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Modification of Shirt Buttons for Retrospective Radiation Dosimetry after a Radiological Event

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Abstract

Preliminary results are presented for a personal radiation dosimeter in the form of a clothing button to provide gamma-ray dose estimation for clinically significant external radiation exposures to the general public due to a radiological incident, such as a Radiological Dispersal Device. Rods of thermoluminescent material (LiF:Mg,Ti and LiF:Mg,Cu,P) were encapsulated in plastic “buttons”, attached to shirts, and subjected to three cycles of home or commercial laundering or dry cleaning, including ironing or pressing. The buttons were subsequently exposed to doses of ¹³⁷Cs gamma rays ranging from 0.75 to 8.2 Gy. The rods were removed from the buttons and their light output compared to their responses when bare or to the responses of a set of calibration rods of the same type and from the same manufacturer. In all three of the comparisons for LiF:Mg,Ti rods the relative responses of the rods in buttons changed by 2–6% relative to the same rods before cleaning. In both comparisons for LiF:Mg,Cu,P rods, the response of laundered rods was 1–3% lower than for the same rods before cleaning. Both these materials are potential candidates for button dosimeters.

Keywords

detector; thermoluminescent; dosimetry; personnel; emergencies; radiological; screening measurements

INTRODUCTION

In the event of a radiological incident, such as a Radiological Dispersal Device (RDD or “dirty bomb”), an Improvised Nuclear Device (IND), a Radioactive Exposure Device (RED) or the release of radioactivity from a nuclear power plant, large numbers of people might receive significant doses of radiation. The dose received by an individual from external exposure would depend on many parameters, such as proximity, length of exposure, and shielding by vehicles or structures, and could range widely. Because it is difficult, time-consuming and imprecise to calculate doses on an individual basis, exposures might have to be assessed by biological dosimetry in order to determine potential risk and triage for

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possible treatment. If the general public wore personal dosimeters, the determination of exposure could be made more quickly, as or more precisely, and probably less expensively since it would require less sophisticated equipment. To this end the design of a personal thermoluminescent dosimeter (TLD) in the form of a clothing button was investigated which would be inexpensive enough that it could be attached to every garment, either during manufacture or at home. The aim of the dosimeter is to estimate clinically significant doses that could require treatment or close observation of the person who was exposed to the radiation.

MATERIALS AND METHODS

Two candidate thermoluminescent (TL) materials were tested: LiF:Mg,Ti and LiF:Mg,Cu,P. LiF:Mg,Ti is about half the cost of LiF:Mg,Cu,P and has been the generally preferred material, however LiF:Mg,Cu,P is starting to replace it in large-scale personnel dosimetry. As of 2006 the U.S. Navy has switched entirely to LiF:Mg,Cu,P, which has superior characteristics relative to LiF:Mg,Ti (Moscovitch 2006) including higher sensitivity, improved energy response for photons, lack of supralinearity, and insignificant fading.

The TLDs were $1 \times 1 \times 6 \text{ mm}^3$ square rods obtained as single batches of 40 Harshaw TLD-100 and TLD-100H from Thermo Electron Corporation (now Thermo Scientific, Cleveland, OH) and as TLD100R and TLD100OR from REXON TLD Systems, Inc. (Beachwood, OH), which for each company were composed of LiF:Mg,Ti and LiF:Mg,Cu,P, respectively. The rods were annealed using the suggested time-temperature profiles for reading listed in Table 1 before initial use with a Harshaw 3500 manual TLD reader (Thermo Electron Corporation). The LiF:Mg,Ti rods were annealed at least once before use and also after being irradiated. Rods that had received larger doses were annealed twice to minimize the remaining signal. The average last annealing reading was 0.06% of the measured dose and never larger than 0.4%. All the LiF:Mg,Cu,P rods were annealed at least twice in the reader before use and between irradiations. The average last annealing reading was 0.5% of the measured dose. In 90% of the measurements the reading was <1% of the measured dose, in 5% (7 measurements) it was 1-1.5%, and in 5% it was 1.5-4.5%. All the readings contributing >1.5% to the signal were for two doses in the same test. These annealing readings are likely overestimates of the contributions to the dose measurements since the signal remaining in the TLD is usually reduced after each reading.

“Buttons” ~11 mm in diameter and 3.2 mm thick, about the size of a standard men’s shirt button, were constructed of polycarbonate or polymethyl methacrylate (PMMA). The buttons consisted of two pieces: a top disc or lid 1 mm thick and a bottom disc 2.2 mm thick with three slots 1.2 mm deep for TLD rods (Fig. 1). The rods were placed in the slots and the lids were adhered to the bottom discs using epoxy, which was applied carefully in order to avoid gluing the rods to the buttons or getting epoxy on the rods, which could alter their responses.

The buttons were sewn onto cotton/polyester shirts and subjected to three cycles of home laundering, commercial laundering, or dry cleaning. For the commercial laundering and dry cleaning, a different cleaning company was used for each cycle. In addition, for the dry cleaning one company used chemical cleaning and two were “organic” (synthetic hydrocarbon solvents). After the laundering cycles, which included ironing or pressing, the buttons were removed from the shirts.

The buttons and TLD rods were irradiated with 0.662 MeV gamma rays using the Gammacell 40 ^{137}Cs irradiator (Best Theratronics Ltd., Ontario, Canada) at the Center for Radiological Research of Columbia University. The irradiator has two sources, one below

and one above the irradiation chamber and had a dose rate of 0.75 Gy min^{-1} at the center of the irradiation chamber. Irradiation times were 1 to 11 minutes. The irradiations were performed by pressing the “On” and “Off” buttons for the source and measured using an external electronic timer.

For convenience, the bare TLD rods were irradiated in a Plexiglas (PMMA) holder purchased from Thermo Electron Corporation. The holder has a top 2 mm thick and a bottom section 4 mm thick with circular holes 1.5 mm deep, so that there was approximately 1 mm more material absorbing the gamma rays on either side of the rods than for the buttons. The extra material would reduce the dose for the dosimeters by less than 1% relative to the buttons.

After irradiation, the buttons were split apart using a razor blade and a custom fixture. A few of the TLDs broke in the removal process, probably because of excess epoxy that got into the TLD slots in the button or too tight a fit in the slot; these were read but not used in the analysis. All the TLDs were read using the planchet for 1 mm rods and the time-temperature heating profile recommended by the Thermo Electron Corporation instructions for the two different types of dosimeter materials (Table 1). No pre-anneal was used for either type of material. The light output readings of the TLDs integrated over all the glow peaks were normalized to the reference light signal of the reader. The reader photomultiplier noise was negligible compared to the light signals for the doses used. Depending on the material, the time to read an individual rod was 1.5-2 minutes, about 1 minute of which was required for cooling.

In the first test, 18 Harshaw TLD-100 rods were put in six buttons (three polycarbonate and three PMMA) and subjected to three cycles of dry cleaning over 18 days. Each cycle was performed by a different cleaner. The rods were in the buttons for total of about one month by the end of the cleaning cycles. The buttons were given a dose of 1.5 Gy about one month after the last cleaning and read four days later. No cohort (bare) rods were irradiated and there were no control (zero dose) TLDs. All the rods were removed from the buttons without breakage. Comparison was made to the light output measured for the same rods when bare for the same dose.

In the second test, 18 Harshaw TLD-100 rods that were used in the buttons for the first test were put in six buttons, three of each type of plastic, and 12 Harshaw TLD-100H rods were put in four buttons, two of each type of plastic. The buttons were immediately subjected to three successive cycles of home machine laundering, machine drying and hand ironing (153°C , as measured using a digital thermometer) over four days. Five days after encapsulation, the buttons and 12-13 bare Harshaw TLD rods of each type were given four doses of gamma rays from 0.75 to 6.0 Gy and read the following day. There were no control TLDs. Five of the 18 TLD-100 rods broke while being removed from the buttons as did two of the TLD-100H rods. Prior to being placed in the buttons, the TLD-100 rods and the 12 TLD-100 calibration rods were irradiated with the same range of doses, providing an element correction coefficient (ECC) for each rod.

The third test consisted of 36 TLD rods. Six polycarbonate buttons contained Rexion LiF:Mg,Ti rods and six PMMA buttons contained Rexion LiF:Mg,Cu,P rods. The buttons were commercially laundered three times over two weeks. Fifteen of the buttons of each type were given six doses from 0.75 to 8.23 Gy on the last day of laundering along with 17 calibration rods of each type. All the rods were read two days later. Three rods of each type in buttons and four rods of each type bare were given no dose and acted as controls. Prior to this test, the TLD rods placed in the buttons and the calibration rods were irradiated with the same range of doses as used in the test so that ECCs could be established for each rod.

RESULTS

The signal level read during the final reader anneal for a LiF:Mg,Ti rod was less than 0.4% of the signal that was read out after the next irradiation of the rods. For 90% of the LiF:Mg,Cu,P TLDs the signal level read during the final anneal was <1% of the signal that was read out after irradiation of the rods, for 5% (7) of the measurements it was 1-1.5%, and for 5% of the measurements it was 1.5-4.5%. These are likely overestimates since the residual signal after reading is almost always smaller than the signal measured during the reading. Each rod was normally given the same dose each time it was irradiated. In addition, all the highest “residual” signals were for TLDs for the same two doses in a single test. Subtracting these residual readings from the final readings makes little or no change in the ratio of the responses and no change in the conclusions.

Because the minimum dose delivered to the TLDs was 0.75 Gy, control TLDs were not used for all tests; the background doses were considered insignificant relative to the given doses due to the relatively short period between annealing and irradiation/reading. The average background level for LiF:Mg,Ti TLD rods that were not irradiated in test 3 was 1.5-6 mGy and for the LiF:Mg,Cu,P rods the background was 0.04-0.6 mGy, which are much less than the smallest dose of gamma rays.

The relative light output per unit dose of TLD rods in buttons for the various methods of cleaning are given in Table 2. These are the averages of the values for individual rods compared to their response at the same dose before being put in buttons, either directly or using ECCs and calibration TLD rods. The various cleaning processes had a 6% or less effect on the response of the LiF:Mg,Ti rods and only a 1-3% effect on the LiF:Mg,Cu,P rods. Of the three comparisons for the LiF:Mg,Ti rods, two showed a decrease in response of 2-3% and one a 6% increase in response. Some of the observed change in response might be reduced by preheating the TLD rods before reading them. This would eliminate the low-temperature peak in the glow curve, which might be more affected by heating effects during cleaning.

The TLD rod light outputs normalized to the sensitivity of the reader as a function of gamma-ray dose are shown in Fig. 2 for LiF:Mg,Ti and Fig. 3 for LiF:Mg,Cu,P. No ECCs have been applied. The lines are second order polynomial least squares fits to the individual rod data for the LiF:Mg,Ti because of its supralinear response at high doses and linear least squares fits for the LiF:Mg,Cu,P data. The points are the average responses for the rods at that dose and the error bars are one standard deviation of the mean. The closeness of the top three (Harshaw TLDs) and bottom three curves (Rexon TLDs) for LiF:Mg,Ti rods in Fig. 1 indicates the small absolute response variation between groups from the same batch and between TLDs that did or did not undergo cleaning. Fig. 2 indicates a 15-20% decrease in the response of the Rexon LiF:Mg,Cu,P rods for Test 3 before and after cleaning, however a similar shift (not shown) was observed in the calibration set, possibly due to a change in sensitivity caused by the annealing procedures used in this study. A difference in response of ~30% was observed between the Harshaw LiF:Mg,Cu,P rods in buttons and another group (cohort in Fig. 2) of rods from the same batch

As expected, the light output of the LiF:Mg,Cu,P rods is ~24 times greater than for the LiF:Mg,Ti rods given the same dose. For the LiF:Mg,Cu,P rods, the light emission per unit dose for the 8.23 Gy dose was significantly below what was obtained for the lower doses, so the data were discarded. The main glow peak was greatly widened and flattened at this dose. Even at 5-6 Gy there was some evidence of broadening of the main glow peak. The measurements made for 8.2 Gy (third test) used a 10:1 neutral density filter to avoid

saturation of the reader photomultiplier tube, so this significant change in response at 8 Gy appears to be due to some problem with the dose response of the TLD rods themselves.

The average response of the REXON Li:Mg,Ti rods was ~10-15% lower than for the Harshaw rods and the REXON LiF:Mg,Cu,P responses were ~5% lower than for the Harshaw rods. Both of these simply could be batch differences. The average weight of the REXON LiF:Mg,Ti rods was about 5% less than that for the Harshaw rods and the REXON LiF:Mg,Cu,P rods were about 15% lighter than the Harshaw rods, so the response difference is not likely to be due solely to mass.

DISCUSSION

Since the TLD rods were sealed in the plastic buttons, it seemed unlikely that the fluids used for cleaning would have an effect on the response of the rods to radiation. The concern was mainly for the effect that heating due to drying and to ironing or pressing might have.

The response of the TL materials can be modified by low temperature annealing. Glow peaks 4 & 5 for LiF:Mg,Ti may be affected by pre-irradiation annealing at 100° C for 30 minutes (Ben Sacher 1992). For LiF:Mg,Cu,P TL materials after annealing at 240° C, “annealing in the temperature region 140–180° C induces strong effects on the glow-curve structure” (Kitis 1996), greatly affecting the main glow peak.

Commercial cleaners seem to use a combination of presses and hand irons to iron clothing. In general, the cleaners used in this study were reluctant to allow temperature measurements of the press or iron and the workers did not know the temperature settings, if any, of the equipment used. One cleaner did allow measurements with a digital thermometer, which indicated a temperature of 110° C for the press, probably relying on steam to remove most of the wrinkles, and a temperature of 105° C for the hand iron, used for parts of the clothing that aren't suitable for the press. These temperatures are much lower than was measured for the iron used for the home cleaning (153° C) and may not be indicative of the temperatures used at all cleaning companies. General information on home irons indicates that they have a range of settings from about 135° C or less to as much as 230° C, depending on the type of clothing and the manufacturer of the iron. The home iron used in this study had a measure maximum temperature of ~185° C. The temperature to which these buttons were exposed and the length of time exposed are poorly controlled variables due to the differences in equipment, requirements of the clothing, and the difference in procedures used by different operators. High temperature exposure during ironing would normally be brief, less than one minute in most cases.

The TLD buttons were exposed to hot-air drying, which also has poorly controlled variables of temperature and length of exposure. This may have a greater effect on the TLD response. A survey of over 20 models of household driers for a report by the United States Consumer Product Safety Commission (USCPSC 2000) indicated that according to the user or service manuals the maximum temperature setting ranged from 125-350° F (~52-177° C). Clothes would normally be exposed up to an hour or more during drying and would not reach these temperatures when damp but could approach these temperatures if greatly over-dried.

Despite these uncertainties of temperature and length of exposure for drying and ironing, no significant effects to the average responses of the TLD rods were observed in the cleaning cycles used for this study. This may not be true for all cleaning conditions.

For convenience, the annealing for the TLD rods was performed using read cycles, as many as three after the initial reading depending on the residual signal. The recommended annealing procedure for LiF:Mg,Ti TL materials is 400° C for 1 hour followed by pre-

irradiation annealing at 75° C for 24 hours (Horowitz 1990), which is considerably more time-consuming. The standard annealing for LiF:Mg,Cu,P TL materials is 240° C for 10 minutes (Moscovitch 1999) or as many as eight read cycles to completely remove the residual signal. For mass production of buttons, these procedures would have to be followed closely to make the responses of the TLD rods as uniform as possible.

For widespread use, it is unlikely that the rods would be calibrated before being embedded in buttons. The criterion for Harshaw-Bicron (now Thermo Scientific) LiF:Ti,Mg TLD materials is that “the evaluated value for any one dosimeter in a batch shall not differ from the evaluated value for any other dosimeter in the batch by more than 30% for a dose equal to ten times the required detection threshold limit” (Velbeck 1999). Responses of rods in the same button or bare rods exposed to the same dose often had differences of 10-20%. The relative variation in response between rods was generally greater for the LiF:Mg,Cu,P rods. For clinically significant doses, the rods could be calibrated after the initial exposure to obtain a better dose estimate.

The purpose of the button TLD is to provide estimated doses for a relatively acute significant exposure (0.5 Gy or more) due to a radiation accident or act of terrorism and not for dose accumulated over a long period of time; therefore fading of the signal is probably not a major concern.

Most button dosimeters would likely be read within a few days after exposure and would probably not be laundered between the time of exposure and when the TLD rods are read. Testing would have to be done on the possible effects of heat from drying and ironing or pressing on a button that has been exposed for cases where the buttons were accidentally subjected to cleaning before being read.

The buttons used were designed to maximize the number of rods in the button and had holes at their edges for attaching the buttons to clothing. In practice, only one or two TLD rods would be placed in each button and the holes would be in the center of the button, as is standard. Each button would be marked with a bar code or serial number to identify it. Buttons could be attached to new garments by the manufacturers and also made available separately for attachment to existing garments. Only one button would be needed per garment, although the use of multiple buttons would provide back-up measurements. To minimize complications of button size, shape and color, a single type of button could be used and attached in an inconspicuous place, such as the back of the bottom of a shirt placket. A single button design would help increase the economy of scale. Alternatively, the button dosimeter could be manufactured in the form of a wafer that could be placed within garment labels. This would be particularly applicable to garments such as T-shirts and tank tops that normally do not have buttons.

TL dosimetry materials come in a number of shapes: discs, chips, rods and powder. For greatest economy, powder would be preferable since the cost of powder is about one tenth the cost of any of the shaped material, however powder would be more difficult to handle when opening the buttons and would have to be weighed before being read. Powder also has to be placed in the reader in some uniform manner since the surface area and thickness affects the reading. Ideally, with a button material that is reasonably transparent and could withstand at least 240° C the powder might be able to be read without being removed from the button.

After exposure, the buttons would be removed by the owner or at a triage center. Since a manual TLD reader system consists of a personal computer, a cylinder of nitrogen and the reader, which is a box $\sim 30 \times 30 \times 45 \text{ cm}^3$, a reading station easily could be set up in the field. For reading dosimeters sent in later, remote centers could be set up specially, contracts

could be set up with one or more radiation dosimetry providers, or a network could be set up consisting of facilities that are already equipped to read TLDs, e.g. radiation oncology centers. The remote centers would have systems that could be loaded with large numbers of TLDs that are read automatically.

The cost of the TLD button, mostly due to the cost of the TLDs, would have to be small relative to the cost of the garments for this system to be economically practical. The cost of the TL materials for the buttons would likely decrease due to economy of scale with demand for thousands of TLDs, making the system, especially for LiF:Mg,Ti TL materials, much less expensive.

CONCLUSIONS

Placement of LiF TLD rods in clothing buttons, either sewn on to garments or placed in labels, appears to be a potentially viable method of estimating the external gamma-ray dose due to an acute exposure, such as from a nuclear reactor accident, Radiological Dispersal Device, Radiological Exposure Device (RED), or Improvised Nuclear Device. Dose assessments made using buttons with TL materials could be used to triage the wearers for treatment, observation or further testing. Changes in response for rods subjected to various methods of cleaning were minimal (6% or less) for both TL materials. The saturation of the LiF:Mg,Cu,P rods above a dose of 5-6Gy will have to be investigated further before the material can be considered for large acute exposures.

Additional testing is necessary to fully assess the potential influence of cleaning processes, especially ironing or pressing, on the response of the TL materials.

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Fig. 1. Photograph of the TLD “buttons”. L-R: top disc; bottom disc with slots for three TLD rods; bottom disc with TLD rods; bottom disc with TLD rods and holes for attaching the button to a shirt.

Fig. 2.

Light readings for LiF:Mg,Ti rods without application of ECCs before and after two methods of cleaning and for the calibration set when irradiated with the buttons. The data was fitted to second order polynomials due to the supralinear response of LiF:Ti,Mg at high doses. The first three curves listed in the legend are for Test 2 (Harshaw) and the second three curves are for Test 3 (Rexon). Error bars are the standard deviations of the readings.

Fig. 3.

Light readings for LiF:Mg,Cu,P rods without application of ECCs before and after two methods of cleaning, for the calibration set when irradiated with the buttons and for a group of rods from the same batch (cohort). The lines are linear least squares fits to the data. The first two curves listed in the legend are for Test 2 (Harshaw) and the bottom three curves are for Test 3 (Rexon). Error bars are the standard deviations of the readings.

Time-temperature profiles used for annealing and reading TLD rods using Harshaw Model 3500 planchet TLD reader (Thermo Electron, 2002; Thermo Electron, 2006).

Table 1

TLD type (Dose type)	Preheat			Acquisition			Anneal	
	Temp. (°C)	Time (s)	Temp. (°C)	Time (s)	Rate (°C s ⁻¹)	Temp. (°C)	Time (s)	
LiF:Mg,Ti (low)	50	0	260	26 2/3	10	260	0	
LiF:Mg,Ti (high)	50	0	300	33 1/3	10	300	0	
LiF:Mg,Cu,P	50	0	240	36 2/3	10	240	10	

Relative light output from TLD rods irradiated in buttons after various forms of cleaning. Uncertainties are ± 1 standard error of the mean

Table 2

Type of cleaning	TLD Supplier	Doses (Gy)	Compared directly to the same rods (no ECCs)				Compared to the same rods using ECCs	
			LiF:Mg,Ti	Li:Mg,Cu,P	LiF:Mg,Ti	Li:Mg,Cu,P	LiF:Mg,Ti	Li:Mg,Cu,P
Dry cleaning (Test 1)	Harshaw	1.5	1.063 $\pm$ 0.024	----	----	----	----	----
Home laundrying (Test 2)	Harshaw	0.75-6.0	----	0.992 $\pm$ 0.062	0.971 $\pm$ 0.020	----	----	----
Commercial laundrying (Test 3)	Rexon	0.75-8.2	----	----	0.984 $\pm$ 0.021	0.973 $\pm$ 0.030	----	----