CHAPTER - VI

DISCUSSION

Thermoluminescence (TL), the emission of light on heating a solid to a temperature of incandescence, has been known for at least three centuries. It has also long been recognised that this phenomenon is thermally stimulated release of stored energy deposited in the solid by prior exposure to some exciting agent such as X-rays, gamma rays or corpuscular radiations. The particular characteristic storage of energy and release of it on heating, whenever desired, made TL very useful and popular in different fields. The application potential of TL has been very well realized during last two decades. Its applications are in many diverse fields like Radiation dosimetry, Archaeology, Geology, Forensic science, Impurity analysis, Solid state physics, Radiation damage in solids, Catalyst evaluation etc.[4-9, 23-25]. Since the present work involves the application of TL of NaCl:Ba and KCl:Ba in radiation dosimetry, a brief account of the present understanding of TL in pure and divalent cation doped alkali halides and their utilities in radiation dosimetry are reviewed separately.

PART - I : THERMOLUMINESCENCE

(a) Short review of present state of understanding of thermoluminescence in alkali halides.

The first observation of TL recorded in literature can be dated as far back as 1633[14]. Robert Boyle reported to the Royal Society, London, on observing a storage glimmering light when he warmed a diamond in the dark. However, the last thirty years have witnessed a rapid and an extensive experimental and theoretical investigation of TL. By far the most sensitive TL materials are dielectric solids. However, it is wide spread phenomenon and exhibited by a host of minerals, in organic crystals, glasses and ceramics, organic compounds such as polymers including polythene and teflon, certain biochemicals and biological materials etc..

Natural as well as artificial TL of pure and impurity doped solids in different physical and/or chemical conditions have been a subject of investigation for a long time. The alkali halides have been selected as ideal host materials for experimental and theoretical studies, on account of the following features.

(i) Simple cubic structure.

(ii) Ionic nature of bonding.

(iii) Large binding energy ($\sim$ 200 K Cal/mole).

(iv) High melting point ($\sim$ 1000°C) which provides vast range of temperature over which the phenomenon can be studied, and

(v) Large electronic band gap ($\sim$10 eV) which offers a wide range of optical transperancy to study the effects of impurities, vacancies and other crystalline defects.

Many luminescene workers have put systematic efforts in examining the TL behaviours of pure and divalent cation impurities doped alkali halides.[25-28]. It has been found that peak location in the glow curve/emission spectrum is influenced by factors such as heating rate, extent of initial excitation (e.g. radiation dose) etc. It was found that the TL characteristics of any material could be specified by the TL glow curve at a given dose, heating rate and the TL emission spectrum at a given temperature. Further, it has been investigated that the phenomenon of TL is sensitive to variety of factors, such as

(i) Impurities in the material.

(ii) Pre and post mechanical and/or thermal irradiation treatments to the material.

(iii) LET of radiation used for excitation etc.

It has been investigated that TL process involves ionic as well as electronic rearrangements and their contributions are difficult to separate. In general, it was concluded that TL is the result of electron hole recombination process. However, the site of recombination could not be specified. Hence, the suggested models for the TL mechanisms have been different. However, the physical significance of the observed peaks in TL glow curve exhibited by any material is usually explained as follows. When the material is excited equal number of electrons and holes are "liberated" within it and some of them get "trapped" at certain "centres" in the material i.e. energy is "stored" in the material. These storage sites are called the electron or hole trap respectively. The physical configuration results on account of capture of an electron/hole in a trap is termed as "colour centre". Under normal storage conditions these traps may be quite stable even for years. However, by proper stimulation (thermal) their probability of getting detrapped can be enormously increased and the trapped charge carriers can come back to their original stable configuration.

This process is accompanied by release of energy as optical radiation, results in the observed TL. If several traps of different depths are involved in the same material, then it is evident that the detrapping will occur at different temperatures and consequently the light emitted will be in the form of several "bursts" of light and will be reflected as several glow peaks in the TL glow curve. When the light is emitted, the nature of emitting spices and its "surrounding" is reflected in the spectral quality of the TL emission as shown by the TL spectral emission curve. The spectral emission peaks may correspond to either different emitting centres or different energy states of the same centres or different environs of the emitting centres.

Many investigators have attributed TL to various forms of impurity luminescence[29-33] and not to colour centre annihilations. However, the reports indicated (F + H) recombination as the determinant for TL emission[34-39]. In fact a good correlation has been observed between glow peaks and various annihilation steps of colour centres for alkali halides[34, 40-44], alkali doped samples[44-46] as well as for other monovalent[39] or divalent[33,35,47] cation impurity activated specimens. Further, they established that TL of irradiated alkali halides arises on account of annihiling of colour centres with suitable radiative recombination at the emission centres. Among the colour centres, F centres are known to play an important role in the TL occuring above room temperatures[29,48-50]. It has been shown that thermal bleaching of F centre electron with subsequent recombination at hole centre give rise to TL above room temperature in pure alkali halides like KCl, KBr and NaCl[51-53].

The experimental result of monovalent (thallium) impurity doped alkali halides on thermoluminescence[31,54-63] have clearly brought out the fact that the exposure to ionizing radiation either excites the electron which gets trapped at the metastable level within the impurity ion or separates the electron from its present impurity ion and electron so released is then trapped at crystalline imperfection other than the impurity ion (forming F type of colour centres). Liberation of the captured electrons during heating and their recombinations with holes at impurity centres results in the emission of the light photon.

The survey of literature of the work on TL of divalent cation impurity (e.g. Ca, Ba, Sr) activated alkali halides indicated[14,22,25] that the general mechanism involved in occurrence of TL remains more or less identical, but the nature of traps, emitters and surrounding physical environments are totally different than pure and monovalent cation impurity doped alkali halides. Since the experimental work in this thesis is mainly concerned with study of TL behaviours of Barium doped NaCl and KCl materials, the upto date present understanding of TL mechanism involved in divalent doped alkali halide is accounter hereafter.

On the basis of experimental results and long experience the crystallographers have established the fact that no crystal, synthetic or natural, is perfect in all respects. In view of this, it is very clear that alkali halide crystals contain primarily cation and anion vacancies, termed as Schottky defects, which are believed to be present inherently. Since the crystals as a whole is electrically neutral, the types of vacancies are present in equal concentrations.

Lidiard[64] and Seitz[65] reported that the deliberately introduced or inherently present impurities in alkali halide crystals mainly occupy substantial positions at the site of alkali ions. Unlike monovalent impurity, the extrapositive charge of the divalent impurity ions produce simultaneously equal number of negatively charged defects in alkali halide lattice to satisfy the requirement of charge neutrality of the crystal. In an undoped alkali halide crystal, the easiest defect to form and satisfy this requirement is the positive ion vacancy or cation vacancy. Thus, the introduction of divalent cation impurity in the spectroscopically pure alkali halide produces an equal number of cation vacancies in alkali halide host lattice. The expected modes of existence of these divalent cation impurities are :

(i) independent of each other as free vacancies and free divalent ions.

(ii) associated in pairs or to form dipoles; Impurity-vacancy (I-V) or

(iv) associated together in higher complexes than pairs.

Alkali halide activated with divalent impurity have been found to contain all the three above mentioned modes of existence. Since the impurity ion has excess positive charge, while the cation vacancy is the centre of negative charge, there is coulombic attraction between them which results in close association of the impurity and vacancy (I-V pair). Such an association is also favourable from energy consideration[65]. The I-V pair is viewed as an electric dipole. It is found that below about 300°C the degree of association in an I-V pair is optimum and decreases with further increase in temperature. Theoretical[66-68] and experimental results reported earlier have shown that at room temperature, most of the divalent cation vacancies are present in the form of dipoles rather than free entities which then aggregate further to form higher complexes in different physical conditions. The experimental work in the field of ionic conductivity[59,65], dielectric loss[59,72], diffusion[65], optical colouration[73], electron spin resonance[74-76] has also confirmed the close association between divalent cation, cation vacancy leading to formation of electric dipoles. Bearing in mind, the types of defects are normally available in a divalent impurity doped alkali halide, as inferred, from the relevant literature referred above, the present work aims at obtaining insight into the TL mechanism of alpha irradiated Ba doped sodium and potassium chloride phosphors.

The TL glow peak observed in pure NaCl are 90 and 240°C. It is very clearly seen that the peaks observed at 180, 200, 220 and 230°C are associated with Ba impurity in NaCl:Ba phosphor. On the other hand pure KCl display small peaks at 50, 100 and a hump at 340°C. The two principle glow peaks observed KCl:Ba are 100 and 170°C. It is important to note that 340°C glow peak remain present in all the samples of KCl in first thermal cycles.

In what follows, the TL results of NaCl and NaCl:Ba materials are presented first. This follows the TL characteristics of pure and Ba doped KCl which are discussed in the following section.

(B) Glow Peaks related to pure NaCl

(1) 90°C glow peak

The typical glow curve displayed by virgin undoped sodium chloride powder specimen, after irradiating it by an alpha dose of 3800 rad are represented in Fig. 1. It is observed that the specimen as-obtained from aqueous solution, undoped sodium chloride, exhibits small but a well defined glow peak at 90°C alongwith the humps at 240 and 340°c during the first heating run. It is interesting that the hump at 340°C observed during the first heating run, disappears in the second run and then remains absent for any further successive run (Fig. 1).

It is clearly seen from Fig. 1 that the intensity of the 90°C peak increases during the second heating run. Further, it is observed that the intensity of this peak stabilizes itself in the third heating run (Fig.1). This feature is observed in all untreated and thermally treated as-obtained pure and Ba^{++} doped sodium chloride specimens (Fig. 1-12).

The another fact observed is that, the above feature is significant in the as-obtained pure and Ba doped NaCl (Fig. 1,4,7 and 10) compared to that in heat treated specimens. It is quite reasonable to presume that in untreated specimens the inherent impurity present is quite small and may be in a separate precipitated phase. During the first heating run, the specimen was already heated 500°C and rapidly cooled to room temperature. This may result in the diffusion of inherent impurity into NaCl lattice and thereby increase the number of TL centres and hence enhanced the intensity of the 90°C glow peak. The dissolution of impurity in solid solution is temperature dependent and expected to be more at higher temperature. The NaCl specimen at 500°C will have a capacity to dissolve certain number of impurity ions. After the dissolution of inherent impurity during first heating run, the impurity available for dissolution at 500°C during subsequent heating runs will continue to decrease. Hence, the enhancement in the intensity of TL will not be pronounced in latter heating runs. The above assumption is well corroborated by the fact that when the specimen is subjected to pre-heat treatment there is no marked change in the intensity of the glow peak between the first and subsequent heating run (Fig. 2 and 3). In the pre-heat treatment,

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the main bulk of precipitated impurity is supposed to have redissolution due to annealing and quenching (Fig. 2 and 3).

Comparison of TL glow curves for thermally untreated specimens and quenched from different temperatures show significant changes in the shape of glow curves. The broad weak peak at 90°C which is obtained in case of untreated specimen enhances in the intensity when the specimen is quenched from 500°C. The intensity of this peak decreases when the specimen is quenched from 750°C. This feature is significantly observed in pure NaCl material.

It is well-known that the rapid quenching of the specimen from high temperature produces thermal strain in lattice. This produces a motion and multiplication of dislocations and consequently a large number of vacancies and dislocations are created in the specimen. The redissolved impurity during quench therefore give rise to impurity dipoles distributed in dislocations and dislocation free regions. It is believed that due to higher dislocation density in the sample quenched from 750°C, the chances of dipoles situated in the dislocation free region are less for this specimen than in one quenched from 500°C.

The 90°C peak is also observed in cases of Ba^{++} doped sodium chloride. In these cases, the peak is observed very weak. In all the Ba^{++} doped specimens the intensity of 90°C is lower than that observed in pure NaCl. It is also found that the intensity of this peak decreases with increase in Ba^{++} concentration (Fig. 1, 4, 7 and 10). These experimental results lead the author to conclude that the peak at 90°C is the inherent property of undoped material.

Since the material is in as-obtained condition from the manufacturers is supposed to have annealed and cooled slowly. The 90°C peak may be associated with the crystallinity of the specimen.

No synthetic or natural material is absolutely pure, however ultrapure it is made, some impurities in the trace quantities are inherently present. It is therefore suggested that the peak may be associated with unavoidably present inherent impurities in host material namely, sodium chloride. As per Maker's data the divalent ions of Ca group and Mg are inherently present in NaCl. It is presumed that these inherent divalent impurities from electric dipoles in the host lattice material (In-V). Since, the glow peak is observed very nicely in untreated NaCl, it is proposed that the electric dipoles present in the perfect region of the host lattice are associated with this glow peak. On the basis of volume considerations, the association of vacancy or vacancies with the impurity ion should minimize the strain energy in the region of impurity. It is suggested that the TL centres responsible for the occurrence of 90°C thermal glow peak is a complex formed by a negative ion vacancy and an inherent impurity dipole (In-V) situated in the perfect region of NaCl lattice.

(2) 240 and 250°C peaks.

The NaCl specimen prepared from aqueous solution exhibit a small peak or a hump at 240°C alongwith the peaks at 90 and 340°C in the first thermal cycle. Among these the peak at 340°C found most intense (Fig. 1). It is very interesting to note that a hump at 240°C changes its shape and appears as a well defined dominant peak at 250°C during the recording of second cycle. The further successive cycles do not produce significant change in glow curve. The another noteworthy fact is that, this peak appears as isolated, well defined and dominant one in 500 and 750°C air-quenched NaCl specimens (Fig. 2 and 3). It is also observed that the intensity of this peak slightly changes when the specimen is quenched from 750°C (Fig. 3). However, the peak is well defined and well intensed, as compared to untreated NaCl. In any of the cases, the intensity of this peak stabilizes itself after the completion of second cycle. It is cleared that, this peak is favoured by thermal treatment. It is a property of heat-treated NaCl. It is important to note that the peak at 240°C is also observed in as-obtained lightly doped NaCl:Ba (10^{-5}m.f.), (Fig. 4). The effect of thermal cycles on the peak in this specimen is identical to that observed in pure untreated NaCl (Fig. 1). This peak remains absent in all other samples of NaCl:Ba with Ba^{++} concentration higher than 10^{-5} m.f. (Fig. 4, 7 and 10). The heat treated NaCl:Ba specimens also did not display 240°C peak. Therefore, it is suggested that the peak at 240°C must be independent of Ba^{++} impurity and associated with TL complex formed by the close association of negative ion vacancy and (In-V) dipole. Since, it is observed predominant one after completion of one cycle, it is believed that the [Vacancy - (In-V)] complex is located in the dislocation region. All observed properties can be explained with the help of the hypothesis proposed for 90°C glow peak.

(C) TL-Peaks related to Ba^{++}impurity in NaCl:Ba Phosphor.
(180, 200, 220 and 230°C peaks)

The examination of TL glow curves exhibited by NaCl:Ba with different Ba^{++} concentrations after excitation with the standard alpha dose (3800 rad) indicates beyond doubts that the peaks at 230, 220, 200 and 180°C (Fig. 4-12) are related with the deliberately introduced divalent cation impurity (Ba^{++}).

The peak at 230°C is observed when lightly doped NaCl:Ba (10^{-5}m.f.) is subjected to pre-heat treatment. It is found from figures 4,5, and 6 that the 230°C peak is the only peak which is well resolved, stabilized and prominent. It's intensity grows with the increase in thermal quenching temperature. It becomes optimum in the 500°C air-quenched NaCl:Ba (10^{-5}m.f.) specimens (Fig.5) and then suddenly drops in the specimens air-quenched from higher temperature, say 750°C (Fig. 6).

It is clearly seen that the peak is the special property of Ba^{++} impurity in NaCl lattice at very low concentration. The intensity of this peak rises substantially after the completion of first cycle and then stabilizes itself for further heating runs.

The thermal cycling effect is not that much significant in the pre-heat treated lightly doped NaCl:Ba specimens.

It is very clear form figures 7,8,9 and 10,11,12 that the increase in Ba^{++} concentration drastically changes the TL glow curves. The peak at 230°C disappears and it comes up at 220°C for 10^{-2}m.f. of Ba concentration. This peak again shifts to 200°C in heavily doped specimens. Not only this, but after pre-heat treatment, the heavily doped NaCl:Ba material exhibits TL peak at 180°C (Fig. 12). The thermal cycling effect is more or less identical in all Ba doped specimens, which is remarkable in un-heat-treated specimens. The intensity of Ba-induced peaks is more or less equal in 500 and 750°C air-quenched specimens.

In short, it is believed that the 230°C peak is the property of lightly doped NaCl:Ba (10^{-5}m.f.) and the characteristic glow peaks at moderately (10^{-2}m.f.) and heavily (10^{-1}m.f.) activated NaCl:Ba materials are 220 and 200°C respectively.

As mentioned earlier for pure NaCl it is suggested that deliberately introduced Ba impurity dipole positioned near the negative ion, that is complex, [(I-V) dipole nearby negative ion vacancy], is associated with the appearance of the Ba induced glow peaks namely 230, 220, 200 and 180°C. It is believed that

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the [(I-V) negative ion vacancy] complexes, differing in the configuration of the components are presumed to be the centres responsible for the above mentioned glow peaks in NaCl:Ba. The statistical probability for the negative ion vacancy to be positioned as nearest neighbour (nn), next nearest neighbour (nnn) and third nearest neighbour (tnn) positions to the (I-V) dipole is closely related to the pre-thermal and pre-history of the phosphors. Since the dislocation cores are reported to be negatively charged[77-79], it is reasonable to assume that for the formation of the proposed (I-V) negative ion vacancy complex, the Ba-dipole should be located in the close proximity to the dislocation to have a negative ion vacancy as its component. Since the 230°C glow peak is observed only in lightly doped NaCl:Ba (10^{-5}m.f.) in which the dipole concentration is low and observed very nicely after thermal treatment, it is believed that second type of situation namely (Ba-dipole) negative vacancy at (nnn) site may be responsible for the occurrence of 230°C glow peak.

Further it is suggested that when the impurity concentration is higher and moderate (10^{-2}m.f. of Ba), large number of dipoles are formed in the matrix resulting in localized symmetric strain. This primarily induces the dipoles to diffuse in the strain energy. Since all Ba-doped glow peaks appear well defined after thermal treatment, it is presumed that the 220°C peak is the property of TL centre located very near or on the core of the dislocation with the component negative ion vacancy occupying the nearest neighbour (nn) position to the Ba dipole. The nearest neighbour position for negative ion vacancy is based on the statistical consideration because these vacancies are presumed to exist in higher concentration near the core of dislocation on the basis of the charged dislocation concept.

The thermal cycling and pre-heat treatment effects observed in the present experiments can be interpreted on the basis of the redistribution and redissolution of the precipitated divalent cation impurity which is limited in content after heat treatment.

It is found that the intensity of the peaks related to Ba-impurity grows with the increase in the content of barium cation impurity in sodium chloride material upto certain moderate concentration (10^{-2}m.f.). Thereafter, the intensity drops down for further increase in the concentration of Ba^{++} impurity, 10^{-1}m.f. of Ba^{++}(First cycles of figures 4,7,10,5,8,11 and 6,9,12). It is believed that the increase in the concentration of Ba^{++}impurity generates larger and larger number of (I-V) dipoles. This developes large number of TL centre available for TL emission. On the other hand, at higher concentration instead of single, pair and higher aggregates of (I-V) dipoles are expected to form. Besides this they will be under the influence of each other, which inturn result concentration quenching centres. On account of which the intensity of glow curve drops even though the concentration of impurity is increased. However, in each case the change in intensity is observed after completion of first thermal cycle or execution of thermal treatment from 500 or 750°C quenching temperature. It is believed that in all the above three conditions the thermal strain produced during heat treatment promotes the diffusion and redistribution of the impurity and vacancies through dislocation motion in host lattice.

(D) Mechanism of TL emission process at 220°C peak in NaCl:Ba (T) phosphor.

The emission wavelength at particular glow peak temperature, 220°C, in the case of 750°C air-quenched NaCl:Ba (10^{-2}m.f.) specimen, referred as NaCl:Ba (T) hereafter, has also been investigated to propose the TL emission

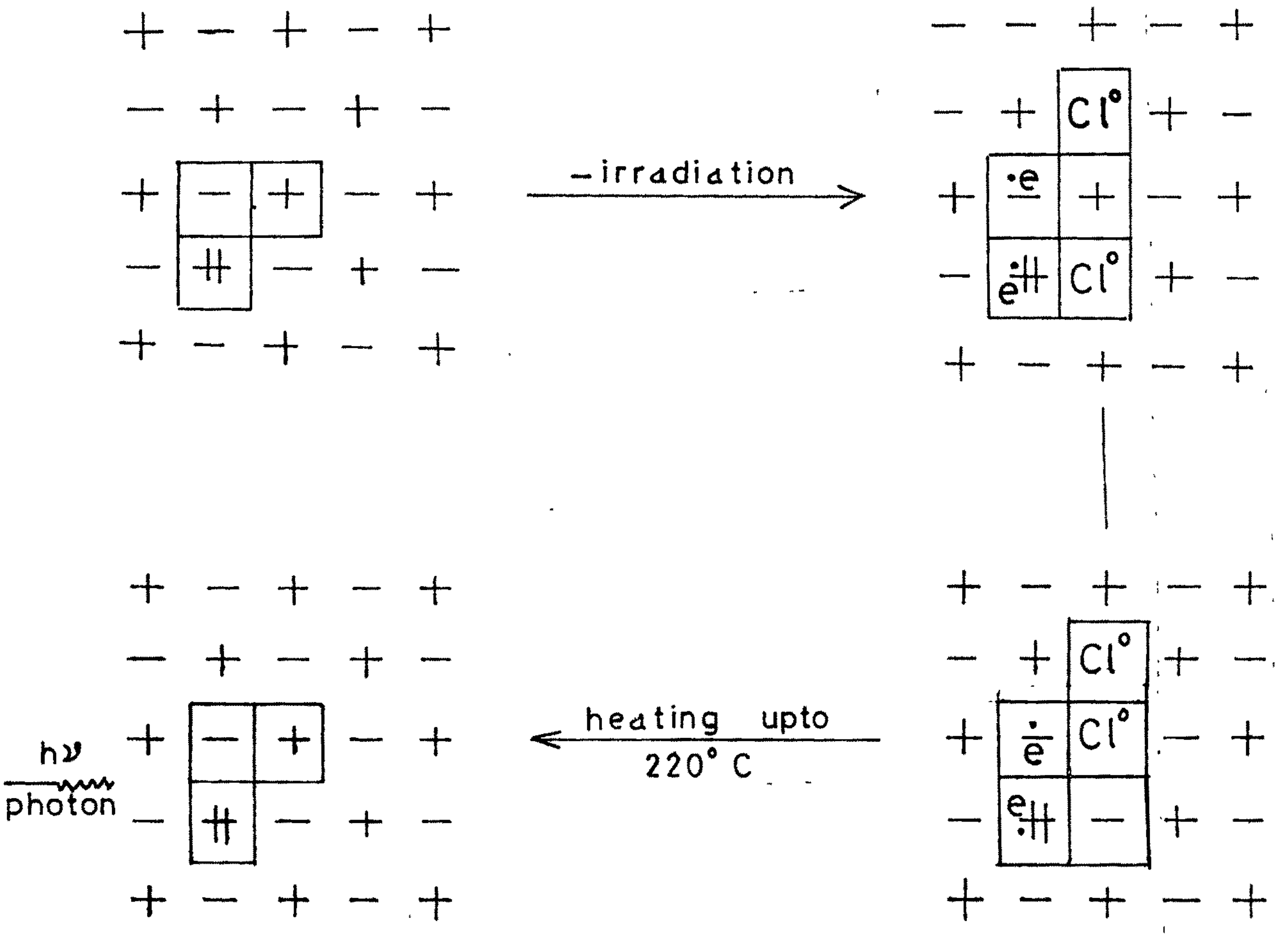

FIG. VI-1: MECHANISM OF TL EMISSION PROCESS AT 220° C PEAK IN NaCl : Ba (T) PHOSPHOR centre and the mechanism of TL at 220°C peak in the NaCl:Ba (T) specimen (Fig. 36). The correlation of the TL emission spectra and glow curves brings out the fact that the TL emission output at 435 and 495 nm is mainly observed in the temperature regions where the well defined TL peaks are observed. Since the emission at 495 nm and 435 nm have been found prominent respectively at 90 and 220°C, it is presumed that 90 and 220°C glow peaks are associated with 495 and 435 nm emission bands respectively. It is also very clear that the peak around 140°C is related to 357 nm. Since the emission spectra of ionized barium atom[80] tally with the TL emission bands observed in the present experiments, the emission centres may be identified with the Ba impurity centre.

The mechanism of TL emission process at 220°C peak is proposed as follows (Fig. VI-1).

(i) The electrons are released from the adjacent Cl^- ions on irradiation of NaCl:Ba (T).

(ii) These electrons then get trapped at Ba^{++}ion and at negative ion vacancy forming the components of the TL centre.

(iii) The relaxation of Cl° atom into positive ion vacancy of Ba- dipole takes place through Crawford-Nelson mechanism[81]. This results the production of Cl_2 molecules near F-centre which are stable above room temperature.[82]

(iv) Heating of the NaCl:Ba (T) specimen with uniform heating rate to 220°C thermally bleaches the electrons from the F-centre. The released electrons recombine with the holes on the Cl° atom of the Cl_2 molecule.

(v) The recombination energy being very low, the excess energy is transferred to the adjacent Ba^{++} ion.

(vi) The transferred energy excites the Ba^+ ion. The characteristic TL emission is exhibited when the excited Ba^+ion returns to the ground state.

It is believed that the steps involved in all other Ba-induced TL glow peaks namely, 230, 200 and 180°C are more or less identical and hence not given separately.

The two peaks at 200 and 180°C appear predominantly in heavily doped specimen (10^{-1}m.f.) when the specimen is subjected to pre-heat treatment at 500 and 750°C respectively (Fig. 11 and 12). The peaks at 200 and 180°C are therefore the special property of heavily doped and thermally treated NaCl:Ba specimens.

The minor increase in the intensity of these peaks is observed in thermally treated specimens, whereas significant change in intensity of these peaks with successive heating runs is seen in untreated specimens.

In case of heavily doped NaCl:Ba the formation of pairs or higher aggregates of dipoles will naturally become more pronounced. Cook and Dryden[84-85] have reported aggregation of dipoles on aging at room temperature. They found the aggregation proceed in two stages, the initial aggregation of dipoles in cluster of three (or trimers), followed by aggregates into higher complexes. It has been reported before that trimers or complexes of more units do not exist in appreciable number beyond 110°C. On the other hand, from their study of precipitates formed in divalent alkali crystals. Miyaki and Suzuki[83] have investigated that divalent ions together with the charge compensating vacancies develop metastable plate like aggregates on $\{1\ 1\ 1\}$ and $\{3\ 1\ 0\}$ planes. The former one was found to be stable at temperatures greater than 150°C while latter below 100°C.

In view of these thermal stabilities of metastable aggregates, it is suggested that aggregates on $\{1\ 1\ 1\}$ planes like say, trimer may be responsible for

200°C glow peaks. The orientation of this trimer in different directions or the location of trimer in different strain region on account of execution of thermal treatment to heavily doped NaCl:Ba specimens may be responsible TL-centre for 180°C. One must note that this peak only appears in a case of 750°C air-quenched heavily doped NaCl:Ba. The shifting of the peak from 200 to 180°C may be due to different environments around aggregates on $\{1\ 1\ 1\}$ in the lattice of 750°C air-quenched heavily doped NaCl:Ba.

(E) Effect of different types of Ionizing radiations on TL of NaCl and NaCl:Ba.

It is generally expected that the impurity (deliberately introduced or inherently present) precipitate out as a separate phase in pure or divalent cation impurity doped alkali halides would get redissolved on application of heat treatment which results stable well defined glow curve. This is coroborated from the TL glow curves presented in figures 13 to 15. Because of this reasons the phosphors of pure and Ba- doped NaCl, air-quenched from 750°C were selected for comparison.

Figures 13 to 18 display the TL glow curves of 750°C air-quenched NaCl and NaCl:Ba with three different concentrations namely, 10^{-5}, 10^{-2} and 10^{-1}m.f. of Ba after one minute exposure to beta (800 rad) and gamma (800 rad) radiations. The examination of these figures brings out the following facts :

1. TL characteristic is sensitive to the nature of excitation sources.
2. The exposure to gamma radiation results maximum TL output while that to alpha radiation gives minimum TL output with higher dose (i.e. 3800 rad)
3. The peaks are well defined and isolated with gamma irradiation.
4. The new peaks in 90-240°C region also come out with excitation with gamma radiation in pure NaCl.

5. Further, it is seen that higher temperature peaks (240°C) are more prominent in gamma irradiation specimens.
6. The peaks related to Ba- impurity, 140, 180, 200 and 220°C are well defined and remain present at a time in single glow curve with beta and gamma irradiation.
7. Like pure NaCl, the doped specimens also display low TL output under alpha excitation. The peaks in the region 140 - 230°C are found more pronounced with gamma irradiation as compared to that with alpha excitation.

According to latest understanding of radiation damage in solids, different type of radiations possess different energies ($E = h\nu$) as frequencies are different. Their characteristics are also different. They interact differently with various materials and produce variety of damage in solid. A particular radiation, also induces different damage on interaction with solid material at different radiation doses. In order to understand this, it is important to know the energy loss of the irradiating particles and photons in a crystal. A survey of literature indicated that the depth of penetration of particle in a crystal is very important in radiation damage. The depth depends on the energy loss. Theoretically, it has been established that the energy absorbed from electrons or photons is very much lower than that from a heavy particles like alpha particles.[86] As a result of this heavy particles do not penetrate deep into the crystal and produce less radiation damage. Besides this, heavy particles are also stopped near the surface of the crystal and the damage is comparatively less. Radiation damage means, creation of vacancy, vacancy pair, dislocations, release the electrons or holes etc.. It is very important to note that electrons or beta particles of 1 MeV energy penetrate a depth of 0.22 cm in a crystal of alkali halide, namely KCl. Whereas on alpha particle of same energy penetrates 2×10^{-4} cm. in the same alkali halide crystal. Thus penetration power of alpha, beta and gamma radiations are different. It is maximum for gamma and minimum for alpha radiation. The beta radiation has moderate penetration power.

Because of this reason, the gamma radiation can excite different types of emitters as well as can excite large number of TL centre associated with 140, 180, 220 and 240°C glow peaks in the specimen of pure and Ba doped NaCl. This offers the interpretations for above listed experimental results (Fig. 13-18). The overall TL emission under 90, 140, 180, 220 and 240°C peaks is much higher when specimen is exposed to gamma than with beta radiation and much lower than beta or gamma when irradiated with alpha radiation. The appearance of most of the peaks under gamma excitation and their absentia under alpha irradiation can also be interpreted on the hypothesis discussed above.

(F) Glow peaks associated with pure and Ba doped KCl.

The thermoluminescence characteristics of pure and Ba^{++} activated KCl have been examined after exposure to a standard alpha dose (3.8×10^3 rad). The specimens were studied in as-obtained, 500 and 750°C air-quenched conditions. The material of KCl:Ba with three different concentrations of Ba^{++}, namely, 10^{-5}, 10^{-2} and 10^{-1} m,f, have been examined for their TL behaviours in all the three above mentioned physical conditions. The experimental data presented in figures 19-30 very clearly brings out the following facts :

1. The pure KCl mateial in all the three physical conditions, namely, as-obtained, 500 and 750°C air-quenched, shows a small peak at 50°C and a hump at 340°C in the first thermal cycle (Fig. 19, 20 and 21).
2. This feature is commonly observed in the first cycle of almost all untreated and heat treated pure as well as Ba doped KCl.
3. The peak at 340°C disappears and a well defined peak at 100°C appears alongwith 50°C glow peak in the second thermal cycle of all KCl specimens.

This glow pattern remains stable for all other subsequent thermal cycles.

4. A dominant glow peak at 100°C is observed in lightly doped KCl:Ba (10^{-5}m.f.) Fig. 22. The 500°C air-quenched thermal treatment developes a new peak at 170°C alongwith 50°C and 100°C (Fig. 23). The relative intensity at the 100°C glow peak is more among these three peaks. It is very interesting that the relative intensity of 170°C glow peak becomes higher than 50 and 100°C glow peaks in 750°C air-quenched lightly doped KCl:Ba phosphors (Fig. 24).

5. It is important to note that the specimens of 500°C air-quenched KCl:Ba with moderate Ba concentration (10^{-2}m.f.), display a well defined prominent peak at 170°C in the first thermal cycle alongwith the peaks at 50 and 340°C (Fig. 26). The intensity of 170°C glow peak rises abruptly in the second cycle and then stabilizes itself.

6. Heavily doped as obtained KCl:Ba (Ba concentration 10^{-1}m.f.) generates a very small peak at 170°C alongwith 50 and 340°C (Fig. 28). The intensity of 170°C peak increases with the application of thermal treatment. Its intensity increases with the rise in temperature of quench upto 500°C. The intensity suddenly drops if the specimen is air-quenched from more than 500°C, say 750°C (curve 1, Fig. 28, 29 and 30).

7. The change in the glow curve pattern with successive thermal cycles in heavily doped KCl:Ba is more or less identical to that observed for lightly doped KCl:Ba. The only difference observed is in the appearance of the dominant glow peak. The prominent peak appears at 170°C in heavily doped specimens (Fig. 28-30), while that exists at 100°C in lightly doped KCl:Ba material (Fig. 19-21).

8. The examination of first thermal cycles of 750°C air-quenched KCl:Ba specimens with different concentrations. Fig. 24, 27 and 30 clearly indicates that 750°C air-quenched KCl:Ba with Ba concentration 10^{-2} m.f., displays intensed peak at 170°C.
9. The intensity of the peak at 50°C does not change with the increase in concentration of Ba^{++} in KCl:Ba specimens (Fig. 19-30).

In what follows, attempts are made to give qualitative interpretation to the above mentioned properties of the glow curves of variously pre-treated KCl:Ba specimens.

It is very clear from above listed results that a small peak at 50°C appears in pure as well as Ba doped KCl specimens. Besides this, the peak intensity is totally independent of Ba concentration. Therefore, it is reasonable to suggest that the peak at 50°C is the property of pure KCl. It may be associated with crystallinity of the specimen. Like NaCl, KCl obtained from market is certified by makers to contain avoidably present inherent impurities such as Barium, Calcium, Magnesium...etc.. The role of these background impurities can not of course, be overlooked.

Since, the intensity of 50°C peak is independent of pre-heat treatment as well as Ba concentration, it is believed that it may be associated with unavoidably present Mg impurity which would provide only a limited number of saturable TL centres responsible for 50°C peak.

The minor variation in the intensity of this peak with successive thermal cycling can be explained on the basis of the change in defect pattern and redistribution of inherent impurity and vacancy on execution of thermal treatment during subsequent heating runs.

The another lower temperature peak say at 100°C is found relatively well defined at lowest barium concentration, in as-received specimen. The as-obtained phosphor can be considered as good as a slowly cooled specimen. Therefore, it is proposed that the TL centres for 100°C peak are located in dislocation free regions. In contrast to this, the peak at 170°C becomes prominent in pre-heat-treated heavily doped KCl:Ba. It is quite obvious that the centres for 170°C are situated in the dislocation region of the KCl host lattice.

In most of the experiments the peaks at 100 and 170°C have appeared in the second cycle with their intensities nearly to their saturation values (Fig. 23, 24, 26, 29). This brings out the fact that heat treatment during the first thermal also plays an important role in generation of 100 and 170°C glow peaks. As proposed earlier, the uniform distribution of impurities and defects in the host lattice of KCl are suggested to be responsible for the observed change in TL properties of KCl:Ba. The thermal treatments, rapid cooling of KCl:Ba material in air from 500°C to room temperature, do not significantly increases the intensity of 100°C peak (Figs. 23-27, first cycles), while that favours the peak at 170°C (Figs. 25-27 , first cycles). These experimental results supports the suggested hypothesis that the TL centres for 100 and 170°C are located respectively in perfect and dislocation regions of host lattice.

In view of the hypothesis developed in previous section for NaCl:Ba, it is also proposed that the TL centres associated with 170°C peak in the specimen of barium doped KCl should be the Ba- dipole studied in dislocation region.

From their correlative study of TL glow curves and TL emission spectra of KCl:Ba[96], they reported that, characteristic emission is absent in TL emission of KCl:Ba. However, they found that the increase in barium impurity grows the

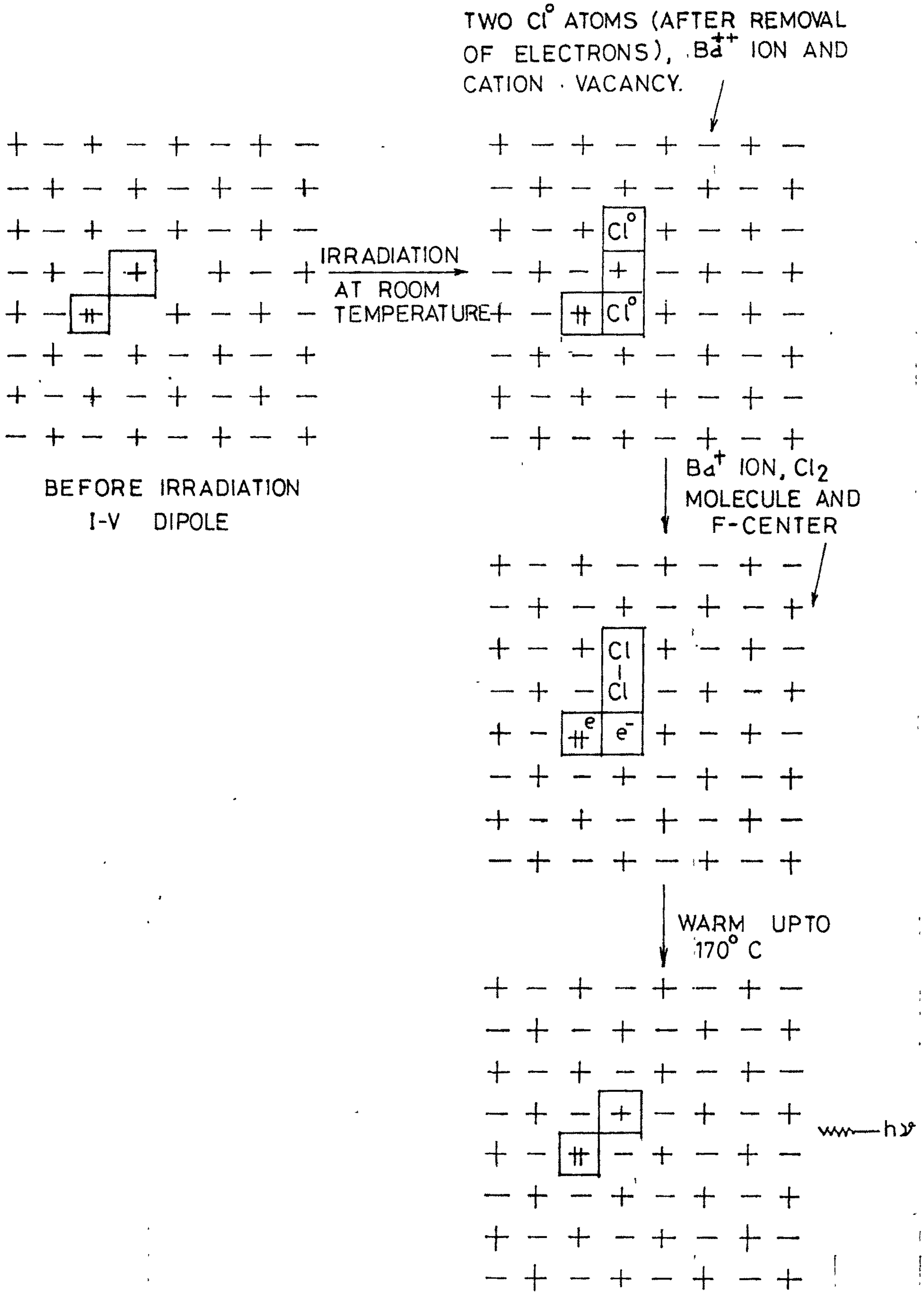

FIG. VI-2: THE SEQUENCE OF STEPS IN THE OCCURRENCE OF 170 C GLOW PEAK.

intensity of TL of KCl:Ba. Therefore, the mechanism of TL at 170°C in KCl:Ba is different than that suggested for 220°C in NaCl:Ba (T).

The mechanism proposed for 170°C glow peak in KCl:Ba (T) is as follows:

1. Exposure to alpha radiation of KCl:Ba, ejects a large number of electrons in the host lattice.
2. This will produce two Cl° atoms in the neighbourhood of dipole.
3. One of the Cl° atom relaxes into the cation-vacancy of the dipole through Crawford and Nelson mechanism[59], and creates a halogen molecular complex oriented along $\langle 1\ 0\ 0 \rangle$.
4. The Ba^+ ion and the generated anion vacancy of the centre, capture one electron each from those released in the lattice.
5. The excited TL centre for 170°C is generated by the association of a Cl_2 molecule, Ba^+ ion and an F centre.
6. The heating of the phosphor to 170°C ejects the holes trapped at the centre whose recombination with the trapped electrons within the centre gives rise to emission of photons.

It is suggested that in the case of impurity doped KCl:Ba the Ba^{++} ions are only few and isolated. They are located here and there. The excitation with alpha radiations produce electrons as well as interstitials in the lattice. The electrons are captured by isolated Ba^{++} ions. On capturing electrons, they become Ba^+ ions. The intenstitial ultimately generate halogen molecules or molecular ions,[33,87-91] which are stable at room temperature. It is presumed that at at about 100°C the interstitials are released from traps and become mobile. The recombination of Ba^+ ions with mobile interstitials results in the emission of light at 100°C. Similar type of mechanism but with inherently present Mg^{++} ions is believed to be responsible for the occurrence of 50°C in pure KCl.

It is concluded that 170°C peak appears as a well developed and prominent peak in 750°C air-quenched NaCl:Ba (10^{-2}m.f.) specimen. In this condition of material the peak intensity and shape do not change significantly with successive thermal cycles. In other words , 750°C air-quenched KCl:Ba with moderate concentration of Ba, (10^{-2}m.f.), designated as KCl:Ba (T) exhibits stable and well defined peak at 170°C.

Since 170°C peak in KCl:Ba (T) and 220°C glow peak in NaCl:Ba (T) are found well defined, isolated and intensed, they are selected for the detail dosimetry work. In what follows, the discussion is given in the next section (Part-II).

(G) Peak commonly observed in pure and Ba activated NaCl and KCl.
(340°C glow peak)

It is very interesting to note that the prominent peak at 340°C is exhibited by almost all pure and Ba doped NaCl and KCl specimens (Figures 1-12 and 19-30). This peak disappears after the completion of first thermal cycle. It always remains absent in second and subsequently heating runs. This peak is found to be shifted to higher temperature side around 370°C, in heavily doped NaCl:Ba and KCl:Ba, (10^{-2}m.f. and higher), Table-I.

The pre-heat treatment does not result the significant change in the position and intensity of the peak. Only the pure NaCl (Figures 1, 2 and 3) and heavily doped NaCl:Ba (10^{-1}m.f.) brings some changes (Table-I). This peak is observed under the excitation with alpha-, beta-, gamma- and UV- radiations. It is clearly seen that the types of excitation do not change the position and intensity of 340°C glow peak. The typical glow curves for 750°C air-quenched NaCl are as shown in Fig. 15A and 15B. In order to investigate the origin of this peak, the 750°C air-quenched NaCl and KCl have also been examined for their TL behaviours even without any excitation with ionizing radiation. It is very important to note that this peak appears in 750°C air-quenched NaCl and KCl:Ba specimens without irradiation (Fig. 35A, 35B). The intensity is quite comparable with that of irradiated 750°C air-quenched specimen. Similar type of results are observed in NaCl:Ba and hence not presented in this.

The last experimental fact indicate that the peak at 340°C is independent of types of radiation and types of alkali halide.

Joshi et al.,[21] observed the same peak in thallium doped NaCl. The survey of literature strengthens the fact that this peak is also independent of impurity, i.e. divalent or monovalent.

It is believed that the excitations of centres responsible for this peak is probably initiated by non-radiative means. This could be mechanical pressure during handling of the specimens. This is also corroborated by other researchers[92-94]. The luminescence from non-irradiated phosphors during heating with uniform heating rate is referred as Tribo or Spurious TL. Kathuria et al.,[96] have observed the spurious TL in commonly used TLD materials like LiF, CaF_2, $CaSO_4$ etc. Joshi et al., examined this in NaCl and NaCl:Ca. It is suggested that the peak observed in present work in pure and Ba doped NaCl and KCl is spurious TL glow peak. The mechanism is as follows :

1. The mechanical disturbances during stirring and spreading of the specimen on heater plate excites the material and generates the electron-hole pairs.
2. These pair are then trapped at defect centres.
3. The warming of the specimen with uniform heating rate release the trapped charge carriers from trapping sites through thermal detrapping.
4. These released charges subsequently recombine at recombination centres by emitting visible photons.

5. It is proposed that during warming of specimen for the record of first TL glow curves at about 300°C the oxygen present in atmosphere begins to interact with the surface ions forming new recombination sites which results in the occurrence of spurious TL glow peak at 340°C.

It is very well established that the 340°C peak appears only in the first cycle. It is suggested that the thermal release of trapped charges and their destruction by recombination take place during heating of the phosphors with uniform heating rate to 400°C in the first thermal cycle. In second thermal cycle the phosphors in the heating pan is not disturbed by mechanical pressure. This will not excite the specimen mechanically before recording the second run. Therefore, no spurious TL is expected in the successive heating runs. The change in the intensity and the position of the spurious glow peak (340°C) with increase in Ba concentration in NaCl:Ba (T) and KCl:Ba (T) specimens, may be on account of the formation of additional recombination centres favoured by distortion induced in the lattice due to the presence of impurity ions.

It is believed that the absence or suppression of this peak with exposure of higher beta or gamma dose is due to overshadowing of the glow peak by neighbouring dominant glow peak. Further, our earlier reported experimental results[95] alongwith present data reveal the fact that the strength of this peak for different alpha, beta, gamma or UV doses reaches saturation of the height which is comparable with that obtained for non-irradiated specimens (Figures 35A, 35B, 1-12 and 19-30). This indicates that the peak at 340°C is sensitive to the magnitude of ionizing radiation dose. This strengthens the independent occurrence of the spurious glow peak on irradiation.

It is necessary to point out the suggestions made by Akolekar et al[96] from our laboratory regarding the origin of 340°C glow peak in the case of pure and Ba- doped KCl. From their experimental results, they proposed that the TL centres for 340°C glow peak are metastable clusters made up of single barium dipoles and vacancies. They have not reported 340°C peak as spurious glow peak. It is believed that their interpretation is based on experimental data of KCl system only. Secondly, they have not examined and presented the TL glow curves of KCl and KCl:Ba without irradiation. The present extensive examination of pure and Ba- doped NaCl and KCl alongwith work published in the recent post encourages the author to believe that the 340°C peak is a spurious TL glow peak.

BEHAVIOURS OF 340°C GLOW PEAK IN PURE AND Ba DOPED NaCl and KCl.

Sr. No.	Fig. No.	SPECIMEN			Glow Peak temp.	TL intensity in Amperes.
		As-obtained	500°C air-quenched	750°C air-quenched		
1	1	NaCl	-	-	340°C	6.5 x 10^{-8}
2	2	-	NaCl	-	340°C	3.0 x 10^{-8}
3	3	-	-	NaCl	340°C	17.0 x 10^{-8}
4	4	NaCl:Ba. 10^{-5}m.f.	-	-	340°C	7.5 x 10^{-8}
5	5	-	NaCl:Ba 10^{-5}m.f.	-	340°C	6.0 x 10^{-8}
6	6	-	-	NaCl:Ba 10^{-5}m.f.	340°C	9.0 x 10^{-8}
7	7	NaCl:Ba 10^{-2}m.f.	-	-	370°C	17.5 x 10^{-8}
8	8	-	NaCl:Ba 10^{-2}m.f.	-	370°C	18.5 x 10^{-8}
9	9	-	-	NaCl:Ba 10^{-2}m.f.	370°C	18.0 x 10^{-8}
10	10	NaCl:Ba 10^{-1}m.f.	-	-	370°C	76.0 x 10^{-8}
11	11	-	NaCl:Ba 10^{-1}m.f.	-	360°C	43.0 x 10^{-8}
12	12	-	-	NaCl:Ba 10^{-1}m.f.	360°C	28.0 x 10^{-8}
13	19	KCl	-	-	340°C	5.0 x 10^{-8}
14	20	-	KCl	-	340°C	5.0 x 10^{-8}
15	21	-	-	KCl	340°C	4.0 x 10^{-8}
16	22	KCl:Ba 10^{-5}m.f.	-	-	340°C	8.8 x 10^{-8}
17	23	-	KCl:Ba 10^{-5}m.f.	-	340°C	7.3 x 10^{-8}
18	24	-	-	KCl:Ba 10^{-5}m.f.	340°C	6.2 x 10^{-8}
19	25	KCl:Ba 10^{-2}m.f.	-	-	370°C	5.3 x 10^{-8}
20	26	-	KCl:Ba	-	340°C	8.4 x 10^{-8}

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Sr. No.	Fig. No.	SPECIMEN As-obtained	SPECIMEN 500°C air-quenched	SPECIMEN 750°C air-quenched	Glow peak temp.	TL intensity in Amperes.
21	27	-	-	KCl:Ba 10^{-2}m.f.	340°C	2.2×10^{-8}
22	28	KCl:Ba 10^{-1}m.f.	-	-	340°C	5.0×10^{-8}
23	29	-	KCl:Ba 10^{-1}m.f.	-	370°C	9.8×10^{-8}
24	30	-	-	KCl:Ba 10^{-1}m.f.	340°C	4.0×10^{-8}

TABLE - 1

PART - II

APPLICATION OF TL OF NaCl:Ba (T) AND KCl:Ba (T) IN RADIATION DOSIMETRY.

(A) Brief account of research in TL dosimetry.

Radiation dosimetry deals with the detection and estimation of radiation exposures. The three major available types of radiation detectors are (i) Physical detectors, (ii) Chemical detectors and (iii) Biological detectors. Thermoluminescent dosimeters fall under the category of physical detectors. The dosimeter (TL material) stores the dose information when put in radiation field. The evaluation of radiation dose is done later by using TL-reader. The intensity of TL gives the magnitude of the dose.

The selection of TLD material for fabrication of dosimeter has been made mainly by trial and error method with the hope that material will be useful for the estimation of ionizing radiations (alpha, beta, gamma, UV and X-rays). The most widely studied TL dosimetric materials are; LiF, CaF_2, $CaSO_4$, Mg_2SiO_4, $Li_2B_4O_7$ and BeO. At present the TL dosimetric materials available are CaF_2(Mn), CaF_2(Dy), $CaSO_4$ (Mn), $CaSO_4$ (T_m), LiF (Mg,Ti), BeO (Na), $Li_2B_4O_7$ (Mn), and Mg_2SiO_4.

Many investigators have undertaken and examined TLD through different aspects[4,5,8,25]. On the basis of their work, they have concluded that a good TLD material must fulfill the following basic requirements.[4,5]

(i) High concentration of trapping sites, simple trap distribution and long storage of trapped charges at normal working temperature.

(ii) High TL-efficiency.

(iii) Resistance to radiation damage of trapping centres, the luminescence centres and the base material itself.

(iv) Matching of the spectral sensitivity of the photomultiplier with the spectral distribution of luminescence emission, and

(v) Low fading, Energy incandescence, Reproducibility, Small size and Low cost of the material.

Amongst the materials that have been investigated for their application as solid state dosimeters for ionizing radiation, two classes, thermoluminescent fluoride and radiophotoluminescent silver activated phosphate glasses have been found extensive practical uses[97,98]. Besides this, dosimetry workers have also explored the wide spread use of pure and RE-doped LiF and CaF_2 in radiation dosimetry over the range 10^{-3} to 10^{6} rads.[89,99,100]

Survey of literature indicated that, till 1978, none of the dosimetry workers reported the use of TL of alkali halides other than LiF in radiation dosimetry. Joshi et al., have undertaken the corroborative research project with Health Physics Division, Bhabha Atomic Research Centre, Bombay to develop the pure and impurity doped sodium chloride as dosimetric material.

As a begining of this work, K.P. Dhake et al.,[17] examined the possibility of NaCl:Ca phosphor as dosimetry material for X-rays. They studied the TL glow curves for NaCl:Ca with various X-irradiation duration from a Philips No. 1009 X-ray generator operated at 35 KV and 20 mA using a tungsten target maintaining all other conditions identical. From their preliminary experiments, they suggested that NaCl:Ca can be of use in X-radiation dosimetry provided it satisfies all the basic conditions for a dosimetry material.

Nehate et al.,[18] prepared the TLD grade NaCl:Tl (T) powder by annealing the NaCl:Tl powder obtained from aqueous solution by the method of recrystllization, at 750°C, for two hours and subsequently rapidly cooled to room temperature in open air. The TL behaviours of NaCl:Tl (T) phosphor have been examined by them in different physical situations. They concluded from their experimental data that due to the high TL efficiency, capability of registering beta dose in the range $10-10^4$ rad, resistance to radiation damage, and negligible fading; the NaCl:Tl (T) phosphors are found suitable for beta dosimetry. Thereafter NaCl:Ca material has also been explored by them[20] as UV dosimetry material.

The use of NaCl:Tl (T) material as a promissing TL material for ultraviolet dosimetry have been investigated by S.P. Barghare et al.,[21]. They have studied the intrinsic TL sensitivity of the material to 253 nm UV radiation. They suggested that, since the material satisfies most of the dosimetric requirements, it is a suitable TL phosphor for UV-dosimetry.

The extensive study of the thermoluminescent behaviours of untreated and thermally pre-treated, undoped and Ca doped NaCl phsophors after exposure to gamma radiation have been made by K.P. Dhake et al.,[15]. They found that 750°C air-quenched NaCl:Ca (10^{-3}m.f.) phosphor display optimum TL output with a dominant and well defined glow peak around 147°C. The study of dosimetric properties of this peak for gamma- radiation has indicated that NaCl:Ca materials fulfill the most basic need of an efficient TLD material and the phosphors can be used in gamma dosimetry for range 10^0-10^4 rad. The material has also been tested for its use in TSL-radiation dosimetry for the low gamma dose of range of 500 m. rad. to 3 rad from the source of ^{226}Ra (9 mCi)[19].

The NaCl:Ba (10^{-2}m.f.) phosphor quenched from 750°C in air, designated as NaCl:Ba (T) has exhibited pronounced TL output with a well defined glow peak at 220°C under excitation with gamma radiation. Mahajan et al., have proposed the TL centres involved in the occurrence of 220°C, they have also recommended the use of NaCl:Ba (T) material in estimation of gamma doses in radiation applications. They also pointed out that the material recommended is hygroscopic and unstable in moist climate, which limits its applications in dosimetry. However, they suggested that proper shielding from the direct exposure to contaminating agents may make the material very useful in radiation dosimetry. Besides this, E.E. Heywood et al.,[22] have studied the TL of pure NaCl after thermal treatment. They have developed the method to prepare TLD grade NaCl. From their experimental results they concluded that NaCl could be a valuable TL material provided that the limitations can be minimized. Further they reported that the high sensitivity ideal glow characteristics and low cost could make NaCl phosphor, a commonly used TL material. They have also used NaCl for dose estimation in the treatment of Carcinoma Cervix.

(B) Ba-doped sodium and potassium chlorides in alpha radiation dosimetry.

The above mentioned survey of the work done in the field of use of pure and impurity doped sodium chloride clearly brings out the fact that the material is suitable for beta-, gamma-, X- and ultraviolet radiation dosimetry.

The estimation of alpha dose through TL technique using sodium chloride has not been undertaken by dosimetry researchers. Looking to the success of NaCl as dosimetry material, it is very interesting to investigate the utility of sodium chloride in alpha dosimetry. Therefore, as a test case, NaCl and KCl doped with Ba impurity are selected for the study of dosimetric properties to find out their suitabilities in alpha radiation dosimetry. In what follows, the experimental results are given below :

It has been shown in first part of the thesis that TL centre associated with 220°C in NaCl:Ba (T) [750°C air-quenched NaCl:Ba, 10^{-2}m.f.] involves Ba dipole and a negative ion vacancy situated in the dislocation region. The glow curve shape clearly indicates that the trap is single valued and a large number of them is involved in the generation of the peak at 220°C. This is the fundamental requirement of the good TLD material [requirement No. (i)].

The dosimetry peak in NaCl:Ba (T) appears at high temperature (Fig. 9 and 37) which indicated the involvement of deeper traps. This leads to the long storage of trapped electrons at normal working temperature. Further, it is seen that the location of 220°C peak does not change with increase in the dose of alpha radiations (Fig. 37). Therefore, it is believed that the phosphor under examination is resistive to radiation damage. Thus, the significant TL intensity of the peak and the resistance to radiation damage, the NaCl:Ba (T) material fulfills the condition numbers (ii) and (iii).

It is very clear from the TL emission study (Fig. 36) that the TL emission displayed by the present material around 220°C is 375 nm. This emission falls in the sensitive region of the photomultiplier of TL reader. Thus the condition No. (iv) listed in previous section is satisfied [Part II-A].

The change in intensity of the 220°C glow peak in NaCl:Ba (T) material has been studied with different doses of alpha radiation. The glow curves obtained are presented in Fig. 37. It is seen that the glow peak at 220°C grows with increase in alpha dose upto certain dose. Thereafter, it does not increase with the same rate. The TL intensity versus incident alpha dose response is found to be supralinear. The plot of TL output at 220°C versus room temperature decay time is plotted in Fig. 39. It is clear that the intensity of the peak does not change drastically with the increase in decay time. This leads to the negligible

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fading which fulfills another conditions of an efficient TLD material.

The thermoluminescence radiation dosimetry study has also been extended to Ba- doped KCl material to find out its use in determination of alpha dose in radiation applications.

TL experimental data presented for KCl:Ba under alpha irradiation (3800 rad) for different concentrations of Ba (Figures 22-30) infers that 750°C air-quenched KCl:Ba (with Ba^{++} concentration 10^{-2}m.f.) designated as KCl:Ba (T), exhibits the optimum TL output. Therefore, the specimen of KCl:Ba (T) has been selected for the dosimetric study. The glow curves for alpha irradiated KCl:Ba (T) after different doses of alpha radiation. They are presented in Fig. 40. It is seen from Fig. 40 that, in the temperature region examined, KCl:Ba (T) phosphor exhibit two main glow peaks at 170°C and 340°C. Out of the two peaks observed the variation of 170°C glow peak, with increase in the magnitude of alpha dose is particularly striking. The TL output versus alpha dose response is exhibited in Fig. 41.

It can be said that the basic requirements listed previously for dosimetric material are fairly well satisfied by KCl:Ba (T) material.

In view of the models presented in previous section, the observed peak at 170°C in KCl:Ba (T) is due to the formation of TL centre consisting of a single dipole in association with a nearby negative ion vacancy located in dislocation region. Since, the peak at 170°C appears thermal treatment and quenching the specimen from elevated temperature will lead to uniform distribution of the impurity ions and their subsequent conversion into single dipoles more numerous in number. Hence, there will be significant rise in the strength of the 170°C glow peak, as observed in the glow peak. The shape of glow curve clearly indicates that the trap is simple, single valued and large number of them are involved in the generation of peak at 170°C. The peak appears at high temperature suggest the involvement of deeper traps. This leads to the long storage of trapped electrons at normal working temperature. This fulfills the condition number one. It is obvious from figure 40 and first cycles of figures 21 and 27 that the substance has significant TL output at 170°C, which is the second basic requirement to be satisfied by dosimetric material. The glow curves for different alpha doses have more or less identical stable characteristics namely, peaks at 170°C and 340°C. Since the increase in the magnitude of incident alpha dose do not change the glow peak position and nature of glow curve significantly; it can be believed that the trapping centres, the luminescence centres and the base material are not affected by increase in dose of alpha irradiation. Thus the requirement of high TL efficiency and resistance to radiation damage are fulfilled in KCl:Ba (T) phosphor.

The another important point is to be taken care of, in the detection of TL output and thereby dose is the matching of the sensitive region of the spectral response of the detector and the wavelength region of TL emission. Of course, unlike NaCl:Ba, the study of TL emission spectra indicated that KCl:Ba does not display Ba characteristic TL emission. However, whatever broad emission is there, it falls in the sensitive region of spectral response of the photomultiplier tube. This fulfills condition (iv).

The energy dependence or the tissue equivalence is the another point to be examined precisely. Heywood et al.,[22] have studied this for NaCl. Unfortunately the energy dependence characteristics of the present material could not be examined systematically for the present materials under investigation, because of limited expected facilities available. However, the response of TL output at 170°C with different known incident alpha doses, in the dose range 3.8×10^2 to 3.8×10^5 rads is represented in Fig. 41. It shows straight line supra linear relation between incident alpha dose and TL intensity of 170°C peak. This fulfills the most important property of the TLD material. The other listed requirements like reproducibility of the glow curve, desirable shape and size of the specimen and very low cost of the materials are additional factors which strengthen the claim of NaCl:Ba (T) and KCl:Ba (T) as alpha radiation TLD material.

Of course, there is one serious limitation namely, atmospheric contamination of the present phosphors. This can however be eliminated by shielding the phosphor.