Chapter 1: INTRODUCTION

1.1 General

In recent years, the number of reactors and high power particle accelerator facilities in operation, being commissioned, designed, or planned in the country has significantly increased. Prompt radiation field in the accelerators are dominated by neutrons and gamma radiation (in the case of positive ion accelerators) or by high energy bremsstrahlung radiation (photons) (for electron accelerators). In reactors the radiation field constitutes of neutrons, gamma radiation and fission fragments.

A shield is a physical entity placed between a source of ionizing radiation and an object to be protected in order to reduce the radiation level at the position of the object. Proper shielding of the radiation facilities is mandatory to provide a safe environment both to living and non-living world and to ensure optimum use of the beam time. It aims to:

(i) reduce the dose to personnel and environment during operation of the facility
(ii) reduce exposure to personnel from highly radioactive targets and other components, and
(iii) prevent contamination of the environment.

Over decades concrete has proved to be the most suitable material for bulk shielding. Shield design in accelerators depends crucially on the concrete composition, because elemental and isotopic composition of the concrete determines its shielding efficiency, strength and durability. It has a strong influence on the cost incurred.

Compositions of concrete used in different parts of India vary significantly, but for all of them the required thickness for effective radiation shielding in high power accelerators is large, which involves a huge cost. Self-compacting concrete (SCC) is one of the preferred
choices for cost-effective shield in these facilities. In SCC, self-compaction ability of the mix ensures uniform pouring during construction, thereby avoiding formation of voids inside the shield wall at the initial stages of construction.

After the lifetime operation of a reactor or a particle accelerator, the concrete shield walls will possess significant amount of low level radioactivity due to nuclear reaction induced in it by neutrons and high energy gammas which constitutes the radiation field inside the facility during the lifetime of the accelerator. Disposing this active concrete in the public domain is environmentally hazardous and hence it needs special disposal provision. The type and level of radioactivity will, however, depend on the elemental composition of concrete and also on the type of accelerator. Estimation of radio-activation of concrete shields in reactor and accelerator facilities is very important during decommissioning (Masumoto, K., et al., 2003).

1.2 Radiation Shielding

Radiation shielding is based on the principle of attenuation, which is the ability to reduce the effect of a particle or ray by blocking or bouncing particles through a barrier material. Charged particles may be attenuated by losing energy to reactions with electrons in the barrier, while x-ray and gamma radiation are attenuated through photoemission, scattering, or pair production. Neutrons can be made less harmful through a combination of elastic and inelastic scattering, and most neutron barriers are constructed with materials that encourage these processes.

There are several factors that influence the selection and use of radioactive shielding materials. Considerations such as attenuation effectiveness, radioactivity production in the
shield due to exposure to radiation field, strength, resistance to damage, thermal properties, and cost efficiency can affect radiation protection in numerous ways. For example, metals are strong and resistant to radiation damage, but they undergo changes in their mechanical properties and degrade in certain ways from radiation exposure. The main types of radiation and their shielding are discussed below:

1.2.1 Alpha and Beta Shielding

Alpha particles are positively charged helium nuclei, and are relatively easy to block, while beta particles are positively or negatively charged electrons that are more difficult to shield against. While density remains an important characteristic for blocking alpha and beta radiation, thickness is less of a concern. A single centimetre of plastic is sufficient for shielding against high energy alpha particles, as is a half-inch of paper.

In some cases, lead is ineffective in stopping beta particles because they can produce secondary radiation when passing through elements with a high atomic number and density. Instead, plastic can be used to form an efficient barrier for dealing with high-energy beta radiation. When negatively charged beta particles hit a high-density material, such as tungsten, the electrons are blocked, but the target which the barrier is intended to protect can actually become irradiated.

1.2.2 Gamma and X-rays Shielding

Gamma and X-rays are forms of electromagnetic radiation that occur with higher energy levels than those displayed by ultraviolet or visible light. In most cases, high-density materials are more effective than low-density alternatives for blocking or reducing the intensity of such radiation. However, low-density materials can compensate for the disparity
with increased thickness, which is as significant as density in shielding applications. Lead is particularly well-suited for lessening the effect of gamma rays and x-rays due to its high atomic number. This number refers to the number of protons within an atom, so a lead atom has a relatively high number of protons along with a corresponding number of electrons. These electrons block many of the gamma and x-ray particles that try to pass through a lead barrier and the degree of protection can be compounded with thicker shielding barriers. However, it is important to remember that there is still potential for some rays making it through the shielding, and that an absolute barrier may not be possible in many situations.

1.2.3 Neutron Shielding

Neutrons are particles that have neither a positive nor a negative charge, and thus provide a wide range of energy and mass levels that must be blocked. Lead is quite ineffective for blocking neutron radiation, as neutrons are uncharged and can simply pass through dense materials. Materials composed of low atomic number elements are preferable for stopping this type of radiation because they have a higher probability of forming cross-sections that will interact with the neutrons. Hydrogen and hydrogen-based materials are well-suited for this task. Compounds with a high concentration of hydrogen atoms, such as water, form efficient neutron barriers in addition to being relatively inexpensive shielding substances. However, low density materials can emit gamma rays when blocking neutrons, meaning that neutron radiation shielding is most effective when it incorporates both high and low atomic number elements. The low-density material can disperse the neutrons through elastic scattering, while the high-density segments block the subsequent gamma rays produced in nuclear reaction.

The different types of radiation and their shielding materials are shown in Fig. 1.1.
1.3 Concrete as Radiation Shield

From the consideration of cost, and where space is not a constraint, conventional concrete of sufficient thickness can be used to construct a satisfactory biological shield. However, in many applications, space is a definite constraint. Hence, to provide adequate shielding, high density concrete using various heavy aggregates is employed. Natural minerals used as aggregates in high density concrete are hematite, magnetite, limonite, barite and some of the artificial aggregates include materials like steel punching and shot. While the density of normal concrete is in the range of 2300-2400 kg/m$^3$, high density concrete has a density upwards of 2600 kg/m$^3$. High density concrete used in Neutron Shielding has a high cement content and aggregates like hydrous ores to increase the percentage of hydrated water (Abo-El-Enein, S. A., et al., 2014).

During batching and mixing of ordinary as well as high density concrete it is important to avoid over-mixing due to the fragility of certain aggregates. To prevent segregation of coarse aggregates the slump should be kept low and over-vibration avoided. Mortar may be placed in layers of specific thickness over which a fixed quantity of coarse aggregate may be placed.
and vibrated into the mortar, a procedure which is called ‘puddling’. As far as curing is concerned there is no major difference from normal weight concrete.

1.3.1 Geology of Coarse Aggregate (CA) samples used in the work

Coarse Aggregate (CA) or rock chips make up the major portion of any concrete. To know more about any concrete it is very essential that one should know more about the CA. The depth and range of studies that are required to be made in respect of aggregate to understand their widely varying effects and influence on the properties of concrete cannot be underrated (Shetty, M. S., 2013).

The CA samples used in the preparation of SCC in the present work were procured from the twelve different major geological formations of the state of Karnataka. The locations for procurement of the CA samples were chosen on the basis of the Report on Geology of Karnataka, obtained from the Department of Mines & Geology, Government of Karnataka (Report No. DMG: SGU: GR: 2009-10:621).

As per the report on the Geology of Karnataka, the rock formations in the state can be broadly divided into the following seven groups, viz. (i) Sargur group (ii) Peninsular gneiss (iii) Dharwar group (iv) Younger granites (v) Proterozoic sediments (vi) Deccan Traps and (vii) Laterites. The geological identification of the rock types are: granites, gneisses, trap, basalts, sandstone, limestone, dolomite, quartzite and laterites, with one or more of these found in the above mentioned groups. A list of the rocks procured, along with the site of their locations is presented in Table 3.1.
1.3.2 Use of SCC at Nuclear Installations in India

From the point of view of employing SCC technology in the construction of nuclear facilities, the specific advantage it has over the normal vibrated concrete (NVC) is probably the ease with which the fresh SCC mix can flow and spread into forms having heavily congested reinforcement, all by itself, and without any need for external compaction. The structures in nuclear facilities are designed with higher safety factors considering the seismic loads. This results in higher percentages of steel reinforcement leading to congestion, especially at the column-beam junctions. To ensure proper placement of concrete under such circumstances the height of concrete pour needs to be restricted. This results in more construction joints and a consequent increase in construction time (Bapat, S. G., et al., 2005).

Keeping these aspects in view, full scale mock-ups and actual use of SCC at their Nuclear Power Plant at Kaiga in the state of Karnataka have been successfully undertaken and implemented by the Nuclear Power Corporation of India Ltd (NPCIL). SCC mix of grade M30 has been achieved and used at this plant with a cement content of about 225 kg/m³ (Bapat, S. G., et al., 2004).

SCC has also been successfully used in the concreting of walls and other structures at the Tarapur Atomic Power Project 3 and 4 (TAPP 3 & 4) of NPCIL in the state of Maharashtra. A volume of about 230 m³ of M40 grade SCC has been produced with a cement content of about 300 kg/m³ and successfully used at this site (Amit Mittal, et al., 2004).

1.3.3 Definitions of terms related to SCC

The following terms, often used in the context of SCC proportioning, may be defined as under (The European Project Group, 2005):

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• Filling ability: The ability of fresh concrete to flow into and fill all spaces within the formwork, under its own weight.

• Flowability: The ease of flow of fresh concrete when unconfined by formwork and/ or reinforcement.

• Paste: The fraction of the concrete comprising powder, water and air, plus admixture, if applicable.

• Powder (Fines): Material of particle size smaller than 0.125 mm.

• Passing ability: The ability of fresh concrete to flow through tight openings such as spaces between steel reinforcing bars without segregation or blocking.

• Robustness: The capacity of concrete to retain its fresh properties when small variations in the properties or quantities of the constituent materials occur.

• Segregation Resistance: The ability of concrete to remain homogeneous in composition while in fresh state.

• Slump-flow: The mean diameter of the spread of fresh concrete using a conventional slump cone.

• Thixotropy: The tendency of the fresh mix to progressive loss of fluidity when allowed to rest undisturbed but to regain its fluidity when energy is applied.

• Viscosity: The resistance to flow of the fresh mix once flow has started.

• Viscosity Modifying Admixture (VMA): Admixture added to fresh concrete to increase cohesion and segregation resistance.

• Workability: A measure of the ease by which fresh concrete can be placed and compacted: it is a complex combination of aspects of fluidity, cohesiveness, transportability, compactability and stickiness.
1.3.4 Advantages of using SCC

Properly proportioned and placed SCC can result in both economic and technological benefits for the end user. The cost savings, performance enhancements, or both, are the driving forces behind the use of SCC. Specifically, SCC can provide the following benefits: *(ACI 237R-07 Report, 2007).*

- Reduce the labour and equipment
- Eliminate the need for vibration to ensure proper consolidation
- Enable the casting of good quality concrete independent of the skill of the workers
- Accelerate the construction rate, which leads to saving of time
- Facilitate and expedite the filling of highly reinforced sections and complex formwork while ensuring good construction quality
- Enable more flexibility in concrete placing and spreading points during casting.
- Reduce noise on the job site and pave the way for ‘silent concrete.’
- Decrease employee injuries by facilitating a safer working environment
- Create smooth surfaces free of honeycombing and signs of bleeding and surface defects.

1.3.5 Induced radioactivity and Radiation transmission in concrete

Radioactivity is a phenomenon of disintegration of an unstable nucleus of an element resulting in emission of energy and a new element. This phenomenon exists in nature among heavier elements, i.e., elements having higher atomic number. However, isotopes of many lighter elements as well as stable heavy isotopes are rendered radioactive by bombardment with charged particles, neutrons or high energy photons. This occurs by the nuclear reaction initiated by the energetic incident particle on the nuclides of the medium. This is called artificial radioactivity. Henry Becquerel, a French physicist, discovered in the year 1896 that a compound of uranium emitted some invisible radiant energy. Madame Curie, a Polish and
naturalized-French physicist studied a large number of such substances and named this phenomenon as “Radioactivity”.

Radioactivity of a sample is quantified in terms of the Activity, $A$, which gives the average number of spontaneous nuclear disintegrations taking place in a radioactive material, per unit time. The SI unit of activity is Becquerel (Bq):

$$1 \text{ Bq} = 1 \text{ disintegration per second} = 1 \text{ dps}.$$ 

The old unit of activity is Curie (Ci).

$$1 \text{ Ci} = 3.7 \times 10^{10} \text{ disintegrations per second} = 3.7 \times 10^{10} \text{ Bq} = 37 \text{ GBq}$$

1 Bq is a small quantity for many applications, hence activity is generally expressed in Mega Bq (1 MBq = $10^6$ Bq) and Giga Bq (1 GBq = $10^9$ Bq).

Induced radioactivity in the concrete shield wall of a radiation facility is the artificial radioactivity produced in the concrete by nuclear reaction induced by the neutrons and / or high energy gamma rays that are produced during the accelerator / reactor operation. Induced activity in the concrete shield due to a given radioisotope is determined from the concentration of the corresponding element, neutron flux and the time during which the concrete shield is exposed to neutrons. As a result of generation of induced activity in the shield, structural decommissioning of the radiation facilities would produce radioactive concrete waste. Depending on the type of the reactor or accelerator hundreds of tons of radioactive concrete waste is produced (Thierfeldt, S., 2010; O’Sullivan, O., et al., 2010; and Choi, W.K., 2009). These wastes often contain long-lived radioactivity. Masumoto, K., et al., (2003), showed that in an accelerator environment $^{152}\text{Eu}$, $^{60}\text{Co}$, $^{134}\text{Cs}$, $^{22}\text{Na}$, $^{54}\text{Mn}$ are some of the important isotopes produced in concrete through neutron activation.
Transmission of neutrons and gammas through any material is governed by the exponential attenuation law. For an incident flux $I_0$ on the inner wall of the concrete shield in an accelerator or a reactor facility, the transmitted intensity outside the shield wall is given by

$$I = I_0 e^{-\mu x}$$

Where $x$ is the thickness of the shield and $\mu$ is the linear attenuation coefficient of concrete. $\mu$ depends on the elemental composition of the concrete used. Mass attenuation coefficient of concrete is given by $\mu_{mass} = \mu/\rho$. where $\rho$ is mass density of the shield material.

In order to optimize the shield design, to minimize residual radioactivity in different upcoming accelerator facilities and also to make them cost-effective, measurement and simulation of neutron dose transmission through, and activation of various variants of SCC needs to be carried out. This in turn also requires elemental analysis of the SCC constituents, and modifying them for better performance. In order to achieve this end, it becomes imperative to determine the neutron attenuation and radioactivity induced through the various constituents of SCC.

1.4 Motivation for carrying out the present work

At the end of the year 2011, about 124 nuclear power reactors were shut down throughout the world (Deju, R., et al., 2013). The decommissioning of nuclear facilities is a subject of growing importance in many of the International Atomic Energy Agency (IAEA) member nations (162 countries as of February 2014), due to the large number of facilities that have attained or soon will attain the end of their service life. As a result of decommissioning operations, a wide range and quantity of radioactive materials (including huge masses of concrete) will need to be managed. Some of these materials may be recycled or reused if they continue to have an economic value, but most must be managed as radioactive waste. Thus,
the development and implementation of appropriate strategies for processing and disposal of decommissioning waste has become an important issue (IAEA-TECDOC-1572, 2007).

Quantifying the extent to which irradiation will change the properties of concrete is close to impossible. This is because the quantification is dependent on many factors, such as variation of material properties, material state of testing, neutron energy spectrum, and neutron dose rate (Hielsdorf H., et al., 1978).

Research and Development of low activation concrete shield has been a thrust area in most countries, and in India it has been a continued endeavour of agencies related to the proliferation of nuclear energy and technology like the Department of Atomic Energy, to constantly look for shielding material that can extend the operational life of nuclear installations. It is in this backdrop that the present work gains importance and significance. This work is a part of the research project entitled, “Study of self-compacting concrete compositions for accelerator shielding”, funded by the Board of Research in Nuclear Sciences (BRNS), Department of Atomic Energy, Government of India, Sanction No. 2009/36/77-BRNS; dated 16-12-2009.

Researchers in India and across the world have worked on neutron activation and subsequent gamma spectroscopic analysis of various different materials of their interest, including soil and rock samples, for obtaining the trace element concentrations in the materials. There is considerable amount of literature available on these topics, and a comprehensive review of the same has been included in Chapter 2 of this thesis.
1.5 Research Gaps

There is practically no work carried out on the coarse aggregate (CA) (stone chips used in preparation of concrete) samples of the state of Karnataka for quantifying the trace elements present in them that give rise to long-lived radionuclides in the radioactive concrete waste, which would need to be handled/disposed of appropriately during the decommissioning of the nuclear facility.

The novelty factor of the present work, therefore, is that it not only documents the trace element concentrations, and radioactivity build-up in the CA samples procured from the twelve different geological formations of Karnataka, but also delves into the various aspects of producing low-activation self-compacting concrete prepared using the CA sample showing least radio-activation. Also, neutron transmission studies were carried out on the SCC slab specimens for obtaining the quantification of attenuation characteristics/properties of SCC mixes prepared using the CA samples of Karnataka.

1.6 Objectives of the work

- The primary objective of this work is to identify the appropriate coarse aggregates (CA) (stone chips used in preparation of concrete) procured from twelve different geological formations in the state of Karnataka, India, that may be employed in the preparation of self-compacting, low radio-activation concrete for neutron shielding.
- To create a database for neutron activation properties of the procured coarse aggregate (CA) samples, and to study the neutron transmission through SCC samples prepared using these CAs.
- To select an appropriate methodology to proportion SCC mix to meet the target strength, and shielding properties (neutron attenuation and residual activities).
- To select the most suitable SCC mix for shield design of nuclear installations in the State of Karnataka.
The above objectives have been achieved through:

- Neutron induced activation measurements for the CA samples collected from the twelve different geological formations in the state of Karnataka.
- Neutron dose transmission measurement through the SCC samples prepared using the above CAs.
- Comparison of the existing mix proportioning techniques available for achieving SCC, and selecting the method that is most appropriate.

1.7 Organization of the thesis

This thesis comprises of six Chapters.

Chapter 1 is on Introduction which contains a brief write-up on the geological formations in the state of Karnataka (from where the twelve CA samples have been procured), preliminary remarks on concrete as radiation shielding material, advantages of using self-compacting concrete, motivation for and significance of carrying out the present study, statement of objectives, and the methodology adopted for actualizing the objectives.

Chapter 2 is on Literature Review. This Chapter contains a comprehensive survey on the literature of relevance to the study, and is split into four sections as follows:

i. Concrete as Radiation Shielding Material
ii. Current Developments in Self-Compacting Concrete (SCC)
iii. Neutron Activation and Gamma Spectroscopic Analysis
iv. Studies on Neutron Transmission through Concrete

Chapter 3 is on Materials and Methods. This Chapter contains a description of the materials used and the methods explored to achieve SCC using different types of admixtures, the procedures adopted in carrying out the neutron activation and gamma spectroscopic analysis of the CA samples procured from the twelve different geological formations of the state of
Karnataka, and also the description of the methodology adopted for carrying out the neutron transmission study through the SCC slabs prepared using the CA samples.

Chapter 4 is on Results and Discussions. This Chapter contains a critical analyses of the results obtained in terms of trace element concentrations, and the activity build-up in the CA samples on which neutron activation has been carried out. The outcome of a comparative study of four different prevailing methods of proportioning SCC mixes, and also attempts at achieving SCC using different types of mineral admixtures is included in this Chapter. The various aspects of the parameters obtained by carrying out the neutron transmission study, namely (i) linear attenuation coefficients, and (ii) half-value layer thicknesses of SCC shields (slabs) prepared using the CA samples are also documented and critically examined in this Chapter.

Chapter 5 is on Summary and Conclusions. This Chapter contains, in brief, the outcome of the study carried out, namely:

i. The compatibility aspects of the twelve types of CA samples in preparing SCC of grades M25 and M30.

ii. Values of trace element concentrations and radioactivity build up in the twelve types of CA samples, determined after carrying out the neutron activation and gamma spectroscopic analysis.

iii. The most suitable/advantageous method that may be employed in proportioning SCC using the CA sample that showed the least activity build up, so as to produce low activation concrete that may be employed in fabricating neutron shields from the point of view of reduction of radioactive wastes generated following decommissioning of the nuclear installations.
iv. The neutron shielding parameters described by linear attenuation coefficient and half value layer thickness of M25 and M30 grade SCC prepared using the twelve different CA samples.

**Chapter 6** is on Scope for Future Work. This Chapter delves into the aspects that may be fruitfully explored for furthering the research work in the area of producing low activation concrete for neutron shielding - namely use of CA samples like high density aggregate and serpentine aggregate; and the possible use of mineral admixtures like silica fumes, rice husk ash, etc. in proportioning SCC for fabricating radiation shields.