STATUS OF RADIATION SOLID STATE PHYSICS RESEARCH AT THE INSTITUTE OF NUCLEAR PHYSICS *)

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1. The history of radiation physics of solids began about 200 years ago with Wallestone's observation of thermoluminescence in natural minerals containing radioactive impurities and Pirsol's discovery of reversible coloration and thermoluminescence of CaF_2 at UV wavelengths. Just a century ago, Becquerel observed the coloration of KCl under exposure to electron beam and X-rays, while Roentgen and Ioffe discovered photo-induced conductivity in irradiated crystals. A bit later, Ramsay found an analogy between electrolysis and radiolysis. And only 50 years later did Pekar, Varley, Shatalov, Molner and Scott formulate the conception of atomic vacancy and interstitial atoms as element radiation defects in the simplest cubic crystal lattice, and also considered ionization and elastic mechanisms for the production of these defects.

2. The Department of Radiation Solid State Physics was established at the Institute of Nuclear Physics (INP) of the Uzbek Academy of Sciences more than 40 years ago by famous scientists I. Kurchatov and S. Starodubtsev, when reactor building and radiation material science was still very young. The department was to do both fundamental research of defect production/evolution and practical material science for atomic power stations, space and radiation technology, etc. Research laboratories and groups were organized by material science disciplines: semiconductors, dielectrics, metals and alloys, polymers, although they all had common problems and tasks to solve, such as:

- identification of as-grown (intrinsic and impurity) and radiation-induced defects;
- elucidation of dominant mechanisms (elastic or inelastic) of radiation damage;
- relaxation of defect structure and electron sub-system under various kinds of irradiation,
- determination of the conditions for radiation stability and durability of materials for nuclear reactors, lasers and luminophores.

Most of these studies were carried out in close collaboration with leading scientific and technological centers in the former Soviet Union.

One should note that the first theoretical models [Dins, Winyard] and experiments [Corbet] on radiation-stimulated hetero- and self-diffusion in Si, CdS, as well as sub-threshold defect production in semiconductors [Hudges] and, later on, in dielectrics were developed here within the decade 1960-1970. It was a successful competition with the related scientific centers of the USA, France and Japan. It was the initiative of former director Ulug Gulyamov to organize a special research group for reactor material science and build up the unique equipment for *in-situ* mechanical testing of fuel assemblies (UO_2 in ZrO_2 or steel cladding).

3. The nuclear-physical facility and equipment of the INP was contemporary (some even unique) and provided researchers with wide possibilities and opportunities for doing experiments in the course of irradiation. Now we have the 10 MWt WWR-SM reactor with ten vertical and horizontal channels; a 14 MeV neutron generator; a cyclotron that can accelerate protons up to 18 MeV and alpha-particles up to 36 MeV with a beam current up to 1 mA/cm^2; a few ~1.25 MeV ^{60}Co gamma ray sources, each capable of delivering 1-2000 R/s; and a special 250 keV co-axial electron accelerator capable of delivering a beam current up 5 mA. The facilities also support in-situ and ex-situ temperature measurements over a wide range, 77-1200 K, for investigations of optical, electrical and mechanical properties of different materials.

4. There is one particular capability that has been developed and featured at INP: the ability to distinguish radiation-induced and radiation-stimulated transitions between stable and unstable

structure states by means of combined and sequential irradiation with different particles and electromagnetic quanta at variable temperatures, doses and energies. Such an approach allows one to experimentally solve two actual problems: I) To separate contributions by energy losses from elastic collisions of projectiles with lattice atoms and ionization processes. The problem was often ignored, and now attracts many researchers, even those among metallurgists. II) To take into account a noticeable contribution from unstable radiation defects when estimating the radiation stability of any material.

5. With developing space technologies (satellites, space stations and ships) in the 1970's, much higher requirements were expected of materials, including semiconductors, optical and laser associated materials, electro- and heat-insulating ceramics, polymers and composites. Almost all components of space radiation around the Earth and within the solar system and their combined influence on materials over a wide temperature range (4-1000 K) could be simulated on the ground for testing materials before designing and making real apparatus. Just about all laboratories led by Generalova, Vakhidov, Niyazova, Yunusov, M. Muminov and Makhkamov applied their efforts to solving space material science problems of the day. Our experimental results and recommendations were used for a wide range of problems, including the manufacture of radiation hard glass for illuminators, laser elements, aboard electronics, electric wires and insulation, construction materials, among others.

6. The main working parameters characterizing irradiated materials were studied together with structure analysis (via neutron and X-ray diffraction techniques). The initial defect character and mechanical strain of structure are crucial for radiation response, and such a correlation allows choice of the proper growth conditions resulting either in radiation hardness or radiation sensitivity in the materials.

7. One of the main expected results of radiation material science is the identification of impurity centers, their charge and energy, form and distribution over the crystal lattice with the use of electric, magnetic, optical and kinetic methods, as most impurities determine the behavior of materials under and after irradiation. For example, impurities of the Au and Pt groups, forming deep levels in p-Si(B) and n-Si(P), result in increasing photo, magneto- or thermal-sensitivity as well as their fast response $\sim 10^{-8}$ s. Doping with Ce, Ti-Cr, Nd-Cr, Yb provide optical and laser materials with radiation hardness, while Fe, Ni, Mn, W, are responsible for radiation coloration and deterioration of laser generation. But, what is bad for lasers and optical windows can be good for radiation detectors. Thus, fused quartz glass and fluorite crystals, which become colored under radiation and luminescent at post-irradiation heating, are suggested as dosimeters of gamma-neutron radiations [Tavshunskii, Vakhidov, Abdukadyrova, Gulamova]. Just the opposite occurs with impurities Eu and Yb in $Ca(Ba,Sr)F_2$ and Mn in MgF_2, causing an increase of radiation hardness. Moreover, Mn in BaFCl increases both radiation stability and light output of radioluminescence [Nuritdinov]. Impurities were found to increase/decrease the rate and efficiency of generation and recombination of defects, owing to decrease/increase of the bound strength with lattice anions and hence decrease/increase of the recombination barrier.

In order to determine which impurities to use, and the methods for inserting them into a crystal lattice, non-destructive activation techniques such as autoradiography and X-ray fluorescence were explored [Vakhidov, Gasanov, Flitsiyan, Mukhamedshina, Gafitulina]. Distributions of Au in Si, transition metals in quartz and diamond were studied in this way.

8. Radiation induced recharging of impurity centers and native structure defects has allowed their visualization in the optical absorption spectra of most oxides and explains the linear growth of the induced absorption at low ionization doses. The following saturation level and the related irradiation dose were found to depend on the concentration and distribution of the centers and defects. Vakhidov and Blinkova have demonstrated such dependencies in silica blocks, tubes and fibers having different degrees of structure disorder and mechanical strains.

9. Radiation induced conductivity of ceramic dielectrics [Muminov, Skripnikov, Kim Gen Chan, Mukhamedjanov], and Ce-doped silica glasses [Gulamova, Alimov] had been studied in the course of gamma-irradiation. The conductivity was shown to proceed either on anion vacancies or interstitials (including alkali metals and hydrogen), depending on the irradiation conditions (temperature, dose, intensity).

10. Combining irradiation with choosing impurities to create either electron or hole trapping centers and varying the temperature, it became possible to distinguish among different electronic processes, such as self-trapping holes, radiative and non-radiative charge recombination, as well as charge localization of impurities or defects by spectra of gamma-luminescence and gamma-stimulated conductivity. Excitonic luminescence at low temperatures and exciton-impurity energy transfer followed by Ce, Eu, Nd, Cr impurity luminescence, are effective channels of releasing the absorbed gamma-quanta energy in the course of electronic subsystem relaxation. These processes prevent the generation of Frenkel pairs (vacancies F - interstitials I). Self-localized hole states have a low activation energy (~ meV) but a high oscillation strength, which cause a high absorption intensity in the visible and near IR ranges. Taking into account this absorption allowed to correct the non-stable optical losses and explain the observed low radiation hardness of most optical and laser materials at low temperatures (77-200 K) in ionizing radiation fields [Ibragimova, Rakov, Nuritdinov].

11. It was found that radiation-induced and radiation-stimulated generation of intrinsic defects by atomic displacements in oxides began after the saturation stage at higher doses. Dose and defect growth rate were determined by the degree of native defects (already existed impurities, vacancies, interstitials, dislocations and/or disordered regions). Atomic displacements could be induced at very high gamma-doses ($>10^9$ R) either in perfect and pure single crystals or in those doped only with Ce or Sm, and stimulated at minimal doses ($<10^4$ R) in deteriorated glasses [Nuritdinov], strained tubes [Blinkova] and stretched optical fibers with a lot of ruptured Si-O bonds, mostly at the surface and interfaces [Ibragimova].

It was proved for the first time in 1981 that the ionization of oxygen defect production plays a dominant role in oxides [Ibragimova, Nuritdinov], as in the case of alkali halides [Luschik, Itoh] and fluorides [Feofilov]. The non-radiative decay of electronic excitation into Frenkel pairs can be suppressed by radiative excitonic relaxation, which transfers its energy to several luminescent impurities, thereby providing radiation optical hardness [Ibragimova].

12. A study of radiation defect production, together with radiation-stimulated diffusion, has allowed the observations of the interactions of intrinsic defects (especially, mobile interstitials or vacancies) and impurity atoms followed by formation of stable defect complexes or aggregates at 600-1200 K. Irradiation changes the p-n transition geometry and defect concentration, which results, in some cases, an increase in the sensitivity and fast-reading of devices [Niyazova, Koyfman, Yunusov, Makhkamov, Karimov]. Consider these two examples: 1) radiation diffusion of OH in LiF creates aggregate centers composed of anion di-vacancies which enable frequency-amplitude modulation of lasing, while tri-vacancies deteriorate this modulation [Ibragimova]; 2) out-diffusion (removal) of Na from SiO_2 results in additional hardening of the crystal lattice [Gasanov]. Radiation enhanced oxygen diffusion in high temperature superconductors $YBa_2Cu_3O_{7-x}$ at low doses causes oxygen ordering in intergrain contacts and hence lowering the normal state resistivity, and at high doses the radiation effect is just the opposite [Gasanov, Ibragimova]. The ortho-tetra-phase transition was not found in this superconductor under gamma-irradiation because of the equilibrium between the absorption and desorption of oxygen [Kalanov].

13. Structure-phase transitions of order-disorder type have been systematically studied in different alloys (both insertion and substitution): hydrogen containing metals, silicon and SiO_2 [Oem, Khidirov, Kalanov]. Hydrogen was found to form different kinds of sublattices; while Si crystal blocks contain small SiO_2 inclusions with radial distribution, which correlates with

resistivity and other working parameters. It was found at first in experiment, and then theoretically confirmed, that long-term gamma+neutron irradiation of quartz results in the formation of small precipitates of the metamict phase. A similar radiation induced mechanism causes formation of diamond precipitates in graphite [Gasanov, Ibragimov]. Thus, radiation induced phase transformations (quartz-silica glass, graphite-diamond) allows modification of structure and obtain valuable properties for different applications.

14. Particular attention has been paid to developing radiation technologies destined for improving or creating new properties. Consider the following: 1) transmutation of silicon under neutron+gamma irradiation [Yunusov, Karimov]. 2) the manufacture of p- or n-diffusion Si structures by means of 6 MeV electron irradiation [Makhkamov]. 3) thermoradiation treatments, as well as sintering and synthesis of crystals, ceramics and glass under gamma-irradiation [Gasanov, Isaev, Nuritdinov, Ibragimova]. Different courses of irradiation accompanied by heating or followed by annealing has led to the following improvements and modifications of material properties: change of the optical transparency window [Tavshunskii, Kim Gen Chan]; an increase in radiation sensitivity and durability of dosimeters and detectors [Abdukadyrova, Gulamova] or radiation hardness of working parameters of solar cells [Makhkamov] and laser elements [Ashurov, Bedilov, Ibragimova]; an increase in the fast-reading or shortening of the response by semiconducting devices [Yunusov, Karimov, Makhkamov] and scintillators [Kim Gen Chan, Ibragimova, Gafarov]; to make noble coloration to semiprecious crystals [Vakhidov, Gasanov]; enhanced structure stability and mechanical toughness of alloys, polymers, ceramics and metal-ceramic contacts [Oem, Kalanov, Khidirov, M. Muminov, U. Gafurov]; an increase in critical current and magnetization of high temperature superconductors [Gasanov, Ibragimova]; an increase in the break-down voltage and decrease of dielectric losses of insulators [M. Muminov, Mukhamedjanov].

15. Parallel to the above-mentioned research, groups of theorists have considered different radiation processes based on the main ideas of solid state physics and the interactions of nuclear particles and ionizing radiation with solids. Kinetic and parametric equations were solved, and expressions of phenomenologic dependencies were suggested for elastic and inelastic defect production processes in simple semiconductors [Kiv, Umarova, Oxengendler], ionic crystals and simple oxides [Yarkulov, Jumanov, Ibragimov]. The radiolysis mechanisms were elaborated for electro-insulating polymers and organic liquids [Lenchenko]. Relaxation of electronic states and photoconductivity, and also radiation enhanced diffusion of impurities and intrinsic defects, were studied theoretically in Si and CdS [Oxengendler, Umarova]. Later, there were considered the radiation induced structure-phase transformations, amorphization and recrystallization of several oxides [Ibragimov], atomic disorder in ceramics [Skripnikov], amorphization and structure ordering of metal alloys [Pressman, Soldatov]. A hypothesis was advanced to explain radiation hardness and noise in non-crystalline materials and multiphase ceramics [Oxengendler, Skripnikov].

Recently, owing to the appearance of powerful computers, it has become possible to develop modern theoretical approaches such as the following: computer simulation of structure defects and modeling of radiation processes in metals [Gurvich]; quantum-chemical modeling of radiation processes in semiconductors [Khakimov, Umarova]; molecular models describing the formation of kink-defects, self-diffusion and destruction of molecular chains in polymers [Gafurov], and microscopic two-step scenarios to explain non-gap superconductivity in Fermi-Bose-liquids [Jumanov].

16. All this radiation research and technology has obvious yet real and positive practical applications for technological, economic and social development:

- unique crystals (blue topaz, ametist, smoky quartz, diamond like zirconia both white and colored) for jewelry yet harmless when worn;

- valuable luminophore BaFCl for medical tomography; and radiation-hardened active elements for powerful lasers (YAG, YAL, LiF) capable of working in outer space;
- optical filters with widened transparency window;
- semiconducting structures and fast-reading devices for electronics and microelectronics;
- hard infrared radiation detectors and solar elements;
- detectors and dosimeters of nuclear and ionizing radiation based on semiconductors and dielectric materials for nuclear monitoring and safety;
- radiation resistant hermetic pressure lead-ins and insulators for nuclear power plants;
- high temperature superconductors with augmented critical parameters (transport and magnetization currents) for the coming generation of energy materials.

The main conclusion to this review is to emphasize that natural radiation can be a very helpful servant, BUT one should never let it be human's master.

One can see below a selected and chronological list of publications from the Institute of Nuclear Physics.

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