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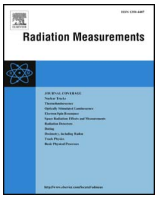

# OSL in NaCl vs. TL in LiF for absorbed dose measurements and radiation quality assessment in the photon energy range 20 keV to 1.3 MeV

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ABSTRACT

The aim of this study was to determine the photon energy dependence of absorbed dose measurements, in a comparison of optically stimulated luminescence (OSL) in NaCl with thermoluminescence (TL) in LiF:Mg,Cu,P. The comparisons were made at exposure to ionizing radiation in the photon energy range 20 keV to 1.3 MeV. Specially designed dosemeter kits containing both NaCl and LiF were used under *i)* laboratory conditions using defined radiation fields, *ii)* laboratory conditions using sealed point sources mimicking unintentional exposures, and *iii)* field conditions in areas in Japan that were affected by the Fukushima Daiichi nuclear disaster in 2011. The dosemeter kits used in Japan showed that absorbed doses as low as 100 μGy can be assessed from the OSL signal in NaCl. The ratio of the dosemeter readings using OSL in NaCl and TL in LiF increases after irradiation at lower photon energies (less than a few hundred keV) as determined under laboratory conditions. Compensating for this energy dependence of the absorbed dose determinations obtained from OSL in NaCl would thus require an energy-dependent conversion factor for photon energies below 600 keV. On the other hand, the difference in the photon energy dependence between NaCl and LiF may be used to assess the mean effective energy of the photon field. The signal ratios between NaCl and LiF after exposure to radiation in the Fukushima Dashii contaminated areas in Japan, 1.67 ± 0.26 (2013) and 1.63 ± 0.32 (2015), indicate that the mean photon energy in this area was 300–400 keV during the years of the survey.

## 1. Introduction

In retrospective dosimetry for emergency dose assessments, optically stimulated luminescence (OSL) or thermoluminescence (TL) in common crystalline materials accessible in public surroundings have proven to be very useful (e.g., Bøtter-Jensen, 1995; Hütt et al., 1996; Sholom and McKeever, 2014; Mesterhazy et al., 2014). In particular, household salt (consisting predominantly of NaCl) exhibits a significantly higher OSL signal per unit of absorbed dose than many other materials after exposure to high energy beta ($^{90}$Sr/$^{90}$Y) and gamma ($^{60}$Co) radiation (e.g., Thomsen et al., 2002; Bernhardsson et al., 2009; Christiansson et al., 2012).

Given the potentially high sensitivity of this technique for absorbed dose determination, studies have been launched to investigate its performance as an environmental and personal dosemeter for external gamma radiation exposures (e.g., Bernhardsson et al., 2011, 2012; Ekendahl et al., 2016). NaCl is a low-cost detector material that enables fast and straightforward sample preparation as well as calibration. For workers such as first responders in a radiological or nuclear emergency who have to operate in very different radiation fields, a dosemeter of this type would be of great value. Also, since radiation dosemeters are often not a budget priority for many employers when the probability of radiation exposure is low (e.g., for public safety personnel), a simple, cost-effective dosiemeter could allow all workers to have a personal dosemeter.

It is important to know the energy response of the detector material for accurate dose determination. Depending on the radiation detector material's effective atomic number and the type of stimulation, the photon energy response can present as either an under-response or an over-response relative to air in the low-energy range (e.g., Harder and Hermann, 1985). For personal dosimetry, it would be an advantage if the material used has the same (or similar) effective atomic number ($Z_{eff}$) as soft tissue ($Z_{eff} = 7.65$) (ICRP) or water ($Z_{eff} = 7.51$) (Bos, 2001). In theory, such detector materials can exhibit a signal response based on the dose absorbed by tissue that is independent of the photon energy, in the range from about 10 keV to 3 MeV. Lithium fluoride (LiF) has been a standard dosemeter material for prospective dosimetry in the past decades (Bos, 2001; Kortov, 2007). In large part this is due to

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its effective atomic number ($Z_{eff} = 8.3$), which makes its energy dependence more similar to that of soft tissue and thereby makes it suitable for personal dosimetry. Photon energy dependence is less important for materials with a $Z_{eff}$ close to that of soft tissue, especially at energies of a few hundred keV where the photoelectric effect is dominant. However, many detector materials are not tissue-equivalent. Household salt (NaCl) has a significantly higher effective atomic number ($Z_{eff} = 15.2$) (Murty, 1965) than that of soft tissue, making it more sensitive to photons with low energy (< 400 keV) than to those with high energy. Other well-known detectors with higher effective atomic numbers are $Al_2O_3$ ($Z_{eff} = 11.3$) and $CaF_2$:natural ($Z_{eff} = 16$). In order to use NaCl for personal dosimetry, it is therefore necessary to investigate the energy dependence of the absorbed dose in relation to that of soft tissue. In a previous study, the OSL signal in NaCl was compared to the TL signal in LiF after exposure to a $^{60}$Co beam ($E_{\gamma1} = 1173$, $E_{\gamma2} = 1333$ keV) at a depth of 7 mm in a polymethylmethacrylate (PMMA) phantom (Christiansson et al., 2014). The ratio of OSL-NaCl to TL-LiF was determined to be 1.0 ± 0.03, which is expected at photon energies where the Compton scattering interaction is dominant.

The aim of this study is to investigate whether NaCl may be used as an alternative to or in combination with LiF to determine absorbed doses at lower photon energies, down to 20 keV (Malthez et al., 2014). This may practically be tested under both laboratory and *in situ* conditions by determining the ratio of NaCl and LiF dosemeter readings for photon energies ranging from 20 keV to 1.3 MeV. The long-term objective is to find a physical design and read-out procedure to retrieve an OSL signal in irradiated NaCl that is suitable for estimating the dose absorbed by tissue for assessment of radiation exposure.

## 2. Materials and methods

### 2.1. Sample preparation

In a previous study, four brands of household salt were investigated to determine both their OSL response to gamma and beta radiation and their OSL signal integrity (Christiansson et al., 2014). In the present study, one of the previously investigated salt brands ("Falksalt fint salt med jod") is used for all OSL measurements in NaCl as it showed a high sensitivity to radiation and is also widely available in Swedish supermarkets. Hereafter, "Falksalt fint salt med jod" will simply be referred to as salt or NaCl.

Special dosemeter kits were constructed for the purpose of testing household salt as a personal dosemeter. Two different types of holders were used for the dosemeter kits. Holder No. 1 (Fig. 1) was previously developed for estimating external effective doses in Russian populations after the Chernobyl accident. This was accomplished by determining the surface absorbed dose and then applying correction factors (Bernhardsson et al., 2012; Thornberg, 2000; Wøhni, 1995). The holder was made of two PMMA plates with dimensions of 58 × 27 × 4 $mm^3$. Between the PMMA plates, each kit was prepared with about 80 mg of NaCl and two LiF chips of LiF:Mg,Cu,P (MCP-N, Mikrolab, Poland). The salt and the LiF chips were protected from environmental humidity and separated from each other by individual rubber O-rings (in total, four O-rings per dosemeter: two with NaCl and two with LiF chips). In order to optimize the amount of salt in the dosemeter holder, and in accordance with previous sampling procedures for salt (Christiansson et al., 2008), the salt was sieved to grains in the size range from 100 to 400 µm. The thickness of the NaCl portions was approximately 2 mm (see Fig. 2).

Holder No. 2 (Fig. 1) was designed especially for the experiment with the reference X-ray beams due to physical constraints. The thinner salt layer (~1 mm) was used to more accurately measure the absorbed dose from radiation components with lower photon energies. Holder No. 2 was made of two PMMA plates. The top plate had dimensions of 10 × 10 × 5 $mm^3$, and the bottom plate had dimensions of 10 × 10 × 6 $mm^3$ (see Fig. 2) Each had four milled holes for NaCl grains (about 50 mg) and two LiF chips. The thickness of the lid of Holder No. 2 was the same as the thickness of the entrance PMMA plate for the calibration phantom used in routine TLD calibrations.

Before preparing the dosemeter kits with NaCl, any remaining OSL signal in the salt was erased by exposing it to light from a fluorescent lamp with an illuminance of 1100 lux (0.16 mW $cm^{-2}$) and a wavelength range of 300–700 nm for at least 2 h (Christiansson et al., 2014). The dosemeter kits with NaCl and LiF were assembled and then covered with black tape to avoid signal loss in the salt due to optical bleaching.

For $^{60}$Co photon energies (1173 and 1333 keV), both the LiF chip (0.9 mm) and salt layers of 1 or 2 mm, respectively, will be relatively accurately approximated as Bragg-Gray cavities positioned at a point with quasi-charged particle equilibrium (provided by the 4 or 5 mm PMMA layer in front of the salt). However, in this study, the design of a 1–2 mm thick NaCl layer was a compromise between the Bragg-Gray cavity conditions and the fixed thickness of the LiF chips used as reference detectors.

### 2.2. Calibration and readout of OSL in NaCl

The OSL measurements were carried out in an automated TL/OSL reader (TL/OSL-DA-15; Technical University of Denmark, Risø campus, Roskilde, Denmark). After each exposure, salt was distributed on stainless steel cups. Small aliquots of about 5 mg salt per sample cup were portioned in a thin layer during darkroom conditions. These conditions were established by using red plastic film (106 primary red, LEE filters, www.leefilters.com) in front of the lamps in the laboratory

**Fig. 1.** Left frame: PMMA Holder No. 1 with two rubber O-rings for the NaCl and two for the LiF chips. Two nylon screws were used to tighten the PMMA plates together and to avoid any moisture from entering the dosemeter cavities. Right frame: PMMA Holder No. 2, with two cavities for the NaCl and two for the LiF chips.

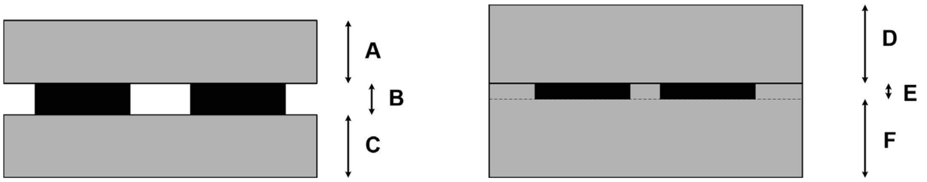

**Fig. 2.** Left frame: PMMA Holder No. 1 in profile. A = 4 mm, B = 2 mm, C = 4 mm. Right frame: PMMA Holder No. 2 in profile. D = 5 mm, E = 1 mm, F = 5 mm.

**Table 1**
Gamma-emitting radiation sources and X-ray units used for laboratory investigations of OSL in NaCl vs. TL in LiF.

| Radionuclide | Photon energy (keV) and branching ratio (% per decay) | Activity and reference date | Holder type |
|---|---|---|---|
| Ba-133 | 81 (34%)<br>276 (7%)<br>303 (19%)<br>356 (62%)<br>384 (9%) | 370 MBq (2006-07-15) | No. 1 and No. 2 |
| Cs-137 | 662 (85%) | 370 MBq (2006-07-03) | No. 1 and No. 2 |
| Co-60 | 1173 (100%)<br>1333 (100%) | 370 MBq (2006-07-14) | No. 1 and No. 2 |
| Mammography 23 kV | 18.1 | | No. 1 and No. 2 |
| | Mean photon energy (keV) | | |
| Mammography 29 kV | 19.1 | | No. 1 and No. 2 |
| X-ray 60 kV | 37 | | No. 1 |
| X-ray 125 kV | 56 | | No. 1 |
| Standard X-ray and gamma ray beams | | | No. 2 |

during the sample preparation. A test dose was used to compensate for possible variations in the sample sensitivity (e.g., originating from a variable amount of mass per aliquot). The test doses were provided by an internal source of $^{90}$Sr/$^{90}$Y (20 MBq, 2009-04-09). The dose rate to quartz at the irradiation position of this source was determined to be 0.80 ± 0.02 mGy s$^{-1}$ (2016-08-03) using sensitized quartz (Batch 101, DTU Nutech) as a standard.

During the readout, all samples were stimulated by continuous wave (CW) light from blue LEDs, with a peak emission at 470 nm. A preheat temperature of 220 °C for 10 s was applied, followed by a CW-OSL readout at 100 °C for 40 s. The NaCl dosemeters were calibrated in a PMMA -phantom (191 × 191 × 77 mm$^3$, 5 mm entrance window) in a $^{60}$Co beam using absorbed doses in water of 0.3, 0.5, 1.1, 2.1, 4.2, 6.4, and 12.7 mGy. The OSL signal of each given dose was determined and a linear regression was adopted for the NaCl calibration curve.

For the particular set-up used in this study the various sources of uncertainties are estimated to be: 5% for the calibration dose of the set-up; c. 5% for the standard deviation in the OSL-signal of replica of the sample; and 7% for the average deviation from the calibration line of each aliquot. The contribution from the fading factor is judged to be negligible due to the use of the same time span between irradiation and OSL-readout for the calibration source as for the experimental irradiations. Using the same uncertainty propagation as presented in Ainsbury et al. (2017) for the OSL readout, a relative uncertainty for the absorbed dose to water using NaCl of 10% have been assigned to the dose determinations.

## 2.3. Calibration and readout of TL in LiF

LiF chips of LiF:Mg,Cu,P (MCP-N, Mikrolab, Poland) with dimensions of 3.2 × 3.2 × 0.9 mm$^3$ were used as a comparison to the NaCl. The LiF chips were heated before each irradiation to 240 °C for 10 min while placed in an aluminum holder (using an annealing oven, Nabertherm, Germany), followed by rapid cooling to room temperature on a large aluminum block. The LiF chips were calibrated in a PMMA phantom (191 × 191 × 77 mm$^3$, 5 mm entrance window) in the earlier mentioned $^{60}$Co beam. Each LiF chip was individually calibrated using an absorbed dose in water of 0.3, 0.6, 1.2, and 6 mGy. The TL signal of each LiF chip was determined and a linear regression was adopted for individual calibration curves. After irradiation, the LiF chips were heated to 100 °C for 10 min prior to readout in the TL/OSL reader. Readout of the TL signals was performed using a linear heating rate of 5 °C s$^{-1}$ up to 240 °C, after which the temperature was held constant for 60 s to assure a high depletion efficiency of the signal.

In analogy with the doses determined by OSL-NaCl measurements, the uncertainty on the absorbed dose to water by means of TL in LiF is estimated as below 5% based on data presented by e.g. Shen et al. (2002) and Glennie (2003).

## 2.4. Irradiation geometries

The dosemeter kits were tested during different irradiation conditions: *i*) positioned on the outside of a solid rectangular PMMA phantom (191 × 191 × 77 mm$^3$) when irradiated in the laboratory, *ii*) positioned on the outside of a solid rectangular PMMA phantom (191 × 191 × 77 mm$^3$) when irradiated in a X-ray unit, *iii*) placed in a calibrated radiation unit at IRSN in France (Holder No. 2 only), or *iv*) placed on the chest of persons working in a contaminated area in Japan (Holder No. 1 only).

### 2.4.1. Dosemeter kits calibrated in the laboratory

The dosemeter kits were positioned on the PMMA phantom described in the previous section and irradiated with the radiation sources given in Table 1. Each exposure was carried out at a distance of 0.5 m. The irradiations with the $^{133}$Ba and $^{137}$Cs sources were conducted in a device that allows several different samples to be homogeneously irradiated at the same time. It used one or more radiation sources at distances ranging up to 0.9 m from the samples (for the purpose of varying the dose rate). As it was possible to rotate the samples around the source and vice versa, different irradiation geometries were possible.

### 2.4.2. Dosemeter kits irradiated in a mammography unit and X-ray unit

The dosemeter kits were also used in a clinical environment by exposing them to radiation from a mammography unit and an X-ray unit. The dosemeter kits were positioned on the PMMA phantom (described above) that in turn was positioned on the X-ray table. The field of view was maximized to cover the whole PMMA phantom and a series of exposures were performed.

### 2.4.3. Dosemeter kits irradiated in the laboratory at IRSN

The dosemeter kits were irradiated in standard X-ray and gamma-ray beams in the calibration laboratory at Institut de Radioprotection et de Sûreté Nucléaire (IRSN, France). The following reference fields were used: N-25, N-40, N-60, W-80, N-80, N-100, Cs-137, and Co-60, as defined in the ISO 4037–1 standard (ISO, 1996). The fields had mean photon energies of 20, 33, 48, 57, 65, and 83 keV, respectively. Air kerma rates at the irradiation positions were converted into the corresponding absorbed dose rate in water using a stopping-power ratio of

1.104–1.138 (depending on energy) between water and air (NIST, https://physics.nist.gov/PhysRefData/Star/Text/ESTAR.html).

#### 2.4.4. Dosemeter kits used in a contaminated area in Japan

In May 2013 and November 2015, five NaCl/LiF dosemeter kits (Holder No. 1) were worn by five staff members who were working five days in the contaminated zone in the Fukushima prefecture, Japan. In addition to their mandatory supply of electronic dosemeters (T404 PED, Tracerco, United Kingdom), the dosemeter kits were placed on their chest. The dosemeter kits used in Japan were assembled in Sweden one day before departure to Japan. One of the kits was positioned inside a hotel room in the center of Fukushima city, which is situated outside the closed and contaminated area, to correct for the natural background radiation and the contribution to the signal by the roundtrip flight.

In 2013, two dosemeter kits were worn by two different persons working in the contaminated zone with varying dose rates. In 2015, dosemeter kits were worn by three persons working in the same contaminated zone. According to the electronic dosemeters carried by the staff, the dose rate in the investigated areas ranged from 5 to 65 μSv $h^{-1}$.

## 3. Results

### 3.1. Dose response of exposure in laboratory conditions

The energy response of LiF:Mg,Cu,P (MCP-N, Mikrolab, Poland) is relatively flat. The measurements show that NaCl has a large over-response for photon energies below 200 keV compared with LiF when the ratio between the NaCl and LiF readouts is plotted as a function of gamma ray energy (Fig. 3). The ratio in the signal readout of the two dosemeter materials (as defined in Sections 2.3 and 2.4) appears to peak at 40 keV, reaching a value close to 13. Above 40 keV, the ratio monotonically decreases as a function of the mean photon energy. This feature can be used to estimate the mean photon energy in the environment surveyed by the dosemeter kits. At photon energies above 200 keV, there appears to be only a slight difference in this ratio between the two holder types. Holder No. 1 has a slightly higher ratio for $^{137}$Cs and $^{60}$Co gamma-ray exposures and a slightly lower ratio for $^{133}$Ba exposures.

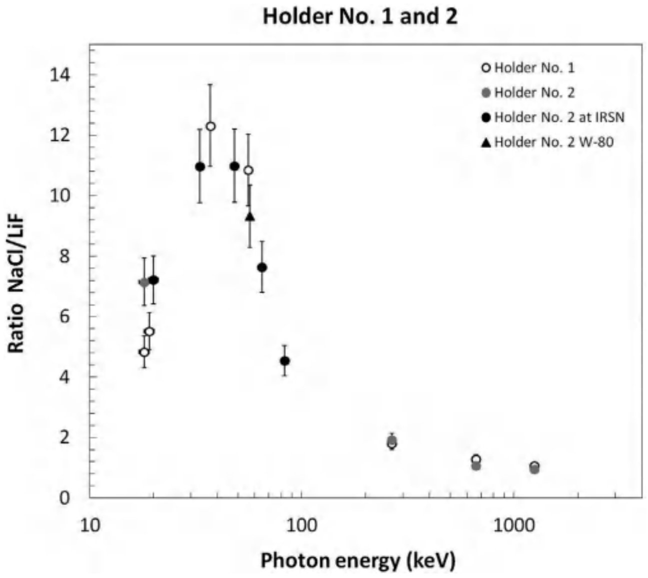

**Fig. 3.** Absorbed dose ratio between NaCl (OSL) and LiF (TLD) using Holders No. 1 and No. 2, as a function of the primary photon energy. The dose absorbed by the NaCl was determined using a calibration curve, and the dose to the LiF chips was determined using individual calibration curves.

**Table 2**
Radiation exposure in terms of the dose absorbed by water for two staff members working in the contaminated zone in Fukushima in 2013, as measured by dosemeter kits positioned at chest height. The background dose (representing the accumulated dose during the entire expedition, from the hotel room and on the flights) is subtracted.

| | Absorbed dose to water OSL-NaCl (mGy) | Absorbed dose to water TL-LiF (mGy) | Ratio OSL-NaCl to TL-LiF |
|---|---|---|---|
| Person A | 0.180 ± 0.018 | 0.104 ± 0.005 | 1.73 ± 0.19 |
| Person B | 0.091 ± 0.009 | 0.057 ± 0.003 | 1.60 ± 0.18 |
| **Mean value** | | | **1.67 ± 0.26** |

**Table 3**
Radiation exposure in terms of the dose absorbed by water for three staff members working in the contaminated zone in Fukushima in 2015, as measured by dosemeter kits positioned at chest height. The background dose (representing the accumulated dose during the entire expedition, from the hotel room and on the flights) is subtracted.

| | Absorbed dose to water OSL-NaCl (mGy) | Absorbed dose to water TL-LiF (mGy) | Ratio OSL-NaCl to TL-LiF |
|---|---|---|---|
| Person A | 0.122 ± 0.012 | 0.070 ± 0.004 | 1.75 ± 0.20 |
| Person B | 0.121 ± 0.012 | 0.070 ± 0.004 | 1.73 ± 0.19 |
| Person C | 0.092 ± 0.018 | 0.066 ± 0.003 | 1.39 ± 0.16 |
| **Mean value** | | | **1.63 ± 0.32** |

### 3.2. Dose response of dosemeter kits under field conditions

The total absorbed doses to the staff members in 2013 and 2015 and the ratio between the NaCl (OSL) and LiF (TLD) absorbed doses are shown in Tables 2 and 3.

The mean ratio between the absorbed doses obtained from NaCl and LiF in the five dosemeter kits worn in the field in 2013 and 2015 was 1.64. The results obtained with the dosemeter kits in Japan indicate that the mean photon energy in the contaminated area was between 300 and 400 keV (by comparing with the ratios in Fig. 3). These results also show that commercial household salt can detect absorbed doses down to 0.1 mGy if using a regenerative dose readout protocol (Duller, 1991; Murray and Roberts, 1998; Murray and Wintle, 2000). However, if only OSL in NaCl readouts are used, without a combination of LiF tablets, then the dosemeter readings need compensation in terms of either a conversion factor (requiring knowledge of the photon energy at the surveyed site) or some filtration using transmission apertures dimensioned to flatten out the over-response observed below 200 keV (Fig. 3).

## 4. Discussion

The energy response of NaCl as a radiation detector material was experimentally evaluated. The energy response of LiF:Mg,Cu,P (MCP-N, Mikrolab, Poland) is relatively flat; however, NaCl has a large over-response compared to the dose absorbed by water. Consequently, a combination of NaCl and LiF dosemeters can be used to assess the mean effective energy of the photon field. As observed in other studies (Malthez et al., 2014), there is a limitation with this method when the ratio is very high, in this case between 20 and 100 keV. In this range, the uncertainty of the mean energy is high.

## 5. Conclusions

- Measurements of the absorbed dose with TL in LiF and with OSL in NaCl agree well for photon energies above 660 keV.
- The results from the dosemeter kits used in Japan show that household salt can be used to detect absorbed doses from photon energies from fresh nuclear power fallout down to 0.1 mGy. However, for accurate dose determination using the current design

of the dosemeter kits, conversion factors are needed for photon energies below $^{137}$Cs primary photon energy (662 keV).

- When used in the field, the absorbed dose by NaCl in the dosemeter kits overestimates the dose retrieved from LiF by at most 75%.
- A combination of NaCl and LiF dosemeters can be used to assess the mean effective energy of the photon field.
- To be able to use NaCl to determine the tissue-equivalent absorbed dose, dosemeter kits with thinner layers of NaCl could be designed. For the purpose of using NaCl as a personal dosemeter for determining the personal dose equivalent, Hp(10), efforts must be directed toward developing a detector design that follows the Bragg-Gray theory.

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## Appendix A. Supplementary data

Supplementary data related to this article can be found at http://dx.doi.org/10.1016/j.radmeas.2018.03.003.

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