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NUCLEAR RADIATION AND THE PROPERTIES OF CONCRETE

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1. EFFECTS OF NUCLEAR RADIATION ON THE PROPERTIES OF CONCRETE

1.1 INTRODUCTION

Concrete is often used for structures in which the concrete is exposed to nuclear radiation e.g. pressure and containment vessels for nuclear reactors, particle accelerators such as cyclotrons, and biological shielding. Exposure to nuclear radiation may affect the properties of concrete.

1.2 TYPES OF NUCLEAR RADIATION

There are several types of nuclear radiation:-

(a) Alpha radiation consists of rays of alpha particles which are the nuclei of helium atoms. It is one of the least penetrating forms of radiation and will pass through only a few centimeters of air.

(b) Beta radiation consists of rays of beta particles which are electrons and positrons. Beta-rays are also not very penetrating but they can travel a few metres in air before their energy is dissipated.

(c) Neutron radiation consists of rays of uncharged neutron particles. They can penetrate deeply into matter before their energy is absorbed.

Neutrons are usually classified according to their energy. Neutrons with energies less then 1 eV are usually called thermal or slow neutrons, those with energies between 1 eV and 0.1 MeV as epithermal neutrons, whilst those with energies greater than 0.1 MeV are referred to as fast neutrons.

The intensity of neutron radiation is described by the neutron flux which is the number of neutrons which cross a sphere of unit cross-sectional area during one second. The integrated flux is the time integral of flux and gives the total number of neutrons which penetrate a sphere of unit cross-sectional area and is usually designated in neutrons per square centimetre, i.e. n/cm^2, Frequently the term n.v.t. (neutrons, velocity, time) is used instead of n/cm^2 for the integrated flux.

(d) Gamma radiation consists essentially of rays of electro-magnetic waves. These rays are very penetrating and they can penetrate through considerable thicknesses of matter. The capture of neutrons by a nucleus of an atom is often accompanied by the emission of gamma radiation which is called secondary or capture gamma radiation. X-rays are similar to gamma rays but have a longer wavelength. As the wavelengths of electromagnetic waves decrease, the waves behave more and more like moving nuclear particles which are referred to as photons.

The energy of gamma radiation may be expressed as the "Energy dose" which is a measure of the total gamma radiation energy given up to, or absorbed by, unit mass. The unit "rad" (radiation absorbed dose) is a general measure of energy absorption. A rad (rd) is defined as an energy absorption of 100 ergs/g of the irradiated material. The "Dose rate" is the energy dose per unit of time. It is expressed as rd/h or rd/sec.

1.3 RADIATION DAMAGE IN CONCRETE

Neutron and gamma radiation can affect the properties of concrete. When assessing the results of investigations into the effects of nuclear radiation on the properties of concrete the following should be considered:

1.3.1 Heat

The absorption of nuclear radiation energy by a material causes an increase in temperature. As neutron radiation penetrates concrete the neutrons are absorbed producing heat and secondary gamma rays are emitted. The absorption of these secondary gamma rays, together with any incident gamma rays, also produce heat. Due to the low thermal conductivity of concrete, this heat can result in a significant temperature rise within the concrete mass and increases of about 250°C have been reported. Elevated temperature affects the properties of concrete and this must be considered when assessing the results of investigations into the effects of nuclear radiation on concrete properties.

1.3.2 Slow and Fast Neutrons

The results of experiments using both slow and fast neutrons have been reported. Published results do not prove conclusively whether fast neutrons produce more radiation damage in concrete than slow neutrons. There is also no published information which establishes whether the effects of high neutron dose rates over short periods may be extrapolated to lower rates over long periods. Higher dose rates will, however, cause greater increases in temperature.

1.3.3 Type of Aggregate

Experimental results (1)(2)(3) have indicated that changes in the properties of concrete exposed to nuclear radiation depend primarily on the behaviour of the concrete aggregate. This appears to be due to expansion of the aggregate which is caused by changes in the lattice structure of the various minerals in the aggregate when exposed to radiation.

Seeberger & Hilsdorf(1) have reported the results of experiments on a large number of concrete aggregates. During radiation the temperature of the specimens increased to about 150°C. Length changes and changes in the dynamic modulus of elasticity were determined.

When exposed to a thermal neutron flux up to 10^{20} n/cm^2 and fast neutrons up to 5 x 10^{18} n/cm^2 the increase in length of most aggregates was not more than about 0.1 per cent. In the case of granite, length changes of almost one per cent were observed and at a thermal neutron flux exceeding 10^{19}n/cm^2 and fast neutrons exceeding 5 x 10^{17}n/cm^2 the specimens disintegrated completely. With the exception of basalt, most aggregates showed a small decrease in the dynamic modulus of elasticity. The greatest decrease was for hornblende and for granite. It was concluded that for concrete to have a high resistance to neutron radiation, aggregates consisting of several different types of mineral components e.g. granite, should be avoided.

Kelly *et al.*(3) have reported on length increases in limestone and flint aggregates exposed to fast neutron doses varying from 1 to 4 x 10^{19} n.v.t. Limestone grew in length from about 0.2 to 2 per cent as the dose increased, whilst flint aggregate increased from about 0.1 to 1.3 per cent. Tests on concrete made with limestone and flint aggregates indicated that the linear changes were similar to those of the aggregates when exposed to neutron radiation. Tensile strength and dynamic modulus of elasticity of the aggregates decreased when exposed to fast neutrons.The strength and modulus of elasticity of the concrete made with the limestone and flint aggregates also decreased. Changes in the thermal conductivity of the aggregates and of the concrete were also ascertained and were found to be similar in magnitude. Tests to ascertain the effects of gamma radiation indicated that changes in the properties of aggregates and concrete exposed to nuclear radiation, were mainly caused by neutrons.

1.3.4 Hardened Cement Paste

Experimental results (1)(2)(3) indicate that nuclear radiation does not cause significant changes in the strength and modulus of elasticity of hardened cement paste. Unlike aggregates,hardened cement paste does not increase in volume when exposed to neutron radiation. The cement paste shrinks and the shrinkage is similar to that due to temperature and is related to the loss in weight which occurs rather than to the neutron dose$^{(3)}$. Cracking may therefore take place in irradiated concrete because of differential dimensional strains between the cement paste and the aggregate. This could be an important factor, in addition to the reduction in strength and modulus of elasticity of aggregates, in causing a decrease in the strength and modulus of elasticity of concrete when irradiated. Because hardened cement paste is relatively unaffected by nuclear radiation, it may be expected that rich concrete mixes will be more resistant to radiation than lean mixes. Investigations reported by Chisholm-Batten$^{(4)}$ tend to support this although the results are not conclusive because of temperature effects.

1.3.5 Type of Cement

In regard to type of cement, Van der Schaff$^{(5)}$ reported the results of tests on mortar specimens made with barytes aggregates and ordinary Portland cement and also with blast furnace slag cement. The specimens were exposed to fast neutron fluxes between 3 and 8 x 10^{19} n/cm^2. The temperature in the centre of the test specimens was between 150° and 200°C. The tensile strength of the Portland cement samples decreased by 31 per cent whilst the blast furnace slag cement samples did not decrease in strength. Tests$^{(4)}$ using ordinary Portland cement and high alumina cement in mortar specimens exposed to irradiation by thermal neutrons indicated that the loss in transverse strength with ordinary Portland cement was less than with high alumina cement. These results are however considered to be inconclusive because of the effects of temperature. Elleuch *et al*$^{(6)}$ reported the effects of irradiation on concrete made with aluminous cement and serpentine aggregates. The test specimens were subjected to fast neutron doses up to 10^{20} n/cm^2 and consequent temperatures of the order of 250°C. Although there were appreciable changes in the mechanical properties of the concrete, it was concluded that the changes in the properties of the alumina cement paste due to irradiation were insignificant and that the magnitude of the shrinkage of the hardened paste was similar to that caused by temperature.

1.3.6 Gas Generation

Gas is generated when concrete is exposed to gamma and neutron radiation(2)(3)(6). The gas generated consists mainly of hydrogen, oxygen, nitrogen, carbon monoxide and carbon dioxide and is caused by radiolysis of the water evaporating from the concrete at elevated temperatures. This gas generation is not considered to have much effect on the properties of concrete although the gas may have corrosive effects on parts of a reactor pressure vessel such as the steel liner and reinforcement. Internal pressure in the concrete may also be developed.

1.4 COMPRESSIVE STRENGTH

As indicated in Section 1.3 there are a number of factors which could affect the results of experiments on the effects of nuclear radiation on the properties of concrete, e.g. the effects of high temperature due to radiation, the type and energy spectrum of the radiation, the type of aggregate, and the mix proportions. Specimen size and shape, and moisture content, could also account for the considerable variation in the test results which have been reported.

1.4.1 Effects of Neutron Radiation

Hilsdorf *et al*(7) have referred to the results obtained by a number of investigators concerning the effects of both fast and slow neutrons on the compressive strength of concrete. In general no clear distinction can be drawn between the effects of fast neutrons and of slow neutrons. At an integrated neutron flux of 5×10^{18} n/cm^2 the compressive strength of uncooled irradiated specimens varied from a decrease of about 20 per cent to an increase of almost 20 per cent when compared with the compressive strength of companion specimens which were neither irradiated nor heated. For a flux of 5×10^{19} n/cm^2 compressive strength varied from a decrease of about 25 per cent to an increase of about 5 per cent. At higher fluxes there was always a reduction in strength.

At a flux of 5×10^{18} n/cm^2 the compressive strength of uncooled irradiated specimens was between 5 and 20 per cent less than that of specimens which had not been irradiated but which were exposed to temperatures similar to those experienced in the irradiated specimens. At a flux of 5 x 10^{19} n/cm^2 there were decreases of up to 30 per cent in compressive strength.

In several investigations the decrease in the compressive strength of the irradiated specimens when compared with that of specimens which were heated but not irradiated was of the same order of magnitude as the decrease when no allowance was made for temperature effects. It would seem that in many cases the decrease in compressive strength could mainly be due to the effects of neutron radiation and not the effects of elevated temperature caused by the absorption of neutrons.

1.4.2 Effects of Gamma Radiation

In some of the tests referred to in Section 1.4.1 the test specimens were simultaneously exposed to gamma radiation. Not many tests have however been reported on the effects on compressive strength of gamma radiation alone. Alexander (8) reported that for gamma radiation doses up to 10^{10} rd there was no reduction in the compressive strength of concrete when compared with the strength of companion specimens which had neither been irradiated nor heated. Sommers (9) however obtained reductions of between 25 and 60 per cent in compressive strength for gamma radiation doses exceeding about 10^{11} rd. In his tests the specimens were immersed in demineralised water to shield them against neutrons. Demineralised water may however cause deterioration in concrete and there was a significant decrease in the compressive strength of the concrete specimens which had not been irradiated but which had been immersed in demineralised water. The results therefore indicate that gamma radiation plus the effects of demineralised water had a greater effect on compressive strength than the effects of immersion in demineralised water only. It should also be noted that tests on concrete exposed simultaneously to neutron and gamma radiation have not shown such large decreases in compressive strength as Sommers' tests did for gamma radiation alone.

1.5 TENSILE STRENGTH

Factors which could influence the results of tests on the effects of nuclear radiation on compressive strength also apply to tensile strength and there is considerable variation in the results which have been reported.

1.5.1 Effects of Neutron Radiation

Gray(2) compared the tensile strength of irradiated concrete specimens with that of "untreated" companion specimens, i.e. specimens which were neither irradiated nor subjected to the elevated temperatures experienced in the irradiated specimens. For concrete with limestone aggregate and at integrated fast neutron fluxes of 2×10^{19}, 3×10^{19} and 4×10^{19} n/cm^2 the tensile strength of the irradiated specimens was 31, 52 and 57 per cent less respectively than that of the untreated specimens. For concrete with flint aggregate, the reductions were 56, 67 and 67 per cent respectively. Elleuch(10) reported a reduction of about 65 per cent for a fast neutron flux of 2×10^{19} n/cm^2 and 80 per cent for fast neutron fluxes between 4×10^{19} and 10^{20} n/cm^2. Reductions in the tensile strength of mortar specimens made with barytes and magnetite aggregates respectively have been reported by Van der Schaaf(5). When exposed to an integrated flux of between 3 and 8×10^{19} n/cm^2 the reductions were 31 and 22 per cent for the barytes and magnetite mortars respectively. Tests carried out at the Atomic Energy Research Establishment at Harwell(11) in the United Kingdom between 1953 and 1956 gave an increase of 17 per cent, and decreases of 6, 4 and 26 per cent in transverse rupture, at integrated slow neutron fluxes of 0.5, 1.6, 3 and 7×10^{19} n/cm^2 respectively.

In the above investigations the tensile strength of irradiated specimens has been compared with that of "untreated" specimens, i.e. neither irradiated nor exposed to temperature. Gray(2) also compared the tensile strength of irradiated specimens with that of companion specimens which were not irradiated, but which were exposed to temperatures similar to those reached in the irradiated specimens. The reductions in strength were of a similar order to those obtained in the comparison with untreated specimens. Elleuch(10) reported decreases in tensile strength, compared with temperature exposed specimens, of about 40 per cent at fluxes between 2 x 10^{19} and 10^{20} n/cm^2. This is significantly less than the decreases of 65 and 80 per cent mentioned above when compared with untreated specimens and indicates that part of the reduction in strength of untreated specimens was due to the effects of temperature. Van der Schaaf(5) reported reductions of 29 and 16 per cent when compared with the strength of barytes and magnetite mortars heated to an average of 200°C. These reductions are also less than in the comparisons with untreated specimens.

In the Harwell tests referred to above (11) the reduction in the tensile strength of the irradiated specimens was 4, 23, 22 and 39 per cent respectively when compared with the tensile strength of companion specimens which were not irradiated but were heated to 50°C. Similar results were obtained when the companion specimens were heated to 100°C. These decreases are larger than those obtained in the comparison with untreated specimens. It is of interest to note that the strengths of the heated specimens were about 20 per cent higher than those of the unheated specimens and this could account for the greater percentage reduction.

From the above results it is difficult to conclude precisely what part temperature plays but it is apparent that at neutron fluxes greater than 10^{19} n/cm^2 there is a significant reduction in tensile strength. The reduction is also greater than in the case of compressive strength

1.5.2 Effects of Gamma Radiation

Results reported by Gray(2) on the effects of gamma radiation alone on the tensile strength of irradiated specimens as compared with that of "untreated" specimens indicate that at a gamma radiation dose between 2 and 4 x 10^{10} rd there was no significant decrease in tensile strength.

1.6 MODULUS OF ELASTICITY

Gray (2) found that, for integrated fast neutron fluxes between 7 x 10^{18} and 3 x 10^{19} n/cm^2, the modulus of elasticity (E) of irradiated concrete was between 10 and 20 per cent less than that of untreated concrete. At a slow neutron flux of about 2 x 10^{19} n/cm^2 Alexander (8) reported similar reductions in E. At an integrated fast neutron flux of about 5 x 10^{19} n/cm^2 Van der Schaaf(5) reported a 42 per cent reduction in dynamic E and 45 per cent in static bending E for mortar specimens consisting of Portland cement and barytes aggregate. With blast-furnace slag cement the reductions were 34 and 18 per cent respectively. For mortar specimens with Portland cement and magnetite aggregate the reductions were 16 and 20 per cent respectively. These decreases were generally, if not always larger than for test specimens which were not irradiated but which were subjected to an elevated temperature of about 200°C. Van der Schaaf concluded that the reductions in E could in some cases have been entirely due to the temperature elevation brought about by irradiation. With fast neutron fluxes greater than 3 x 10^{19} n/cm^2, Elleuch *et al*(6) have reported reductions in E of more than 30 per cent, as indicated by ultrasonic pulse tests, for concrete made with aluminous cement and serpentine aggregate. These reductions were considerably greater than those for companion specimens heated to 200°C but which were not irradiated.

No information has been obtained regarding the effects of gamma radiation on the modulus of elasticity.

1.7 DIMENSIONAL CHANGE

As indicated in Section 1.3.3 concrete aggregates expand when exposed to nuclear radiation. The expansion varies depending on the type of aggregate. Concrete also expands and increases in volume when exposed to a sufficiently large neutron flux. The expansion of concrete also depends primarily on the type of aggregate used in the concrete. Gray(2) and Kelly *et al.*(5) have reported the results of dimensional changes in concrete specimens when exposed to fast neutrons. For concrete incorporating limestone aggregate there was very little change in volume for integrated fast neutron fluxes up to 2 x 10^{19} n/cm^2.

At a flux of about 4×10^{19} n/cm^2 the increase in concrete volume was approximately 1 per cent. Concrete with flint aggregates increased more rapidly in volume. At a neutron flux of 2×10^{19} n/cm^2 the increase was about one per cent and at 4×10^{19} n/cm^2 about four per cent. As previously mentioned (2) the decrease in tensile strength of limestone concrete was less than for flint aggregate concrete.

Reported results indicate that at neutron fluxes less than 10^{19} n/cm^2 dimensional changes in concrete are insignificant. Up to this order of neutron flux the dimensional change is approximately equal to the shrinkage of concrete which may be expected due to thermal effects.

1.8 THERMAL EXPANSION

As mentioned by Hilsdorf *et al*(7) the available information indicates that for neutron fluxes up to 5×10^{19} n/cm^2 there does not appear to be any significant difference between the coefficient of thermal expansion of irradiated concrete and that of companion specimens exposed to temperature but not to irradiation. Results for neutron fluxes greater than 5×10^{19} n/cm^2 and for the effects of gamma radiation have not been published.

1.9 THERMAL CONDUCTIVITY

The heat flow properties of concrete are important in regard to the determination of temperature gradients and therefore of thermal stresses. Gray(2) has reported the effects of neutron radiation on the thermal conductivity of concrete made with limestone and flint aggregates as well as lightweight aggregates. It appears that at neutron fluxes greater than 0.5×10^{19} n/cm^2 the thermal conductivity may be reduced by between 20 and 50 per cent when compared with the thermal conductivity of untreated concrete. Kelly *et al.*(3) reported increases in the thermal resistivity (which is the reciprocal of thermal conductivity) for concrete made with limestone and flint aggregates and exposed to fast neutron doses between 1 and 3×10^{19} n/cm^2.

The increases in thermal resistivity, and therefore the decrease in thermal conductivity, of the concrete were similar in magnitude to those of the aggregate.

1.10 CONCLUSION

If the neutron and gamma radiation doses to which concrete is exposed exceed certain values, then the properties of concrete may be deleteriously affected. The type of aggregate used in the concrete plays a dominant rôle in the effects of nuclear radiation on the properties of concrete.

The British Specification for Prestressed Concrete Pressure Vessels for Nuclear Reactors(12) states that from the experimental data available it is not possible to quantify precisely the effects of neutron irradiation on the properties of concrete but that for neutron doses up to 0.5×10^{18} n/cm^2 the effects of irradiation are considered to be insignificant.

The American Code for Concrete Reactor Vessels and Containments(13) states that for concrete reactor vessels the radiation limit for concrete shall not exceed 10×10^{20} n.v.t. As previously indicated the term n.v.t. is used instead of n/cm^2 for integrated neutron flux.

2. RADIATION SHIELDING

2.1 BIOLOGICAL HAZARDS OF NUCLEAR RADIATION

The biological hazards of nuclear radiation are due to the fact that the radiations interact with human tissues, losing some of their energy in the process. This energy loss is sufficient to ionise atoms in living cells and thereby disturb their delicate chemical balance. Depending on the radiation dose received the proportion of cells damaged by radiation may be sufficiently large to cause the organism to die. Radiation must therefore be reduced or attenuated sufficiently so that what is left cannot cause permanent harm to persons exposed to it.

2.2 ATTENUATION OF NUCLEAR RADIATION

2.2.1 Neutron Radiation

Attenuation of neutrons is accomplished chiefly by causing them to lose energy or to be slowed down in collision with the nuclei of atoms. The slowed-down neutrons are captured by nuclei and this usually brings about the emission of gamma rays. The loss of energy is due to elastic and inelastic scattering and the greatest loss occurs when the target nucleus has a mass which is nearly the same as that of the neutron. The nuclei of hydrogen atoms have approximately the same mass as neutrons and are therefore the most efficient for the slowing down of neutrons. The slowing down process is aimed at reducing fast neutrons to thermal neutrons of low energy when diffusion and capture will accomplish the desired attenuation. The relatively high efficiency of hydrogen and oxygen in the slowing down process makes the presence of these elements very desirable.

2.2.2 Gamma Radiation

The attenuation of gamma rays can be brought about by a number of processes. The three main processes are photoelectric absorption electron pair production, and Compton absorption and scattering. The contribution of each of the above processes to the total attenuation depends on the photon energy and the kind of atoms in the absorbing material. The first two processes are critically dependent on the atomic number of the absorber, and are particularly important for the heavy elements, e.g. lead. For concrete the photoelectric and pair production effects are small and for gamma ray energies in the 1 to 10 MeV range, attenuation is almost entirely due to Compton absorption and scattering(14). At energies greater than 10 MeV, the pair production process becomes increasingly important in concrete.

Compton absorption and scattering results from collisions between photons and electrons. As a result of the collision the photon gives a portion of its energy to the electron and has its direction changed. The loss of energy is called Compton absorption and the change in direction is called Compton scattering. Both lower the penetrating power of the photon. The number of photons that will collide with electrons and the number of successive collisions which a photon will experience depends approximately on the total number of electrons in the path of the gamma rays. The number of electrons in an atom is roughly proportional to the mass of the atom. The mass and density of the shielding material are thus very important in regard to the attenuation of gamma radiation.

2.3 CONCRETE AS A SHIELDING MATERIAL

Concrete is considered to be an excellent material for radiation shielding. It has good structural properties, it is workable and adaptable and normally requires little maintenance. Because it contains hydrogen in both free and chemically bound water, as well as other light nuclei, it is a good neutron shield. Its relatively high density, particularly if heavy aggregates are used, also makes it suitable for the attenuation of gamma rays. A disadvantage is the low thermal conductivity of concrete which makes it difficult to remove the heat generated in the shield.

The thickness of a concrete radiation shield depends on the intensity, energy and nature of the radiation source, the permissible level of radiation outside the shield, and the radiation shielding properties of the concrete in the shield.

In designing the shield, stresses due to temperature must be carefully considered. In the shielding of nuclear reactors the inner face of the concrete shield is exposed to heat from the core of the reactor. Heat is also generated within the shield as it absorbs neutron and gamma radiation. Temperature distribution across the shield is usually non-linear and thermal stresses due to temperature gradients are an important design problem. Stresses due to shrinkage and creep must also be considered. Increase in temperature due to nuclear radiation causes a loss of moisture in the concrete and this may reduce the effectiveness of the shield in regard to neutron attenuation.

Where it is necessary to reduce the thickness of a radiation shield because of space considerations, high density concrete is often used. A wide variety of materials has been used as aggregate in heavy concrete, e.g. barytes, magnetite, limonite, ferrophosphorous and ilmenite. The density of concrete made with these aggregates varies between 3.54 and 3.94 gm/cm^3. Iron and steel shot have also been used as aggregates and result in concrete with a density between 5.47 and 5.96 gm/cm^3. Caution is, however, required in the use of ferrophosphorous aggregates(15).

As mentioned, high density is of primary importance for attenuation of gamma radiation, but for neutron shielding, light atoms, such as hydrogen, are required in the concrete. Gamma radiation arising from the capture of neutrons must also be considered. In the design of concrete mixes for radiation shielding a compromise must therefore be made to meet these conflicting requirements and the optimum composition of the concrete requires careful consideration.

Heavy-aggregate concretes also give rise to problems in construction. Special falsework and shuttering is required and problems concerning segregation arise in the course of placing the concrete. It is important that voids in radiation shields be prevented and to this end use has been made in construction of the grout intrusion or pre-placed aggregate method.

Polivka(16) has published an annotated bibliography on concrete for radiation shielding which lists 365 selected references pertaining to the types and properties of concretes used in radiation shields. Reference should also be made to the "Engineering Compendium on Radiation Shielding"(17) as well as to the publications of Price *et al.*(11), and of Jaeger(18).

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