



GAMMA RADIOLYTIC DECOMPOSITION OF ACIDIC STANNOUS CHLORIDE SYSTEMS

KALE S.M.^{1,2}, DAS SARMA I.B.³, PARWATE D.V.^{1*} AND BATRA R.J.⁴

¹PGTD Chemistry, Rashtrasant Tukadoji Maharaj Nagpur University, Nagpur, MS, India.

²New Arts, Commerce and Science College, Parner, Ahmednagar, MS, India.

³Jhulelal Institute of Technology, Nagpur, MS, India.

⁴Ramdeobaba College of Engineering and Management, Nagpur, MS, India.

*Corresponding Author: Email- dilip58parwate@gmail.com

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Abstract- The investigations involving the effect of ionization on the yields of radiation- chemical reaction in aqueous solutions suggest radical diffusion model for various radiation-induced chemical reactions in aqueous media. Gamma radiolysis of acidic stannous chloride solution at various absorbed doses and in presence of additives has been investigated. G(-Sn²⁺) is higher at the lower gamma dose. At the higher doses beyond 10 kGy the G values remain constant. The additives enhance the radiolytic transformation of stannous by energy transfer process. Presumably the additives exhibit sensitization effect. Efficiency of sensitization depends on the nature of the additives, concentration in the admixture and the absorbed dose. The additives were used in high concentration in contrast to the stannous chloride solutions. Thus, protection might be due to the shielding effect rather than scavenging.

Keywords- G-value, stannous chloride, additives, potassium aluminium sulphate, sodium aluminate, methacrylic acid

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Introduction

Ionizing radiations interact with matter and can cause formation as well as destruction of species. Radiolysis of aqueous systems has evoked much interest in radiolytic products of water which may then interact with solute causing complex reactions. Radiation chemical reaction taking place in water and in aqueous solutions have been the subject of numerous investigations [1-3].

The investigations involving the effect of ionization on the yields of radiation- chemical reaction in aqueous solutions suggest radical diffusion model for various radiation-induced chemical reactions in aqueous media. According to this model, radiation-induced chemical reactions in aqueous solution are initiated by radicals generated from water as a result of energy deposition [4-6].

In the present study, gamma radiolysis of stannous chloride in aqueous hydrochloric acid at various absorbed doses and in presence of additives has been investigated. Various additives used were methacrylic acid (MA), potassium aluminium sulphate (PAS) and sodium aluminate (SA).

Experimental

All the solutions were prepared in doubly distilled water with A.R./ G.R. grade chemicals. The stock solutions of the additives and stannous chloride were used to prepare the working standard of required concentrations. The stannous chloride solutions were freshly prepared to avoid atmospheric oxidation.

Stoppered corning glass tubes made of B24 joints were used as irradiation cells to minimize solvent evaporation. The solutions were irradiated in Gamma Chamber 900 (GC-900) housed in Nuclear Chemistry laboratory, Department of Chemistry, Rashtrasant Tukadoji Maharaj Nagpur University, Nagpur. The dose rate of the Co-60 source during the course of work was 80 krad/hr.

About 10-15 mL of the solutions was irradiated for different time periods. Post irradiation chemical changes were determined spectrophotometrically using Shimadzu UV-240/Elico SL-150 UV-Visible spectrophotometer.

Results and Discussion

Gamma irradiation of deaerated solution of the acidic stannous chloride was studied. The effects of dose, initial Sn^{2+} concentration and in presence and absence of dissolved O_2 on the yield were investigated. The gamma radiolytic transformations of stannous chloride was carried out over a dose range of 0.2 to 15 kGy with and without the additives at varying concentrations at the dose rate 0.84 kGy hr^{-1} .

The $G(-\text{Sn}^{2+})$ values were found to be higher at lower gamma doses and gradually decreases with increase in the dose absorbed and then remain constant beyond 10kGy. The variation of concentration of Sn^{2+} formed and $G(-\text{Sn}^{2+})$ values against absorbed dose, of stannous chloride with additives of different concentration have been plotted and shown in Fig.1a and 1b respectively.

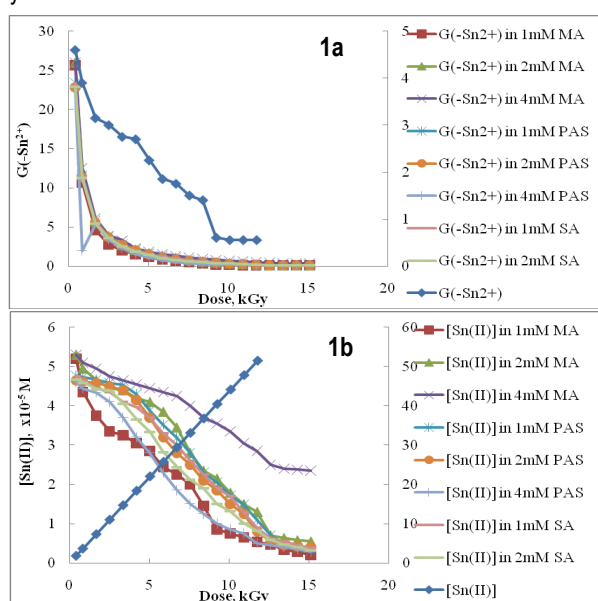


Fig. 1- Graph depicting (a) Variation of stannous ion concentration against dose absorbed in presence of various additives (b) Variation of G-values against dose absorbed in presence of various additives

It has been observed that the $G(-\text{Sn}^{2+})$ values i.e. the amount of stannous ion disappeared (converted in Sn^{4+}) is higher at lower gamma doses and yield gradually decreases as the dose is increased, showing less formation of Sn^{4+} . At the higher doses beyond 15kGy the values are found to remain constant (not shown in figures as there is no change beyond 15kGy). $G(-\text{Sn}^{2+})$ values are found to vary when irradiated in presence of additives and the increase is found to be dependent on the quantity of the additive used. For the dose of 0.42 kGy , $G(-\text{Sn}^{2+})$ in presence of PAS, SA and MA are found to be 22.59, 22.68 and 26.03 respectively. Also it has been observed that the G-values increase initially, due to the additive but decrease rapidly when exposed to higher dose but around 10 kGy remain constant. Thus in MA, when concentration of 1mM, 2mM and 4mM are used $G(-\text{Sn}^{2+})$ is found to change from 25.63 to 26.03. The surfactant MA is found to have very little effect on G-values. However with increase in dose all of them show decreasing trends.

In case of PAS and SA 1mM concentration has shown the highest

value. The higher $G(-\text{Sn}^{2+})$ values of Sn^{2+} in the case of additives are due to the energy absorbed in the bulk of the additives from where it is effectively transferred to the intimately close Sn^{2+} resulting in oxidation. The transfer of the energy leads to increase in G-values. Similarly, the opposite must be decreasing the G-values which happens in the case of MA.

The systems purged with nitrogen showed better protection against oxidation as was expected due to comparatively less numbers of the oxygen containing species. It is clear that with the absence of oxygen the possibility of the formation of oxidizing radicals such as O_2^- , O^- and so on are eliminated thereby reducing the extent of decomposition which would have otherwise occurred. On the addition of additives, in general, energy will be shared by both the constituents proportionately. However, the energy absorbed by the additives may be transferred to the stannous either instantaneously or at a later stage.

Energy transfer by inert system is well known phenomenon. It is a general agreement that G-value is enhanced by the additives such as MA, PAS and SA. Sagert - Robinson and Wong - Willard have explained the energy transfer process in terms of electron migration, whereas Rabe et al and Khare-Johnson have suggested excitation transfer mechanism [7-11]. It therefore appears that minimum $\sim 4 \text{ eV}$ is required to cause dissociation of stannic ion. It has been shown that matrix materials having an energy gap $< 4 \text{ eV}$ is ineffective in causing the energy transfer.

Conclusion

On the basis of the present study it is concluded that $G(-\text{Sn}^{2+})$ is higher at the lower gamma dose. At the higher doses beyond 10 kGy the G value remain constant. The additives enhance the radiolytic transformation of stannous by energy transfer process. Presumably the additives exhibit sensitization effect. Efficiency of sensitization depends on the nature of the additives, concentration in the admixture and the absorbed dose. The additives were used in high concentration in contrast to the stannous chloride solutions. Thus protection might be due to the shielding effect rather than scavenging.

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