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# Heavy Ion Radiation Damage Track Studies in SSNTDs (Polymers, Glasses and Minerals) and Single Activation Energy Model

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My training in Marie and Pierre Curie University, Paris for the identification of relativistic particle tracks in nuclear emulsions came handy for shifting to Heavy ion track studies in solid state nuclear track detectors (SSNTDs). Track formation mechanism in both may be different, but the basic nature of phenomenon by which the tracks are produced remains essentially the same. All charged particles produce ionisation of the medium and lose energy following Bohr's ionisation loss formula. The stopping power is a basic parameter to be calculated which depends upon the charge and velocity of the particle, and nature of the medium. The other parameters of our interest were range, energy and annealing characteristics of heavy ion tracks in SSNTDs. In 1975, the trio of R. L. Fleischer, P. B. Price and R. M. Walker, considered as a "Father of SSNTD Technique", published their book "*Nuclear Tracks in Solids and their Applications*", which may be considered as a Bible for researchers in the SSNTD field. I had the good fortune to meet this trio and visit their research laboratories after publication of this book.

The counting of fossil and induced fission tracks was required in Geochronology to find the FT ages of minerals. The counting of Alpha tracks was obligatory in polymer detectors to determine Radon and Thoron concentrations in the media. But in both these areas, we never bothered to study the nature of track formation. Heavy ion beams became available in India by the end of 1990 after the installation of 16 MeV Pelletron facility under inter-University Accelerator Centre (IUAC) at New Delhi. During the years, I planned the study of Heavy Ion tracks in SSNTDs we had to depend upon borrowed beam time from UNILAC, GSI, Darmstadt (Germany). I was called to attend the User Committee meeting in GSI held in 1985, which opened the door for entering this domain with acceptance of my proposal.

During 1990, Reimer Spohr was invited as a UGC visiting scientist for a month in our university. We held discussions to start a multi-faculty project using Heavy Ions for irradiation of SSNTDs. We prepared a multi-million Rupees project for funding by the DST, which was approved in first instance. Dr. Thangraj of Applied Physics Department was my research collaborator. We hoped that the sanction of an electron microscope will boost research facilities of the science faculty, but we were surprised that it was cancelled after a review by an excuse that this facility is available in institutions of Chandigarh and can be availed of on sharing basis.

We had been using Fission Fragments in lieu of heavy ions since 1981. SK Modgil left his job as school teacher to continue his research leading to Ph.D. By end of 1981, I was successful in importing a standard fission fragment (FF) source (Californium- 252) from USA for our irradiation experiments. This facility proved to be a great boost for our experimental investigations in the field of radiation damage studies. Modgil took initiative under my supervision to expose soda-lime and other glass samples, and minerals samples to record FF tracks. After irradiation in air and vacuum, the samples were etched using suitable etchants. The track density was recorded by counting tracks under appropriate magnification using a binocular microscope. We published nearly half a dozen papers on the stability of FF tracks under different thermal conditions. Our papers "*Efficiency calibration and effect of etchant temperature on fission fragment tracks in soda glass detector*", and "*Thermal stability of fission tracks in sodalime micro-slide glass*" were published in International Journal of Applied Radiation and Isotopes in 1982.

The highlights of our irradiation experiments were annealing studies of radiation damage in SSNTDs for next twenty years. I guided four more students to study radiation damage in SSNTDs using heavy ion beams from UNILAC facility at GSI, Darmstadt. The beginning was made by Modgil in 1981 using FF source as stated above. Our investigations led to formulation of empirical relations (equations) for bulk and track etch rates in SSNTDs (glasses and minerals). When the data was compiled into a paper for submission to a journal, we had the good fortune to invite Prof. Shankar Mukherjee from IIT Kanpur as Visiting Professor in 1982. He examined our paper and took its pre-print to Kanpur. After a week, he informed that an identical formulation has been proposed by a Hungarian Physicist, G. Somogyi, and published his Paper in Nuclear Instruments and Methods B. First, we thought our labour has gone waste? But Modgil did not lose heart, he had a review of track-etch rate data and analysis of isothermal and isochronic curves of his experiments. He hit upon a new formulation for annealing of radiation damage in SSNTDs, which is known as Single Activation Energy Model of radiation damage annealing in literature. This discovery was reported in 12th SSNTD Conference held at Acapulco in Mexico in 1983, which appeared in the Conference Proceedings under the title "*Track annealing studies in glasses and minerals*" in nuclear tracks and radiation measurements in 1984. Our paper, "*Annealing of fission fragment tracks in inorganic solids*", was published in Nuclear Instruments and Methods in Physics Research B in 1985, which was later on used in its modified form by Prof. P.B. Price and his co-workers in University of Berkeley in their space shuttle cosmic ray experiment.

Before we embarked upon the formulation of single activation energy model of radiation damage in SSNTDs, our laboratory had been engaged in the study of annealing of fossil fission tracks in minerals since 1975 in connection with fission track dating investigations. Annealing of latent radiation damage tracks in various SSNTDs was known to considerably influence etch rates and etchable range of tracks, the critical angle of track registration, and fission track ages of minerals. It was well established that heating of minerals results in reduction of track densities as well as track lengths and diameters. Thus the study of annealing phenomenon was important for track analysis and understanding mechanism of track formation in SSNTDs.

The study of polymer samples exposed to heavy ion irradiation was started after some success with glass samples. Gurmukh Singh guided a M.Phil. student Shkuntala Devi and her paper "*Track etch rate characteristics of Makrofol polycarbonate plastic detectors exposed to Xe ions*" was published in Nuclear Tracks and Radiation measurements in 1986. Ravi Chand undertook the investigations of FF and heavy ion tracks in polymers using electrochemical etching technique after developing his own Etch Cell for this purpose. R.K. Bhatia started working on annealing of heavy ion tracks recorded in polymers, while A.S. Sandhu shifted from Geochronology to annealing investigations of heavy ion tracks in minerals. Sandhu was instrumental in studying anisotropy of Heavy Ion tracks in mineral SSNTDs after his paper "*Anisotropic etching and annealing studies of fission tracks in quartz*" published in Mineralogical Journal of Japan. Our choice of Journals was based on two factors i.e. submission to journals which accept papers without any publication charge, and where publication process is rapid. Thus, we had to compromise quality with our convenience, and as a consequence, our publications did not catch the attention of leading groups in SSNTDs. Hence, our original contributions were ignored and our reputation was not established in the field for long.

Gurinder Singh took initiative to study Heavy Ion radiation damage effects in glasses using optical absorption spectroscopy. He obtained a modified Best Fit Model of radiation damage annealing in glasses which was an improvement of Modgil-Virk formulation. This formula was tested for annealing experiments in soda-lime, phosphate and silicate glass detectors to corroborate the concept of a single activation energy in the annealing of radiation damage in glass (SSNTDs). Our paper "*Annealing characteristics of nuclear tracks in glass detectors using optical absorption spectroscopy*" was published in Journal of Radioanalytical and Nuclear Chemistry in 1994. On the other side of the fence, Rajinder Kumar (R.K) Bhatia working on Heavy Ion radiation damage annealing in polymers challenged the formulation of Berkeley group and obtained an equivalent version of single activation energy model of Modgil-Virk. An inter-comparison and experimental verification of all these approaches has been reported in my review paper "*Modgil-Virk Formulation of Single Activation Energy Model of Radiation Damage Annealing in SSNTDs: A Critical Appraisal*" published in Solid State Phenomena Series, Trans Tech Publications (2015).

Without going into the rigorous approach, I may like to introduce the special features of Modgil-Virk formulation of radiation damage in SSNTDs. Annealing experiments were carried out to study the dependence of track annealing rate, $V_a$ ($V_a$ = dl/dt or dD/dt), on the temperature and time of annealing. The experimental results prove the exponential dependence of $V_a$ on temperature and a power law type of variation with annealing time. The empirical formulation of this model relates track annealing rate, $V_a$, explicitly with time and temperature as follows:

$$V_a = At_a^{-n} \exp(-E_a/kT) \qquad (1)$$

where both A and n are ion-dependent constants and $E_a$, the activation energy, is a unique parameter for a given SSNTD. The special features of this model are:

(i) It predicts single activation energy of annealing for all heavy ions and fission fragment tracks as required by the Arrhenius equation.
(ii) It may be used for revealing thermal history of track-recording SSNTDs (minerals, meteorites and lunar rocks), as annealing rate is given explicitly in terms of both time and temperature.
(iii) It explains the partial fading of tracks due to environmental annealing as track length is used as a parameter in place of track density.
(iv) It has universal application. Its validity has been tested for all types of SSNTDs (both crystalline and amorphous) using a variety of heavy ion-beams and fission fragments.

It is unfortunate that Modgil-Virk formulation for Single Activation Energy Model has not been exploited by the research workers to the extent we wished for it. The obvious reasons may be lack of theoretical basis of our empirical formulation as well as lack of publicity. Instead of support, the theoretical group of GNDU Amritsar were involved in a conspiracy in pulling down this Model on the basis of dimensionality problem of its equation (1). However, Prof. Jayant Narlikar, Director IUCAA, Pune came to our rescue and suggested a way out of this predicament. I am pleased to report that a group in PINSTECH (Pakistan) has corroborated our Single Activation Energy Model of radiation damage in SSNTDs in their publication "*Activation energy for the annealing of nuclear tracks in SSNTDs*" published in Nuclear Instruments and Methods in Physics Research B (2001). Their conclusions are as follows:

(i) Activation energy of annealing of nuclear tracks is a characteristic property of the detector material, and
(ii) Single activation energy concept is a good approximation, are identical to what we proposed in our model.

Heavy ions was used as a micro-structuring tool by Reimer Spohr. During his visit to GNDU Amritsar, he left a copy of his unpublished manuscript in our laboratory. When DST project "*Heavy Ion Radiation Effects in Insulators*" was sanctioned, our research team of Gurpartap Singh Randhawa and Sanjit Amrita Kaur was encouraged by me to follow in the footsteps of Reimer Spohr to develop micro-devices using heavy ion irradiation of polymers and muscovite mica. Ion track filters (ITFs) were prepared and used for environmental pollution studies and filtration of cancer blood cells of cancer patients. We had collaborated with group of S.K. Chakarvarty in NIT Kurukshetra and Sanjit Amrita got her initial training in fabrication of ITFs. We were not aware that application of ITFs can prove to be a stepping stone to Nanotechnology of future in India. It was Prof. Brandt of Marburg University (Germany), examiner of Sanjit's Ph.D. thesis, who remarked that this work can lead to development of new technologies in India. We published nearly a dozen papers highlighting our contribution. The following two, published in 1998, need to be mentioned in this regard: "*Effects on insulators of swift-heavy-ions radiation: Ion track technology*" published in Journal of Physics D (Applied Physics), and "*Ion Track Filters: Properties, Development and Applications*" published in Current Science.

Gurpartap Randhawa undertook the study of Heavy Ion ranges and stopping power in SSNTDs and his paper "*Stopping power and range of heavy ions in solids: A comparative study*" was a classic one, which was published in Radiation Measurements (1996) and presented as an Invited Talk by me in the opening session of 3rd International Conference on Material Science Applications of Ion Beam Techniques held at Seeheim, Germany in Sept. 1997. Randhawa was a proficient researcher who got training in Atomic Force Microscopy in CSIO Chandigarh, using this prototype model for the study of radiation damage effects in Insulators (SSNTDs). When R.K. Jain and Ajit Kumar Srivastava from BHU Varanasi joined as Post Doctoral Fellows (PDFs) in our group, we ventured into more diversified areas such as "*Swift heavy ion beam induced modifications in polymers*" using IUAC beams. In addition to physical and chemical property changes, optical and electrical response of Heavy Ion irradiated polymers (PVDF, PET, Kapton-H, PMMA, CR-39) was investigated using UV, Vis and FTIR spectroscopy. Paramdeep Singh Chandi was a great help in analysing data of physical and electrical parameters.

After my retirement from GNDU Amritsar in June 2002, I started my collaboration with Rajesh Kumar of AMU Aligarh, presently working in Guru Gobind Singh Indraprastha University, New Delhi. I donated my Heavy Ion irradiated polymer samples along with some unpublished data which was re-investigated by Rajesh and his brilliant research scholar, Paramjit Singh. This collaboration resulted in nine papers in area of polymer research, such as "*Study of optical band gap and carbon cluster sizes formed in 100 MeV $Si^{8+}$ and 145 MeV $Ne^{6+}$ ion irradiated Polypropylene Polymer*" published in Indian Journal of Physics (2009). I may remark in passing that my collaborators in GNDU Amritsar proved to be ungrateful after my retirement and never bothered to acknowledge the guidance or help in their research activity but my collaborators from AMU Aligarh, my alma-mater, never let me down. I learnt many lessons of human frailty during my research career of nearly forty years!

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