Material selection and mix design of radiation shielding concrete

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Abstract. Based on the compiled literature and the availability of materials that could be used, concrete was selected as the best shielding material. Further work was carried out to develop a specific mixture that would shield the radioactive energies. The important special concrete ingredients that were considered in the mix design were high density aggregates and boron-containing aggregates. Various high density concrete mixtures of w/c (water/cementitious ratio) = 0.42, 0.45, 0.5 and 0.6, were prepared and adjusted appropriately in order to obtain the desired mix characteristics.

The final (high density shielding concrete) mix produced was workable and cohesive with average 28-day compressive cube strength of 30 MPa, w/c = 0.51 and density of 4231 kg/m^3. The concrete had high slump with a height and spread of 230 mm and 510 mm respectively. It was composed of CEM 52.5 N, silica fume, water, hematite sand, hematite stones, steel shots, colemanite and chemical admixtures.

Keywords. Radiation shielding, curing, retardation, workability

Introduction

The nuclear industry has traditionally been regarded as the originator of high density concrete where it is used for radiation shielding. Early work of the 1950s in USA, resulted in the development of some of these shielding materials. Further significant gains were made during the 1980s, mainly at the Sella field reprocessing site where a range of concrete relative densities between 3.4 and 8.75 together with a range of grouts between 2.8 and 6.6 were successfully designed and used in significant quantities [1]. In general the shielding effectiveness is proportional to the concrete density and is used where space is premium; however, it can additionally be affected by the nature of radiation though this is a very specialist topic beyond the realm of the concrete technologist.

The typically used aggregate in shielding (high density) concrete may be naturally occurring or they may be materials used in other industries for various purposes. The common aggregate types used include: Barytes (naturally occurring barium sulphate mineral ore), magnetite and haematite (Iron ores), iron and steel shots, various sizes and types of scrap iron and steel, ferroslag, iron silicon slag, lead shots.

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The advantages of using concrete as a shielding material are related to its good compromise thickness requirements for neutron and photon shielding; it can also be cast into almost any complex shape [2]. Its use as a radiation shielding material is well-established as indicated by the availability of existing knowledge and literature [1-3]. Other benefits of using concrete include the local availability of the required high density aggregates, its versatility, composite nature, economic savings, low maintenance, ease of manufacture and structural integrity. Concretes are highly durable and provide permanent shielding installations [3], unlike materials such as lead which may lack structural integrity, or use of water that might cause complications such as rusting and leakage of containers. Materials have different shielding properties for different types of radiation and therefore the selection of shielding material is a function of the radiation type and the energies of the specific radiation.

This paper is limited to discussion and identification of suitable raw materials for high density shielding concrete (HSDC), its mix design and optimization, testing of mechanical properties. The evaluation of the shielding properties of HSDC is presented in another paper [4].

1. Use of concrete for radiation shielding

1.1 Galena aggregates

In a study conducted by Mortazavi,et al. [5], where the focus was on production of an economic high-density concrete for shielding of megavoltage radiotherapy room and nuclear reactors, galena was used as the only heavy-weight aggregate in the mix. In their investigation, two types of concrete mixes were produced. These were the control and galena mixes of w/c (water/cementitious ratio) of 0.53 and 0.25 respectively. The galena used in this study had a density of 7400 kg/m^3 and was obtained from a mine in Firouzabad in Iran. It was reported that while the control mix yielded a density of 2350 kg/m^3 and 30 MPa, the galena high density concrete had a density of 4800 kg/m^{3}and 50 MPa. It was reported that the galena concrete gave good shielding properties.

It is well known from nuclear physics that only lighter element such as hydrogen and boron are capable of shielding neutrons. Neutrons penetrate through lead quite easily, and since lead was the only special aggregate used in the mix [5], the concrete would not be able to stop the neutrons. The reason for the good shielding properties obtained in the Mortazavi et al. study [5] is that a gamma ray source in form of a narrow beam emitted from a cobalt-60 therapy unit was used. The results were therefore exceptional since lead is good in shielding gamma rays but inadequate for neutron shilelding.

1.2 Effect of colemanite

Gencel, et al. [6] conducted a study on the effect of colemanite on physical and mechanical properties of concrete, when used as a replacement aggregate. Concretes containing different ratios of 10, 20, 30, 40 and 50% colemanite were incorporated into the mix as fine and as coarse aggregate. Concrete mixes of cement content 400 kg/m^3 and w/c = 0.42 were used in the experiments. It was found that the slump of concrete significantly decreased with addition of colemanite into the mix. Further still, the concrete tended to flocculate upon addition of colemanite into the mixes. The slump reduction and related flocculation, were attributed to a possible chemical reaction between colemanite and the cement paste. The overall conclusion of this study was that using 10 to 50 % colemanite as aggregates in concrete negatively affects the concrete in respect to both its physical and mechanical properties. Colemanite is a water soluble boron-containing material, and is known to delay the setting of concrete even when added in small quantities [4].

1.3 Iron ore and steel shots

Iron ore and steel shots are normally included in the HSDC mixes in order to obtain the desired high density which helps in attenuation of photons and slowing down of fast neutron. Several studies and developments have been carried out using these aggregates where magnetite and hematite have been used as the main sources of natural iron.

In a study carried out by Dubrovskii, et al. (1970) [7], hematite was used as the natural source of iron and was incorporated in concrete as fine and coarse aggregate. The mix design had a density of 3030 kg/m^3 and showed good shielding properties. In another study by Kharita, et al. (2007) [8], hematite was used together with black coastal sand to produce special shielding concrete that was tested using two different gamma sources and a neutron source. At the conclusion of the study, hematite samples were considered the best for shielding gamma rays as compared to those mixes which contained no hematite. It was also found that the samples showed good results for shielding neutrons and this was suspected to have resulted from the high iron content of hematite and the presence of iron hydroxide (Kharita, et al., 2007) [8]. Mahdy, Speare and Abdel-Reheem (2002) [9] conducted a study to investigate the effect of transient high temperature on magnetite based heavy-weight and high strength concrete. In their study, twelve mixes with slumps of over 100 mm and strengths of 140 Mpa at 180 days were used. The mixes contained combinations of cement, silica fume, coarse magnetite with a maximum size of 16 mm, fine magnetite and natural fine sand. It was found that concrete strength decreased when temperatures were raised to 100°C. With further increase in temperature, the loss in strength recovered and reached peak strength of 10 % to 30% above the corresponding strength at room temperature. At temperatures of 500 and 700 °C, the strength dropped sharply (Mahdy, Speare & Abdel-Reheem, 2002) [8].

Warnke, et al. (2001) [10] used steel granules to develop a concrete shielding material for casting of low cost, storage concrete containers for waste management. The iron granules were used in the concrete mix at a proportion of 50%. The ion aggregates raised the mix densities from the normal 2400 kg/m^3 to 4000 kg/m^3; and gave concrete compressive strengths reaching up to 65 MPa.

2. Special concrete ingredients for high density shielding concrete

Most of material considerations for HSDC have physical and chemical property requirements which can be challenging to traditional mix design methods. Therefore careful evaluation of these issues is necessary both before and during use of the concretes and grouts. Designers and specifiers of HDSC need to be aware that aggregate grading, will frequently fail to comply with more traditional specifications but high quality concrete can still be produced using these materials. It is generally appropriate to design HDSC mixes starting from basics of the mix characteristics in terms of aggregate/cement ratios and fines content, which will often appear to be extreme and unconventional. Water contents need to be minimized to prevent segregation and full use of superplasticisers is normally recommended (in order to achieve workable mixes), though magnetite has been used to produce self-compacting concrete.

The HSDC concrete developed in this research was required to be of sufficient high density to be of a special type needed to fulfill the purpose of neutron and gamma-rays shielding. Normal weight concrete would be too thick if it was considered for this purpose, which would result in an excessive shield size well beyond the space limitations available; it would also be uneconomical. Each identified ingredient used in the mix development had a certain role to play. The following materials were identified for use in this investigation:

- Ordinary Portland cement (OPC), CEM 52.5 N.
- Hematite (natural high density aggregate).
- Iron/steel shots (artificial high density aggregate).
- Municipal water.
- Colemanite (boron containing aggregate).
- Galena (natural high density Lead containing aggregate).

The aggregates were divided into two categories consisting of:- high density aggregates which produce HDSC, attenuate (absorbs) photons (gamma-rays) and scatters neutrons (change the energy from fast to thermal), and the boron containing aggregate that attenuates thermal neutrons.

3. Chemical analyses of aggregates

Small samples were obtained from the identified suppliers and tested for chemical compositions. The purpose of this testing was to ensure that ingredients that could become radioactive due to elements that have long decaying half-lives (i.e. Cobalt, Copper, Nickel, Zinc etc.) were not significantly present in the concrete mix. These tests were also used to confirm the guarantees presented on the suppliers' product data sheets. The chemical composition analyses of aggregates were conducted using ICP (Inductively Coupled Plasma) and XRF (X-ray Fluorescence methods). It was confirmed that none of the selected aggregates for mix design of HDSC had long half-life elements.

4. Practical mix design of high density concretes

Various trial concrete mixtures of w/c's of 0.42, 0.45, 0.5 and 0.6 were prepared. In the process of developing a suitable HDSC, adjustments were made to the various mixtures so as to obtain the desired material properties, especially:- workability, cohesion, density and compressive strength. The various mixtures and their results are shown in Table 1. As indicated in table, the TM1 was too stony and lacked cohesion. The mix was therefore modified and colemanite was also introduced. It was decided to start with two different proportions of 5 % colemanite (TM1) and 10 % colemanite (TM3), thus satisfying the requirements of the Monte Carlo Neutron Particle (MCNP) simulation outputs [4]. In addition to the above results, a delay in setting time of TM2 was also noticed. As a result of this retardation effect of colemanite, cubes disintegrated when placed under water after they had been demoulded (see Figure 1). The results of TM3 were similar to those obtained for TM2. The mix (TM3) yielded no slump, was not workable nor cohesive, and had delay in setting time. However, the early strength of TM3 was much higher than that obtained for TM2. This resulted due to the different demoulding times of the cubes. The TM2 cubes were demoulded after 24 hours while the TM3 cubes were demoulded after 48 hours. It was evident that the use of colemanite in the mix resulted in the two effects of very low workability with zero slump and delay in setting time of the concrete, which confirms findings in the literature [6].

Figure 1. Disintegrated cubes after being placed under water following the use of colemnite

Given the two difficulties posed by use of colemanite in mixtures, it was decided to introduce two admixtures to improve results. In order to achieve a high slump, a superplasticiser was introduced. To improve the setting time of concrete, an accelerator was added into the mix. This accelerator needed to be free from chlorides due to the high percentage of steel shots used in the mix. A 1 % dosage of accelerator was used in TM 4.

5. Further mix adjustments

The 10 mm slump obtained from TM4 indicated that the dosage of the superplasticiser was not effective enough. In addition to the 10 mm slump obtained, the mix lacked finer aggregates and as a result, its cohesion was poor. The delay in setting time of the TM4 mix, still posed a problem. TM4 mix was therefore modified by adjusting the stone/sand blends from 55/45 (percent ratio of superfine to fine aggregates) in TM 4 to 60/40 blend into TM5. The dosages of chemical admixtures were also adjusted.

As a result, the slump improved to 25 mm for TM5, and the mix was generally cohesive and workable. The required high slump was however still not achieved. From TM5, it was determined that increasing the finer particles of FA (fine aggregates) and reducing the high content of the CA (coarse aggregate) had an influence on the consistency, workability and cohesion of the mix. TM5 mix was further modified to

TM 6 by replacing 60/40 FA blend with 70/30 blend and reducing the w/c and stone content as shown in Table 1. The dosages of the superplasticiser and accelerator used in TM 5 were also increased. Due to the extended setting of the concrete, cubes were cast and only demoulded after 48 hours, cured at 23°C and 65% RH for the first seven days and then under water for the rest of the 28 days. At demoulding of the cubes at 48 hours, it was noticed that the concrete had not completely set. This was attributed to the high dosages of the chemical admixtures. TM 6 was modified into TM7 by changing the type of superplasticizer and using it at a lower dosage, incorporating silica fume and high alumina cement (HAC); no accelerator was used. The TM7 mix gave satisfactory results, setting fully within 24 hours. It also had good cohesion and strength gain at both, the early and late ages. Further adjustments were done on TM7 by removing HAC from the mix and re-introducing the accelerator. This gave TM 8, which required three days for proper setting of the mix samples before demoulding. Use of HAC in the mixtures was not favoured due to its potential to cause *conversion*, a chemical alteration process leading to long-term strength loss in concretes made with HAC. TM8 was considered to be the suitable mix design. The results of its evaluation for radiation shielding are presented in [4].

Table 1. HDSC concrete mix design

			TM1	**TM2**	**TM3**	**TM4**	**TM5**	**TM6**	**TM7**	**TM8**
Ingredients mass %	CEM I 52.5N- PPC		8.75	8.98	8.48	8.07	8.07	10.39	7.96	7.88
	Water		4.38	4.03	4.24	4.86	4.86	4.41	4.41	4.36
	Hematite Stones		46.02	44.89	43.58	37.22	33.52	28.96	28.96	28.66
	Hematite Sand		21.77	21.39	20.97	19.65	23.38	19.73	19.73	19.53
	Steel shots		19.08	18.62	18.37	27.92	27.93	35.77	35.77	35.40
	Colemanite		-	2.30	4.36	2.07	2.07	2.31	2.31	2.28
	Superplasticiser 1		-	-	-	0.06	0.05	0.27	0.17	0.12
	Superplasticiser 2		-	-	-	-	-	-	0.14	0.05
	Accelerator		-	-	-	0.16	0.12	0.36	-	0.19
	Silica fume		-	-	-	-	-	-	0.69	0.69
	High alumina cement		-	-	-	-	-	-	1.73	-
	W/C		0.5	0.45	0.5	0.5	0.6	0.42	0.42	0.51
Results	Density (kg/m^3)		4514	4421	4071	4287	4292	4372	4220	4231
	Slump	Height (mm)	50	No slump	No slump	10	25	190	210	230
		Spread (mm)	-	-	-	-	-	-	530	510
	Cohesion		Poor	Poor	Poor	Poor	Good	Good	Good	Good
	7day strength (Mpa)		39.35	2.64	12.6	-	-	2.6	20	2.51
	28day strength (Mpa)		54	41.1	33.8	-	-	38.9	48	29.94

6. Conclusions

The final mix design of the high density shielding concrete was workable and cohesive with average 28- day compressive cube strength of 30 MPa, water to cement ratio of 0.51 and density of 4231 kg/m^3. The concrete had a high slump with a height and spread of 230 mm and 510 mm respectively. The main special aggregates used in the mix were hematite, steel shots and colemanite. It was observed that colemanite had a strong effect of retarding the setting of concrete. The retardation could be offset by use of high alumina cement, however, consideration should be given to potential *conversion* of concrete as a result of using high alumina cement. It may be appropriate to avoid using high alumina cement in shielding concrete and instead compensate for set retardation by allowing a long period of setting before demoulding or removal of formwork.

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