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Role of Neutron Beam Applications in the Sustainable Socio-Economic Development of Pakistan

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A B S T R A C T

Pakistan Institute of Nuclear Science and Technology (PINSTECH) is the largest multidisciplinary research and development center in Pakistan. It has two research reactors namely PARR-1 and PARR-2. Neutron beam facilities have been installed only around PARR-1. It is a multipurpose research reactor that has been mostly utilized for the production of radioisotope and the training of scientists, engineers and technicians. The reactor is also utilized in the studies of neutron reaction cross-sections, nuclear structure, fission physics, radiation damage in crystals and semi-conductors, studies of geological, biological and environmental samples by neutron activation techniques, gemstone coloration, neutron radiography and neutron scattering. We have been investigating lattice dynamics, crystal and magnetic structure of materials and residual stress measurement using neutron scattering. In this manner, PARR-1 is effectively contributing to the socio-economic development of Pakistan.

Keywords: *Research reactors, neutron beam, neutron scattering, materials testing.*

1. Introduction

Realizing the importance of nuclear technology and its potential applications in various fields, the Pakistan Atomic Energy Commission (PAEC) was formed in 1956. Since its establishment, PAEC has made considerable efforts for the socio-economic development of the country through the peaceful use of nuclear technology. In 1965, the first nuclear research reactor and in 1972 the first nuclear power plant was established in Islamabad and Karachi, respectively. The research reactor was installed at the Pakistan Institute of Nuclear Science and Technology (PINSTECH) and named Pakistan Research Reactor-1 (PARR-1). Later another reactor PARR-2 was installed at PINSTECH. PARR-1 is a 10 MW multipurpose research reactor whereas PARR-2 is a 30 kW low-power miniature neutron source reactor (MNSR).

PARR-1 is a swimming pool MTR fuel-type reactor that became critical in 1966. In 1990, it was upgraded from 5 MW to 10 MW and its fuel was also converted from high-enriched uranium (HEU) to low-enriched uranium (LEU). PARR-1 is a medium flux facility with a maximum neutron flux of 1.5×10^{14} $ncm^{-2}s^{-1}$ at the core. It has six radials and one tangential through neutron beam tubes. Three out of six neutron beam tubes are dedicated to neutron scattering instruments. The other two neutron beam tubes have neutron activation analysis (NAA) and neutron radiography (NR) facilities whereas one beam tube could not be utilized due to space limitations.

2. Neutron Beam Applications

Neutron beams are produced either in constant wavelength sources which are research reactors (RRs) or in pulsed sources such as spallation sources. Neutron beams have numerous applications in the fields of materials science, natural sciences, life sciences, earth sciences, industry and medicine such as isotope production and boron neutron capture therapy (BCNT). Applications of neutron beams in these areas are effectively utilized in research and development, which constructively contribute to the improvement of quality of life, societal well-being and consequently, in the overall socio-economic development of the country.

2.1. Industrial Applications

2.1.1. Neutron Transmutation Doping

The neutron transmutation, which is initiated by interaction with neutrons, changes the nucleus to another or multiple nuclides through a nuclear reaction. The resulting unstable nucleus state undergoes some processes to become stable, which has different properties as compared to the original one [1]. Doping is purposely introducing impurity atoms into the material to obtain the desired properties. Therefore, neutron transmutation doping (NTD) is a process to create impurities in the intrinsic or extrinsic semiconductor by neutron irradiation to improve its electrical properties. There are several candidate materials for NTD such as Si, Ge, GaAs, GaN, GaP, InP, InSe and HgCdTe [1]. However, commercial scale NTD is only viable with Si due to several reasons: high quality single crystal ingots are available in the market, extremely uniform and ease of doping and great demand for doped Si in power devices and sensors [2, 3].

In NTD, the n-type semiconductor is produced by the conversion of ^{30}Si to ^{31}P by the following reaction [4]:

$$\text{Si-30}(n,\gamma)\text{Si-31} \longrightarrow \text{P-31}$$

The total cost to develop an NTD facility can reach half a million dollars, which includes the installation of irradiation rigs, handling equipment and QC instruments [2]. Si ingots are brittle therefore, careful handling before and after irradiation is required. In RRs, having NTD facility storage area of at least one ton of Si ingot should be available and in order to clean the irradiated ingot, an ultrasonic bath connected to radioactive waste water tanks is necessary [4].

Thermal neutrons are required for NTD as defects produced by fast neutron irradiation impair the electrical properties, especially at high resistivity values [1]. Therefore, thermal neutron density along the central axis is crucial and typically thermal neutron flux ~ $5 \times 10^{12} - 10^{13}$ $cm^{-2}s^{-1}$ to obtain resistivity

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of 200 Ω.cm along with a high Cd ratio is desirable. For instance, Si ingot irradiated for 17 hours at 10^{13} $cm^{-2}s^{-1}$ thermal neutron flux can produce 50 Ω.cm resistivity and large facilities with a flux >10^{14} $cm^{-2}s^{-1}$ can irradiate 10-15 tons of Si ingots [1]. The time required for irradiation depends on several factors such as the choice of an appropriate irradiation position, neutronic and thermohydraulic calculations, design of the irradiation rig, calibration measurements to obtain the desired parameters and trained manpower [2].

In the early 2010s, 120-150 tons of Si ingot were doped in RRs at the cost of US $ 70-100 /kg. It is estimated if annual production of hybrid vehicles reaches 50 million by 2030 this demand will rise to 2000 tons per annum [1].

2.1.2. Gemstone Coloration

The majority of natural gems are single crystals of naturally occurring minerals, but some are amorphous (some types of opal and natural glass), others are solid solutions (e.g., garnets and peridot), some are rocks (jade and lapis etc.) and some are made entirely or mostly of organic materials (such as amber, pearls, coral) [5]. The price of a gemstone depends on several factors: color, luminosity and durability. The value of the gemstone can be enhanced with coloration. Different techniques such as neutron irradiation, ion beam and laser beam are used for gemstone coloration. Localized heating is required for inducing color in gems; however, laser beams do not produce localized heating, whereas low-mass ions produce localized heating but result in poor color [6]. Blue coloring in the colorless gemstones topaz and diamond is possible with neutron irradiation and coloration can enhance 30 times the natural value of these gemstones [1]. Gemstones are irradiated with fast neutrons therefore; the container can be shielded with B or Cd. The aluminum container holding up to 2 kg of gemstones is used for irradiation and placed in either the grid position or in the beam tube [1]. Fluence of 10^{17} - 10^{18} fast neutrons is desirable which is equivalent to an irradiation time 50-100 hours in 2MW RR. The temperature of gemstones typically varies between 100-150 °C during irradiation but damage can occur in topaz as discoloration if the temperature soars above 300 °C [2]. In some cases, 70% stones can be transported after 2 months of cooling time but β-emission should be checked with a multi-channel analyzer before releasing gemstones to ensure the dose is not more than the acceptable value of 2 nCi/g [1].

The gemstone market is volatile and difficult to access due to unpredictable trends in the fashion industry. The behavior of gemstone dealers is also erratic, primarily due to the long radiological cooling time of gemstones, which puts their investment at stake. Gemstone's market worth was US$ 14.34 billion in 2021 [7]. Last year, Pakistan exported US$ 13.73 million, which is only 0.09% of the global share [8]. Pakistan can increase its share in the gemstone market as we can exploit the natural reserves of topaz, which are found mostly in Gilgit-Baltistan [9].

2.1.3. Residual Stress Measurement

Residual stress is one of the primary factors influencing the mechanical characteristics of a material, such as strength, plasticity and surface integrity. For instance, tensile stress conditions can severely reduce component life, while compressive stress conditions can improve material performance under fatigue [10]. Therefore, the distribution of residual stress can predict the service life of the materials. Residual stress is produced in materials during fabrication processes such as plastic deformation and heat treatment. Residual stress can impair the mechanical properties and the service life of the material is reduced. Therefore, knowledge of the residual stress is extremely important to improve the thermo-mechanical processing of metals and alloys, thereby increasing the service life of a material. This technique is also utilized in the inspection of the reliability of weld and post-weld heat treatment in the high-tech industries such as the nuclear industry.

At the microscopic scale, residual stress changes the distance between atomic planes which is reflected in the shift of Bragg reflection. This change in the lattice plane spacing "*d*" can be accurately measured with neutron diffraction (ND) and X-ray diffraction (XRD). In Fig. 1A, variation of peak (020) as a result of change in *d* spacing of different brass samples can be easily seen. B2 was annealed at 200 °C, B3 at 300 °C and so on. The corresponding residual stress is presented as a function of annealing temperature in Fig. 1B.

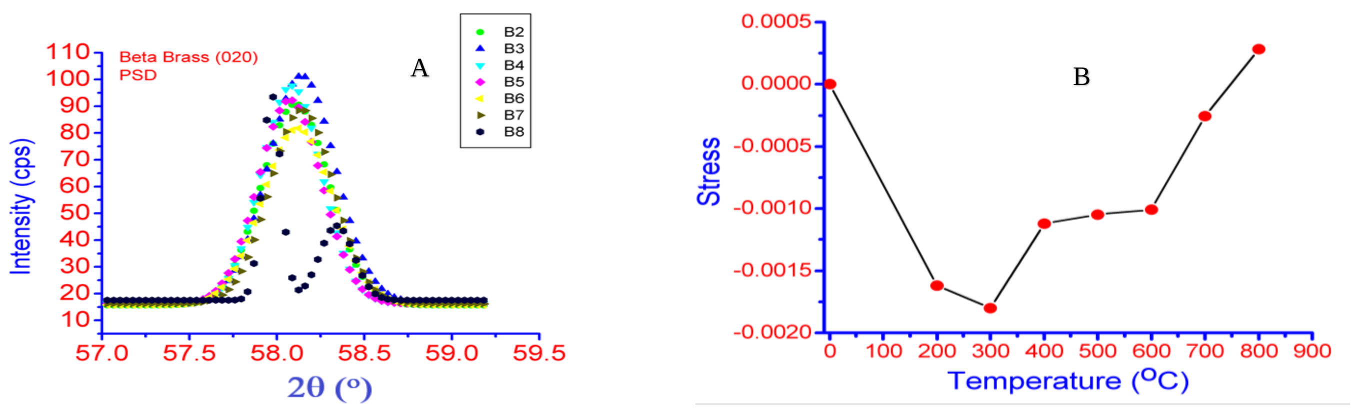

Fig. 1: (A) Change in the 2θ position of Bragg peaks in brass sample measured at different temperatures, (B) calculated residual stress as a function of temperature.

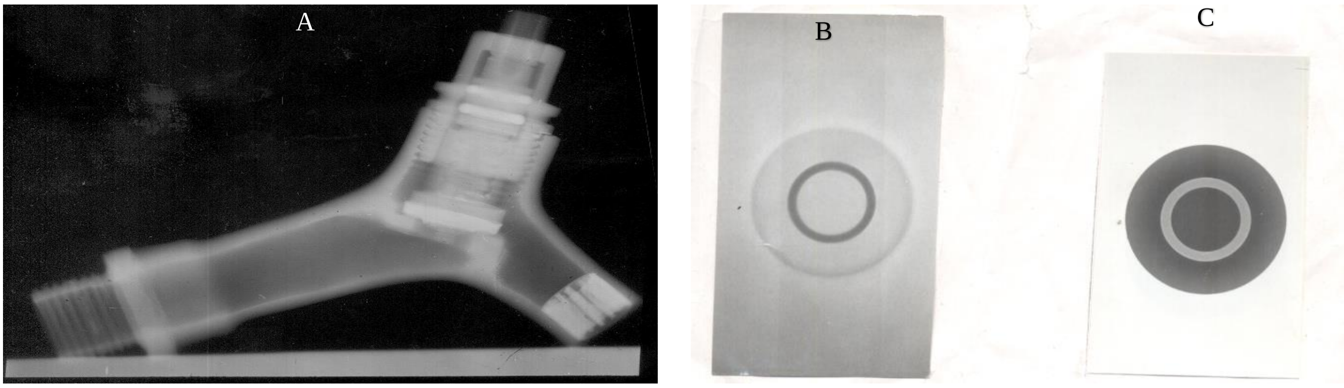

Fig. 2: Neutron radiography image of (A) water tap, (B) O-ring in grooved aluminum, (C) X-ray radiograph of O-ring in grooved aluminum.

The ND technique has the advantage of large penetration depth due to charge neutrality and small absorption of neutrons in most of the elements compared with XRD. Therefore, 3-D mapping of residual stress in the bulk of a component using a non-destructive technique is possible only with ND [11, 12].

2.1.4. Neutron Radiography

Neutron radiography (NR) is one of the most important non-destructive techniques that has applications in the fields of materials science and engineering, cultural heritage, geology, paleontology, etc. [13]. Neutrons' ability to see light elements, especially those surrounded by heavy elements makes NR superior to other techniques such as X-ray, ɤ-ray and ultrasonic testing. A variety of samples such as airplane turbine blades, oil in the engine, detonating fuse, plant root, irradiated fuel, metal fluid, boiling of high-pressure liquid, etc., can be tested with NR.

The NR setup is installed at beam tube 6 at PARR-1. Images of the water tap and O-ring are shown in Fig. 2. In Fig. 2A, the details inside the tap can be easily seen including metallic and polymeric components. Similarly, in Fig. 2B, the O-ring can be seen sitting in groves of aluminum, however, the X-ray image of the same object's O-ring is not visible (Fig. 1C).

2.2. Medical Applications

2.2.1. Radioisotope Production

A number of radioisotopes can be produced at PARR-1 on demand by medical centers that are spread across the country. Some of these radioisotopes are used to make freeze-dried kits at PINSTECH. Technetium generators, PAKGEN 99mTc, are regularly supplied to cancer hospitals all over the country. Last year revenue of more than 300 PKR million was generated from the production of radioisotopes and radiopharmaceuticals.

2.2.1. Boron Neutron Capture Therapy

^{10}B absorbs a neutron and emits α-particle by the following reaction:

$$n + {}^{10}B \longrightarrow {}^{4}He + {}^{6}Li$$

α-particle is highly ionizing, having a range in human tissue nearly equal to the diameter of a cell. Boron neutron capture therapy (BNCT) is to inject a tumor with a borated compound and irradiate it with either thermal or epithermal neutrons [2]. Surface or shallow tumors can be irradiated with thermal neutrons whereas deep tumors require epithermal neutrons. Therefore, the success of the technique is to produce better boron compounds for drug delivery. Other elements such as Gd-based compounds can also be used in this kind of therapy [2]. The reactors with powers ranging from 100 kW to 1 MW are specially adapted for BNCT [1]. Even a low-power, fast reactor of 5 kW can produce epithermal neutrons of the desired intensity [2]. Neutrons having energy 0.5 eV to 10 keV and flux 10^8-10^9 $ncm^{-2}s^{-1}$ are needed for BNCT. A filtered collimated beam free from fast neutrons and gamma rays, which can damage healthy tissues, is required for deep-seated tumors such as glioblastoma multiform. BNCT also demands the development of medical treatment protocols, which require approval from the government health authority.

2.3. Forensic Applications

2.3.1. Neutron Activation Analysis

Neutron activation analysis (NAA) is the simplest and wide application of RRs. NAA can be performed almost in any reactor, even with a power of only a few kW, which can generate a neutron flux of 10^{11} $cm^{-2}s^{-1}$. It requires post-irradiation facilities and HPGe detector with a Compton suppression system. NAA lab can analyze 10 samples per day assuming two counting facilities. At large facilities, 4000 samples per year can be performed but only 10% are mostly utilized in their annual capacity [1].

NAA has non-destructive capability, which simultaneously provides quantitative analysis of multiple elements. This technique is well suited for trace element analysis and typical detection limits of some elements are in the sub-ppm range [14]. NAA is utilized in almost every area of science and technology including archeology, studies of air-polluting, aerosol particulates, sediment studies, sulfur in coal and sugar, boron in steel, uranium, thorium and potassium exploration in salt mines, cadmium in cigarettes, determination of toxic and essential trace elements in food [15]. On the basis of performance and expertise, the NAA laboratory in PINSTECH has been awarded Regional Resource Unit status for the South Asian Region by the IAEA.

2.3.2. Prompt Gamma Neutron Activation Analysis

Prompt gamma neutron activation analysis (PGNAA) setup is installed at beam-tube 2 at PARR-1. The advantages of PGNAA as compared to NAA are that the resulting product is stable and emits no intense gamma radiation. Further, isotopes with very short half-life or small isotopic abundance can easily be measured.

PGNAA can also perform a variety of analyses such as analysis of 316-L stainless steel, sediment studies, Cd in cigarettes and B analysis in steel [15]. Sulfur in coal from the Baluchistan coal mine has been studied with PGNAA at PINSTECH.

2.4. R&D applications

2.4.1. Neutron Scattering

Compared to other techniques, neutron scattering provides a distinct advantage for studying a wide range of materials. Thermal neutrons have a wavelength that is ideal for diffraction investigations or elastic scattering. Conversely, because the energies of phonons and neutrons are similar, lattice dynamics are analysed using inelastic scattering. Selective absorption and the absence of charge on neutrons allow for deep penetration into the materials to study their bulk properties [16]. Due to their distinct scattering lengths, neutrons can even distinguish between hydrogen and deuterium. Determining low- and high-density ice, as well as creating the extraordinarily intricate temperature and pressure phase diagram of water (H_2O-D_2O), would not have been possible without neutron scattering [17]. These advantages of neutrons also make them suitable for in-situ, operando and non-ambient measurements [18]. The magnetic moment of neutrons is another tool used to study magnetic structures of the magnetic materials. Some of these advantages of neutron scattering techniques are utilized at PARR-1.

2.4.1.1. Triple-axis diffractometer

The triple-axis diffractometer TKSN-400 displayed in Fig. 3, used for inelastic neutron scattering experiments, was installed in the early 1970s. This instrument has been extensively used for elastic scattering as well as inelastic scattering experiments. Instrumental and experimental details are provided elsewhere [11]. It has a bent Cu (220) single crystal as a monochromator and its wavelength is selectable. This diffractometer was later upgraded with IAEA technical and financial support. The up-gradation work included designing and developing of multicounter assembly consisting of 8 He-3 detectors with inter detector separation of 12° which covers the 2θ range of 5-110°, instrument control and data acquisition system. This instrument played a vital role in enhancing neutron scattering activities at PARR-1.

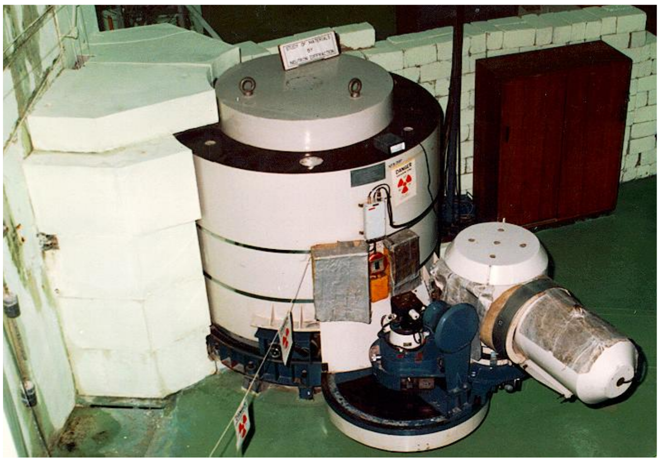

Fig. 3: Triple-axis diffractometer at beamtube 3, PARR-1.

2.4.1.2. Double-axis diffractometer

A high-resolution double-axis neutron powder diffractometer (NPD) for elastic scattering experiments is shown in Fig. 4. NPD was installed after the upgradation of PARR-1 from 5 to 10 MW. It has a Cu (220) single crystal as a monochromator which provides neutrons of 1.27Å wavelength. 30’:20’:10’ collimation produces $1x10^5$ $n.cm^{-2}.s^{-1}$ thermal neutron flux at the sample position. It has two types of detectors, a single BF_3 counter can cover the 2θ range of 5-125° and a position sensitive detector (PSD) has active area of 2.5cmx60cm with a 2θ range of 40° are installed on the NPD.

Fig. 4: PARR-1 double-axis diffractometer installed at beamtube 4.

This diffractometer has been used to study crystal structure by determining the accurate atomic positions of light elements as well as the ordering of elements in the unit cell and magnetic structure. The properties of the materials depend on their crystal structure. Therefore, accurate crystal structure determination is pivotal to both comprehend the properties and create novel materials with enhanced properties.

2.4.1.3. Small Angle Neutron Scattering

A small angle neutron scattering (SANS) instrument shown in Fig. 5 is installed at beamtube 5. Generally, a cold neutron source produces low-energy neutrons for SANS experiments. But SANS at PINSTECH is unique in the sense

Fig. 5: Small angle neutron scattering instrument installed at beam tube 5, PARR-1.

that it utilizes thermal neutrons instead of cold neutrons. In the early 1990s, the idea was proposed that thermal neutrons could also be used for SANS experiments [19]. The SANS instrument has two monolithic channel crystals, which decouple the resolution from the divergence of the incident beam. This decoupling improves angular resolution by two orders of magnitude and a divergent beam can generate sufficient neutron intensity for experiments. In our case, the flux at the sample position is ~$5x10^4$ $n.s^{-1}.cm^{-2}$. This instrument has a monochromator of perfect bent Si (111) crystal which gives a wavelength of 2.1 Å. It has a fully asymmetrically bent perfect crystal Si (111) as an analyzer having Q-resolution = 10^{-4} - 10^{-3} $Å^{-1}$ and Q-range = $2x10^{-4}$ - $2x10^{-2}$ $Å^{-1}$ and ~ 1 mm resolution 1-D PSD. The maximum sample size, which can be measured is $5x25mm^2$. SANS is used for studying aging and fatigue effects, precipitates, cracks, voids and other inhomogeneities in materials.

2.4.2. Materials Testing

Materials' testing is also one of the important neutron beam applications. Materials testing can be applied to nuclear fuels and nuclear reactors' structural components. The testing can be performed inside the core, at the periphery or at an external location using neutron beam guides.

2.4.2.1 Structural Components Testing

Structural components of nuclear reactors are usually irradiated with neutrons, especially fast neutrons to study their aging behavior and predict their service life under the neutron fluence. Fast neutrons produce defects in the materials, which not only impair their mechanical properties but also their corrosion resistance. Typically, neutron fluence of 10^{17} cm^{-2} is required for testing electronic components and organic gaskets whereas more than 10^{21} cm^{-2} is needed for testing metallic components [2].

2.4.2.2 Nuclear Fuel Testing

Nuclear fuel testing is performed with thermal neutrons and generally, two types of testing are carried out in RRs. The first type of testing is the aging of nuclear fuel in which fuel burn up is measured at steady-state power conditions. The second type of testing is carried out under power transient conditions to investigate fuel and cladding behavior. A specialized device is needed for this type of testing in order to control changes in the neutron flux on the fuel sample. The minimum amount of thermal neutron flux 10^{13} $cm^{-2}s^{-1}$ is typically required at the location of the specimen [2].

3. Conclusion

PINSTECH is playing its part along with other contributions in the field of neutron beam applications to achieve sustainable socio-economic development in Pakistan. Large-scale radioisotope production not only reduces import costs but also has the potential to generate significant income from exporting them to other nations. NTD and gemstone color can both produce a sizeable sum of foreign exchange. On the other side, residual stress measurement analysis can save huge costs by extending the service life of the materials, whereas R&D generally improves the overall socio-economic condition. There is a lot of potential in the field of neutron beam applications and more neutron beam applications should be explored.

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