

## Physicochemical Study of Some Gamma-Irradiated pH-Indicator

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**A**QUEOUS SOLUTIONS of pH indicators congo red (CR) and quinaldine red (QR) dyes were titrated by different concentrations of (H<sup>+</sup>) resulting from irradiation decomposition of chloral hydrate (CH) solution for possible use as a monitoring device (label) for gamma radiation. The titrated solutions were readout spectrophotometrically. The useful measuring range was found to be of 20 -100 kGy. Further calculation such as rate of reaction (r) and have been assigned. kinetic constant (k)

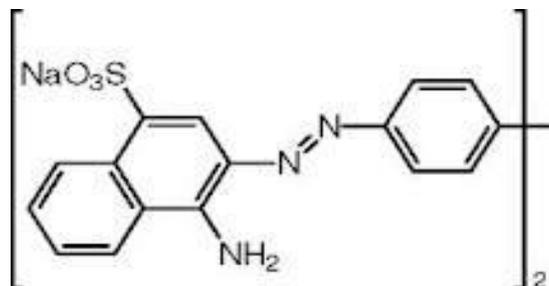
**Keywords:** Congo red, quinaldine red, chloral hydrate, poly(vinyl alcohol), gamma ray dosimeter.

Many solutions undergo  $\gamma$ -irradiation for effective sterilization such as medical solutions, cosmetic lotions, ointments and others need dose detection. Using liquid dosimeters or labels the absorbed doses were detected without any absorption extension coefficient corrections. Many radiation sensitive indicators for qualitative dose measurements have been manufactured to distinguish processed irradiation units from unprocessed irradiation units (Abdel-Rehim and Abdel-fattah, 1993 and Abdel-Rehim *et al.*, 1996). These indicators may be labels, papers and inks which undergo a visual colour change when exposed to ionizing radiation (Abdel-Rehim *et al.*, 1985). These indicators based mainly on radiochromic dye (Akhava *et al.*, 2002, Butson *et al.*, 2001 and Kovács *et al.*, 2000), pH-indicator dye (Abdel-Fattah and El-Kelany, 1998, Abdel-fattah *et al.*, 2002, Ogawa, 1995 and Sidney *et al.*, 1990), They are not used for quantitative dose measurements while label dosimeters may be used for quantitative dose measurements during radiation processing.

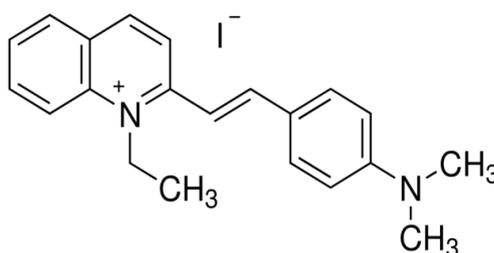
In the present work, solutions of CR and QR were titrated by irradiated CH have different doses were studied for use in high-dose dosimetry through back titration followed by colour measurements using spectrophotometer.

**Materials**

Congo red dye,  $C_{32}H_{24}N_6O_6S_2$ ,  $M = 652.7$  mol/l and quinaldine red,  $C_{21}H_{23}IN_2$ ,  $M = 430.3$  mol/l (Aldrich, analytical grade) and chloral hydrate (Merck Germany) are shown in scheme 1,2 respectively.



**Scheme 1. Congo red.**



**Scheme 2. Quinaldine red.**

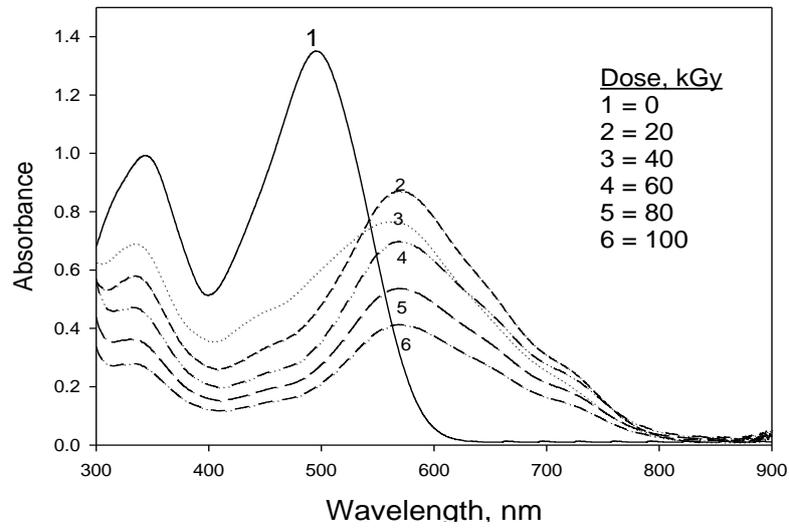
**Apparatus**

Irradiation was carried with a  $^{60}Co$  gamma chamber 4000 Å (product of India). The absorbed dose rate was 5 kGy/ h. The absorption spectra of un-irradiated and irradiated solutions of both dyes were measured using a UV4-visible spectrophotometer. The irradiation of chloral hydrate done by (using several ampoules of CH 5 ml each) exposed by different doses. After irradiation, proceeds titration (0.1 ml of irradiated CH solution [1N] with 2 ml of dye solution).

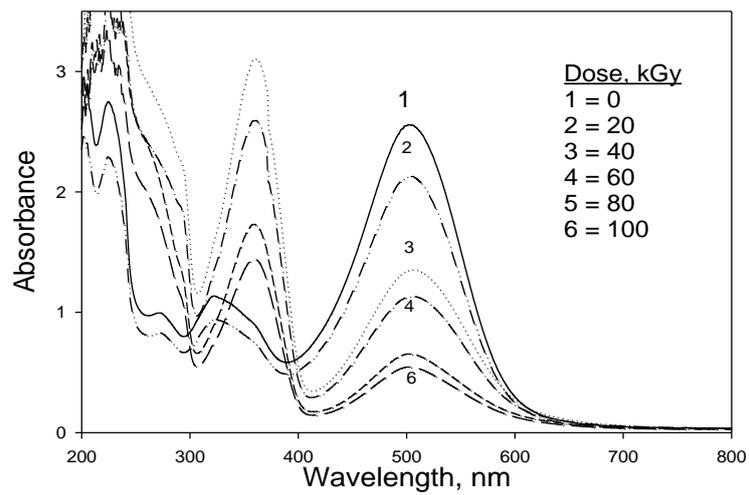
**Results and Discussion****Absorption spectra**

Depending on avoid exposure of both CR and QR pH indicator proceed back titration with irradiated chloral hydrate (1 N) applying different doses of  
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$\gamma$ -irradiation (20, 40, 60, 80, 100 kGy) and using the irradiated resulting  $[H^+]$  concentrations in titration of 0.35 phr concentration for both dyes.



**Fig. 1.** The absorption spectra of CR-CH aqueous solution unirradiated and irradiated to different absorbed doses.

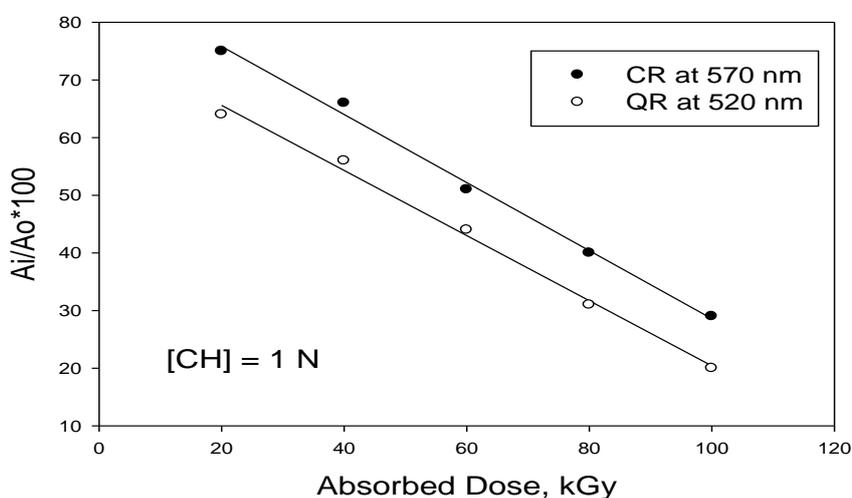


**Fig. 2.** The absorption spectra of QR-CH aqueous solution unirradiated and irradiated to different absorbed doses.

Upon irradiation the amplitude of the peak was found to be decreases gradually with increase of applying gamma rays on CH solutions, starting from 20 kGy to 100 kGy as shown in Fig. 1-2. This is due to the consequent lowering of  $H^+$  concentration caused by the HCl generated from radiolysis of chloral hydrate. Both dyes reacts with consequent  $H^+$  increase according to absorbed dose leads to colour changes and decrease in peak intensity at 570 nm (red-blue) for CR and 520 nm (red-colourless) for QR.

### Response curves

The response curves of CR or QR/CH solutions at different doses are shown in Fig. 3. The response were established in terms of change in relative percentage of absorbance  $[(A_i/A_o) \times 100]$ , (where  $A_o$  and  $A_i$  are the relative absorbencies of unirradiated solutions respectively) at 570, 520 nm against the absorbed dose. It can be noticed that both responses have the same trend in the range of 20-100 kGy with different slope.



**Fig. 3.** Change of response of two dye-CH solutions  $[(A_i/A_o) \times 100]$  as a function of absorbed dose.

### Calculation of concentration of $H^+$

The effect of  $H^+$  at different pH solutions of CR and QR at a dose range from 20-100 kGy has been studied. The change in pH as a function of absorbed dose can conclude that the dye in acidic form is increase with decrease pH

(acid-base titration). This phenomenon is a new development for the presence excess of  $H^+$  in the medium. Fig. 4. shows the total amount of acid formed in solutions of both dyes containing chloral hydrate as a function of absorbed dose. It can be seen that the amount of  $H^+$  increases gradually with increase of absorbed dose.

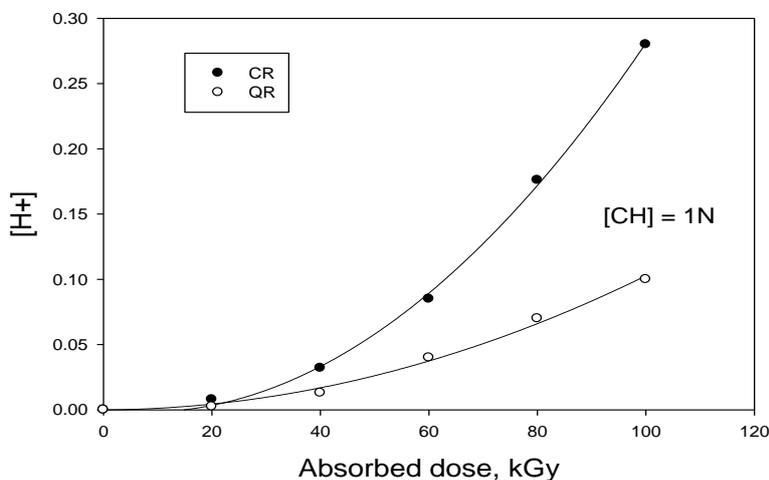
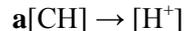


Fig. 4. Variation of  $H^+$  in two-dye solutions as a function of absorbed dose.

#### *Kinetics of $H^+$ formation*

In these solutions CR and QR containing CH, the  $H^+$  is formed due to the irradiation of CH. This reaction may be considered as follows:



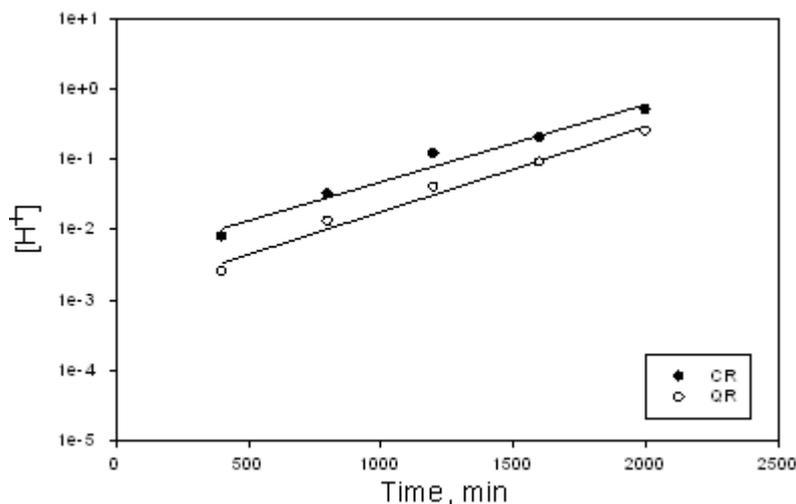
where,  $H^+$  is the concentration of radiation-formed hydrogen ions, the rate law of this reaction may be written as follows:

$$d[H^+] / dt = k [H^+]^a$$

$$\text{Or: Rate of reaction} = k [H^+]^a$$

where, **a** is the order of the reaction (relationship between the concentration of  $H^+$  to the rate of reaction) and  $d[H^+] / dt$  is the rate of reaction. The order **a** can be determined by using the initial slopes method (Atkins 1982). Plotting change

in concentration of  $H^+$  (on log-log scale) against various times gives the rate and order of reaction from the slope show in Fig. 5. Straight lines were obtained, slopes are 0.531 and 0.522 for hydrogen concentration which detected by dye solution of CR and QR, respectively, the order of these reactions are  $\frac{1}{2}$ .



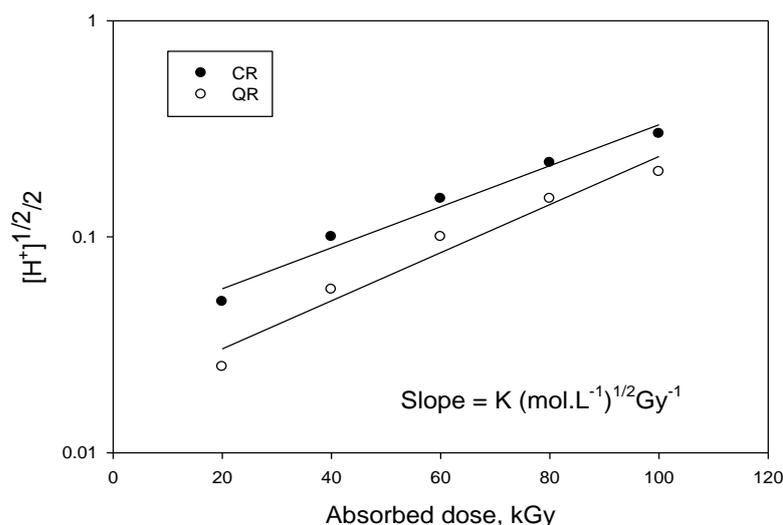
**Fig. 5.** Variation of  $[H^+]$  in aqueous solutions of (CR and QR-CH) as a function of time.

Accordingly, the rate law of  $H^+$  in this acid-base titration reaction, may be expressed as follows:

$$d[H^+] / dt = k [H^+]^{1/2}$$

$$\text{Or: } d[H^+] / dD = k [H^+]^{1/2}$$

Fig. 6. shows the increase of concentration of  $[H^+]^{1/2} / 2$ , as a function of absorbed dose. Straight lines were obtained in accordance with half-order kinetics (El-Kelany, 1997). The kinetic constant,  $k$ , is obtained as the slope of straight lines mentioned above. The values of  $k$  for CR and QR solutions are  $9.27 \times 10^{-3} (\text{mol.L}^{-1})^{1/2} \text{Gy}^{-1}$  and  $9.27 \times 10^{-3} (\text{mol.L}^{-1})^{1/2} \text{Gy}^{-1}$ , respectively.



**Fig. 6. Increase of concentration of acid in CR and QR solutions containing CH various the absorbed doses.**

### Conclusion

This approach for preparing new radiation sensitive indicators by titrating acid-sensitive dyes with irradiated chloral hydrate may be useful. These solutions can be used qualitatively as routine indicators in the 20 – 100 kGy range because they have stable change in colour for CR from red to blue at 570 nm and for QR from red to colourless' at 520 nm depending on irradiated chloral hydrate H<sup>+</sup>. Using the phenomenon of generated HCl from CH under irradiation, hydrogen ion concentrations as a function of irradiated time used to calculate kinetic constant of reaction ( $\approx 9.3 \times 10^{-3} (\text{mol.L}^{-1})^{1/2}\text{Gy}^{-1}$ ).

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**التشعيع الجامى لكاشف إشعاعى حساس**

مشيرة الكيلانى و سيف الدين إبراهيم

قسم الوقاية و الجرعات الإشعاعية ، المركز القومى لبحوث و تكنولوجيا الإشعاع  
، ص. ب. ٢٩ مدينة نصر ، مصر.

هذا البحث تم تحضير محاليل مائية من صبغتي الكونجو الحمراء و الكينولدين الحمراء و معايرتهما بمحلول الكلورال هيدرات المشع عند جرعات مختلفة وذلك بهدف استخدامها كمقياس لأشعة جاما. وقد تم تقدير حساسية هذه المحاليل بواسطة جهاز الأطياف الضوئية. واتضح أن المدى الإشعاعى المناسب لهذه المحاليل يتراوح بين ٢٠-١٠٠ كيلو جراى معتمدا على تركيز أيون الهيدروجين المتكون نتيجة التشعيع لمحلول الكلورال هيدرات. وتم حساب معدل ثابت حركة التفاعل.