Characterization of the Effect of $\gamma$ -Radiation Absorbed Dose on the Electrical Conductivity of Aqueous Urea Solutions in the Presence of Silica Gel : A Preliminary Dosimetric Study

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$\gamma$-radiolysis of 6M-aqueous urea solutions, in the presence of silica gel (SG) significantly increases the values of electrical conductivity (RIC) of the solutions, relative to that in the absence of SG. Some dosimetric characteristics were studied, such as the effects of the absorbed dose, absorbed dose rate, and post-irradiation storage time on the RIC of the irradiated 6M-aqueous urea solutions in the presence of SG (>0.16mm - 0.2mm). A linear relationship between the RIC of the solutions and the absorbed dose ($R^2 = 0.998$) was observed from 0.240 to 17.670 kGy. The uncertainty in the dose-response function was found to be in the range 0.040-0.320 at 95% confidence level. As the dose rate increases, the RIC decreases in the range of 0.298 -1.19 kGy/h. The stability of the response (RIC) of the irradiated solutions is acceptable during post irradiation storage (14 days at 0°C) where the response (RIC) decreased at the end of the storage period, 0.49 -2.8%. The effective atomic number of this solution (7.08) was found to be compatible with biological tissues. The results show that the studied system can be considered a promising dosimeter for low dose applications in the range 0.240 to 17.670 kGy, such as medicine and food irradiation.

Keywords: Urea, Electrical conductivity, $\gamma$ - Radiation, Silica gel

INTRODUCTION

$\gamma$-radiation has emerged in the technology of medical sterilization [1, 2] and food irradiation [3-9]. In the medical sterilization field, whole blood and freeze-dried plasma were irradiated in the range 2.5-25 kGy [1,2]. In the food irradiation field, it was found that, $\gamma$-radiolysis, in the range of 3-10 kGy, reduces the total aerobic viable cell counts in highly contaminated spices and dry herbs [3]. Irradiation at a dose of 300 Gy for quarantine treatment against all insects other than fruit flies of the family Tephritidae is currently recommended [4]. Generally, insect disinfestation of fruits can be controlled by irradiation in the dose range 0.1 -1 kGy [5]. Inactivation of some pathogenic parasites of public health significance, such as tapeworm and trichina in meat, can be achieved at doses in the range 0.3–1 kGy [6]. In order to sterilize ground beef, it must be subjected to ionizing radiation to receive a dosage that is no less than 1.35 kGy and no more than 3.0 kGy [7]. Moreover, irradiation at doses up to 1.5 kGy can be used to extend shelf life of fresh-cut onions [8]. Radiation doses of 1.5 kGy, using photon sources such as X-rays or gamma rays, have been proposed to be sufficient to render eggs free of pathogenic microorganisms such as Salmonella, Listeria monocytogenes, and Campylobacter jejuni [9]. The radiation-induced conductivity (RIC) of urea aqueous solutions was previously studied by the
CHARACTERIZATION OF THE EFFECT OF $\gamma$-RADIATION …

The author of the present work\cite{10,11}, where the author attributed the RIC to the formation of urea – H$_2$O$_2$ adduct during $\gamma$- radiolysis of urea aqueous solutions \cite{12a}. It should be mentioned that significant RIC values were reported for irradiated 6M urea aqueous solutions\cite{10,11}. Silica gel has been found to exhibit high catalytic activity under ionizing irradiation \cite{12b}. So, the author aimed to evaluate the suitability of 6M aqueous urea solution in the presence of silica gel (>0.16mm - 0.2mm) system as a dosimeter for medical and foods $\gamma$ radiation sterilization. Therefore, in the current work some dosimetric characteristics of 6M urea aqueous solution/ silica gel (>0.16mm - 0.2mm) system will be discussed. Therefore, the absorbed radiation dose, and the dose rate effects on the RIC (response) of the solutions, as well as, the post – irradiation stability of the response were studied.

Experimental
The Co-60 $\gamma$ -rays source of NCCRT (India Gamma chamber 4000 A.) was used for the irradiation of the samples. The dose rate was 1.19 kGy/h which was determined by Alanine dosimeter. All the irradiations were carried out at ambient temperature. Urea (98%), have been obtained from Sigma-Aldrich and used as received. Urea was dissolved in water to a stock concentration of 6 M. Distilled water was prepared by distillation from alkaline permanganate and from sulfuric acid followed by a third distillation (Its specific conductivity does not exceed 0.1 $\mu$S.cm$^{-1}$). This pure water is used in the preparation of samples and for final rinsing of all glass ware. Four samples were irradiated for each dose. Each sample is composed of 2g of SG (>0.16mm - 0.2mm) and 20 ml 6M urea aqueous solution in a 50 ml bottle with stopper. The electrical conductivity was measured at room temperature using a Kent EIL5007 conductivity meter (Kent industrial measurements—Brown Boveri). The conductivity meter, with a cell constant of 1 cm$^{-1}$, was calibrated using NaCl standard solutions \cite{13}. The electrical conductivity of the irradiated solutions was measured immediately after irradiation. Storage of the irradiated samples was carried out in a freezer at about 0 °C.

Results and Discussion
The effect of $\gamma$ -radiation on the RIC of urea aqueous solution has been already investigated \cite{10,11}, where the author attributed the observed RIC to the formation of urea – H$_2$O$_2$ adduct during $\gamma$- radiolysis of urea aqueous solutions \cite{12}. The author observed that the irradiation of 6M urea aqueous solutions solution in the presence of SG (>0.16mm - 0.2mm), at 4.2 and 19.1 kGy, increases RIC up to 18.8 and 89.8%, relative to RIC in the absence of SG, respectively (Table 1). Therefore, in order to evaluate the possibility of using 6M aqueous urea solutions / SG (>0.16mm - 0.2mm) system as a dosimeter, some dosimetric characteristics should be studied. These dosimetric characteristics include the effects of the absorbed dose, absorbed dose rate, stability of the response (RIC) over a storage period, and the uncertainty of the measurements of the dose response function.

Dose response
Figure (1) shows the effect of the absorbed radiation dose (kGy) on RIC ($\mu$S.cm$^{-1}$) of 6M aqueous urea solutions in the presence of silica gel (>0.16mm - 0.2mm). In the studied dose range (0.240 to 17.670) four samples were irradiated, per dose, using Co-60 $\gamma$-radiation. The RIC of the samples increases with increasing absorbed dose linearly up to the maximum of the dose range ($r^2=0.9987$). The linear dose response function with a high $r^2$ value confirms the validity of the system for radiation dose measurements.

Dose rate effect
In order to study the effect of the absorbed dose rate on the RIC of the studied system, 50 and 75% attenuators were used during radiolysis. Figure (2) shows that the RIC increases as the absorbed dose rate decreases with a slope; -1.5611 $\mu$S.cm$^{-1}$/kGy.h$^{-1}$ in the dose rate range 0.298 -1.19 kGy/h. Therefore, the sensitivity of the current study system is at the maximum value at 0.289 kGy/h (75% attenuation). Hence, it is expected that this system can be successfully used at low dose rates. Therefore, it can be concluded that the formation of H$_2$O$_2$ and urea – H$_2$O$_2$ adduct \cite{12}, which are related to the RIC, happened mainly in the bulk of the medium, rather than the spurs.

Effective atomic number
The effective atomic number is closely related to the electron density, expressed in number of electrons per unit mass. Compton scattering is the main interaction process for low- and medium-Z elements in a certain energy range, typically between 0.05 and 5 MeV.

Table (1): Effect of silica gel (>0.16mm - 0.2mm) on the RIC of 6.0M urea aqueous solutions

<table>
<thead>
<tr>
<th>Absorbed dose (kGy)</th>
<th>RIC in the absence of SG</th>
<th>RIC in the presence of SG</th>
<th>Δ RIC</th>
<th>Δ RIC%</th>
</tr>
</thead>
<tbody>
<tr>
<td>4.2</td>
<td>10.23</td>
<td>12.15</td>
<td>1.92</td>
<td>18.8</td>
</tr>
<tr>
<td>19.1</td>
<td>15.71</td>
<td>29.81</td>
<td>14.1</td>
<td>89.8</td>
</tr>
</tbody>
</table>

In this energy region, incoherent or Compton scattering accounts for practically all photon interactions.

In this work, the experiment has been performed with two γ-quanta with energies of 1.173 and 1.332 MeV, released by Co-60 source, where Compton scattering is the main interaction process. It is known that the dosimeter should be made from biological tissue equivalent materials, when it is needed in medical and food sterilization fields.

For the Compton scattering, effective atomic numbers ($Z_{\text{Compt}}$) of 6M aqueous urea solution have been determined to be 7.08 according to the following equation [14]:

$$Z_{\text{Compt}} = \frac{\sum n_i A_i Z_i}{\sum n_i A_i}, \quad i = 1,...,N$$

Where, “$A$” is the mass number, “$Z$” is the atomic number, and “$n$” is the number of atoms for each element (i) in the solution. Consequently, $Z_{\text{Compt}}$ of 6M aqueous urea solution seems to match those of soft biological tissues [15-17].

**Post-Irradiation Stability of the response**

The stability of the response (RIC) of 6M aqueous urea solution in the presence of silica gel (>0.16mm - 0.2mm ps1) was studied after irradiation at different “post-irradiation time periods”. Therefore, the irradiated samples were stored under 0°C storage conditions over a period of 14 days. The RIC of the irradiated samples was measured at different time intervals (Fig.3) during the post irradiation storage period. Fig.(3) shows that at the end of the storage period, a limited decrease in the response (RIC) was observed (0.49-2.8%). So, it can be seen that the studied system has a good stability of the response (RIC).

**Uncertainties**

When a set of several repeated readings has been taken, for a Type A estimate of uncertainty, the mean, x, and estimated standard deviation, $\sigma$, should be calculated for the set. The estimated standard uncertainty, $u$, of the mean is calculated from the following equation:

$$u = \frac{\sigma}{\sqrt{n}}$$

where $n$ is the number of measurements in the set.

The standard deviation of the mean represents the uncertainty in the mean. In order to evaluate the batch variability, every dose point, was averaged and its coefficient of variation (at one standard deviation, 1σ) was calculated. From the data obtained (Table.2), it was found that the coefficient of variation (CV%) ranges from 0.265-3.340%, reflecting the precession and the reproducibility of the measurements of the studied system. The type “A” uncertainties (at two standard deviation, 2σ) of the RIC measurements over the dose range of 0.240-17.670 kGy, corresponding to 95%-confidence level, was found to be in the range 0.040-0.320.
CHARACTERIZATION OF THE EFFECT OF $\gamma$-RADIATION …..

Fig. (3): Change in the RIC ($\mu$S.cm$^{-1}$) after irradiation of 6M aqueous urea solutions in the presence of silica gel (>0.16mm - 0.2mm), stored at 0 °C, as a function of storage time (days), ●0.240, ○ 1.260, ▼ 3.130, △ 7.960, ■11.710, □ 14.900, ♦ 17.670kGy

Table (2): Determination of the Standard uncertainty of the mean, Uncertainty type A (95%), and Coefficient of Variation (CV%) for the readout of RIC values at different absorbed doses (4 samples for each dose) for 6M aqueous urea solutions / SG (>0.16mm - 0.2mm) system

<table>
<thead>
<tr>
<th>Absorbed dose (kGy)</th>
<th>Standard uncertainty of the mean (1σ)</th>
<th>Uncertainty type A (95%) (2σ)</th>
<th>CV%</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.240</td>
<td>0.087</td>
<td>0.174</td>
<td>2.400</td>
</tr>
<tr>
<td>1.260</td>
<td>0.139</td>
<td>0.278</td>
<td>3.340</td>
</tr>
<tr>
<td>3.130</td>
<td>0.041</td>
<td>0.082</td>
<td>0.804</td>
</tr>
<tr>
<td>7.960</td>
<td>0.020</td>
<td>0.040</td>
<td>0.265</td>
</tr>
<tr>
<td>11.710</td>
<td>0.090</td>
<td>0.180</td>
<td>0.896</td>
</tr>
<tr>
<td>14.900</td>
<td>0.160</td>
<td>0.320</td>
<td>1.342</td>
</tr>
<tr>
<td>17.670</td>
<td>0.061</td>
<td>0.122</td>
<td>0.450</td>
</tr>
</tbody>
</table>

Conclusion
RIC of $\gamma$-irradiated 6M aqueous urea solutions / SG (>0.16mm - 0.2mm) system, as a function of the absorbed dose, can be used to innovate a novel dosimeter. Where, the RIC of this system significantly increases as the absorbed dose increases with an excellent linearity up to 17.670kGy. The RIC of the irradiated samples (0.240-17.670kGy) has also shown good post-irradiation stability for 2 weeks, when the samples were stored at 0°C where the fading % was found to be in the range 0.49-2.8% at the end of the storage period. Moreover, there is a good compatibility of this system with the biological tissues, which was determined via calculation of the effective atomic number of the studied system (7.08). The uncertainty and the coefficient of variation of the readout values of the response

(RIC) curve was also determined, which reflects a good precision and reproducibility of the measurements, in the range 0.240-17.670 kGy. The dose response range of the presented system covers the absorbed dose range of interest in medical sterilization and food irradiation. The advantages of this system include the stability of its linear response, its reproducibility, small size, low cost, ease of use, good sensitivity, and ease of preparation. Moreover, the studied system covers a wider dose range compared to that of Fricke dosimetry, which is unsuitable to measure doses above 400Gy.

References