could be applied to such problems. In the worst case, a bounding analysis on gross dose estimates would likely be sufficient to determine if the environmental dosimetric records agree with operator declarations. Residual evidence of spoofing itself may be actionable without source reconstruction whatsoever but details would be specific to particular scenarios.

4. Discussion

The utility of measuring the electronic properties of nonconducting materials enables these to become effective dosimeters. Further, the techniques described herein are, in general, applicable to samples collected from bulk materials (i.e. bricks and tiles) down to single grain samples (aerosolized particulate matter). As one might expect, the information available varies with sample size (lower doses generally requiring more sample). As current particulate analysis protocols consider only chemical, isotopic and morphological properties, adding in electronic property characterization with TL, OSL and EPR can provide additional information previously overlooked.

For example, with bulk sample analysis, differential attenuation in the material provides a means by which the energy of the radiation field can then be obtained (albeit at a low effective resolution). Even using multiple samples from a gridded array offers the potential to reconstruct the location of the nuclear material (via inverse square and any shielding considerations) such that the technique can serve as a gamma camera [2].

On the other hand, combining SSD in particulate matter with plume reconstruction techniques may allow for forensic analysis at large standoff distances. To the extent that the atmospheric transport of nonconductive particulate matter mirrors the transport of other materials released by nuclear facilities, one can envision performing dosimetric analysis on particles released by such facilities. In this way, it may be possible to gather information about the activities being performed within. Realization of this capability would allow verification of nuclear operations from a distance, without the need to even step foot inside the facility itself.

As a general statement, most organic materials are better suited for EPR measurement. Alternatively, inorganic insulators tend to work better with TL and OSL characterization. TL and OSL do require the samples to be at least partially transparent to transport the recombination photons with OSL requiring the stimulation light to also penetrate the grains be analyzed. Most materials have some signal which can be measured with all methods but to widely varying sensitivity, linearity and native signal contributions from each such that initial testing or literature recommendations will often guide the user to a preferable measurement protocol. Even with this, particular variations

in a given material type could in principle warrant customization of any portion of the sample preparation approach and/or measurement sequence.

By combining the applications of EPR with TL and OSL, all insulator materials can be utilized. One drawback with common environmental materials is the potential for a large geological dose background. This can be partially mitigated using various techniques presented by others [24] although at the cost of decreased quality in the resulting dose estimate. However, given the variety in materials that this combination of techniques may utilize, there is reasonable likelihood that in any given sample, some useful material could be scavenged.

5. Conclusions

The potential applicability for SSD in nuclear forensics is effectively in its infancy. The realization that the electronic populations of ubiquitous, insulating materials can be used to reconstruct the gross position and energy source material, retrospectively, has implications which are lightly reviewed in this work If the success of prior studies could be extended to particulate matter from environmental smears or air samples, then it would be possible to perform dosimetric assay for treaty verification without direct access to the facility. This would mark a significant improvement in the current suite of tools currently employed for treaty verification and nonproliferation.

6. Acknowledgements

7. References

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