BETOANE CU CAPACITATE DE ECRANARE A RADIAȚIILOR GAMA PENTRU CONSTRUCȚII SPECIALE
CONCRETES WITH GAMMA RADIATION SHIELDING CAPACITY FOR SPECIAL CONSTRUCTION APPLICATIONS

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The article presents the influence of the heavy aggregates and the water/binder ratios on the engineering and gamma shielding properties of the concrete. The elements with great atomic number, Z as Pb (Z = 82) and Ba (Z = 56) contented in the used heavy aggregate, namely lead slag, cathode ray tube (CRT) glass waste and barite, showed a decisive contribution to the increase of the concrete gamma radiation shielding. Also, the high density of the concrete was significantly influenced by the steel shot aggregate as partially substitute of the siliceous sand.

The concrete with CRT glass waste recycled sand had 360 days compressive strength (88.3 MPa) higher than the river sand concrete (83.5 MPa), at the same aggregate volumes and the same water/cement, w/c, ratio (0.38).

The results showed that concrete with complex aggregate based on barite, steel shot and CRT waste glass have had the highest gamma linear attenuation coefficient of 0.192 cm^{-1} for Co-60, higher by 18.2 % than concrete with barite as total aggregate.

Articolul prezintă influența agregatelor speciale, de densitate ridicată și a raportului apă/liant asupra proprietăților caracteristice și de ecranare a radiațiilor gama ale betonului. Elementele cu număr atomic, Z, mare cum sunt Pb (Z = 82) și Ba (Z = 56) conținute în agregatele grele utilizate – zgură plumbică, deşeuri de sticlă de la tuburile catodice (CRT) și barită de haldă – au prezentat o contribuție decisivă la creșterea coeficientului liniar de atenuare a radiației gama. De asemenea, substituirea nisipului silicios cu alice de oțel a condus la creșterea densității betonului și, implicit, a capacității de ecranare.

Betonul cu nisip din deşeu de sticlă CRT a avut rezistența la compresiune la 360 de zile (88,3 MPa) mai mare decât cea a betonului cu nisip de râu (83,5 MPa), pentru același volum de agregat și același raport apă/ciment (0,38).

Rezultatele au arătat că betonul cu agregat complex pe bază de barită, alice de oțel și deșeu de sticlă CRT are cea mai bună capacitate de ecranare a radiației gama, concretizată de o creștere a coeficientului de atenuare liniară, de 0,192 cm^{-1} pentru Co-60, cu 18,2 % mai mare decât betonul cu agregat total de barită.

Keywords: *Heavyweight concrete, density, linear attenuation coefficient, compressive strength*

1. Introduction

The concrete based on heavyweight aggregates has been defined as advanced composite materials with a high density, dense microstructure, high energy absorption, low shrinkage and long-term durability [1]. Heavyweight concrete (HWC) with a density higher than 2600 kg/m^3 is widely used in nuclear facilities radio therapy rooms, and for storing and transporting short lived low and intermediate level radioactive wastes. Heavyweight concrete is cheap, easy to produce in different compositions and versatile shielding material if it meets the properties required. The higher the concrete density (greater than 2600 kg/m^3), the smaller thickness of concrete is required to provide radionuclide diffusion barrier and ionizing radiation shielding [2-4]. Concrete contains 75-80 vol.% aggregates and 15-20 vol.% hardened cement. The fact that at least 75% of the concrete is aggregate shows its importance on the ability of the barrier to diffusion of gamma radiation [5-7].

For achieving higher density traditional aggregates of concrete (sand, gravel, crushed rocks) are partially replaced with materials characterized by high specific gravity such as ferrous materials (magnetite, hematite, limonite, siderite, steel shot) and barite [8-19].

The shielding capacity is significantly influenced by the density of the material. Thus, a technical criterion for the selection of aggregates is the content of atoms with high atomic numbers, which determines the high density of the material.

Barite rock based on high content of $BaSO_4$ is the most widely used material in the heavyweight concrete production, which explains the high number a research works. The use of barite in concrete with radiation shielding properties allows obtaining of high density and suitable mechanical

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strengths [20, 21]. The shrinkage of concrete with barite content is 70-75% of conventional concrete, thermal expansion coefficient measured at 4–38°C is nearly twice that of the conventional concrete, the modulus of elasticity and Poisson`s ratio is nearly equal or even higher to those of the normal concrete although some researchers have reported higher values of modulus of elasticity for barite concrete compared to normal concrete, specific thermal coefficient and heat transfer in heavyweight concrete are significantly less than the corresponding quantities in normal concrete [22, 23]. In some cases, fine barite aggregates delay the setting and hardening of concrete [22].

Throughout time many researchers have an increasing interest in finding new suitable materials, optimizing the mixture proportion and assessing the effect on the microstructure, mechanical strengths, and durability and shielding properties. The scope for using alternative materials in concrete production is twofold; it can provide some technical improvements to the final product, but it is also expected to be beneficial from an environmental point of view.

The article presents the influence of the aggregates type - barite, CRT waste glass, lead slag, hematite and steel shot, the filler type - limestone, barite and the different water to binder ratios on the some engineering characteristics and the linear attenuation coefficient (μ, cm^{-1}) of gamma shielding of the concrete.

2. Experimentals

2.1. Materials

The heavyweight concrete (HWC) was designed with a binder system composed of powders including cement and a filler type (limestone, barite), five aggregate types (barite, CRT glass, lead slag, hematite and steel shot), superplasticizer and water.

Portland cement type CEM II/A-V 42.5N, according to [24] was used.

Limestone filler used in concrete has the following characteristics: - CaO-53.5 wt.%, residue on 0.125 mm sieve- 9.8%, density- 2760 kg/m^3. The limestone filler was required in the concrete composition to increase segregation stability and shielding capacity.

Crushed barite ore includes $BaSO_4$ is a heavyweight aggregate used in the heavy concrete for special applications. Barite ore used was a residue from barite rock processing for barite mineral obtain. The $BaSO_4$ content varied from 18.12 to 46.21% and the density from 2.426 to 3.620 g/cm^3. The lower density values are the effect of including silicate rocks (gneisses) in varying proportions in barite mineral.

The leaching properties of the barite ore and the lead slag were tested as according with Column test procedure included in CEN/TS 14405 [25]. The results obtained, as released chemical species, at various liquid/solid (L/S) ratio, are showed in Table 1 and Table 2.

Table 1

The concentrations of released chemical species from barite ore / *Concentrațiile speciilor chimice eliberate de baritină*

Chemical composition/ *Compoziție chimică*	Concentration/*Concentrație* (mg/l)		
	L/S=0.1	L/S=1	L/S=2
Zn	1.08	0.02	0
Mo	0	0.2	0
Mn	14.6	0.22	0.191
Cr	0	0	0.002
Ca	0.04	64.8	30.8
SiO_2	31	0.15	24
NH_3	10.9	0.267	0.16
Cu	0.56	0.047	0.03
Mg	53.46	0.036	4.86
PO_4^{3-}	0.8	0.6	0.8
NO_2^-	0	0	0
NO_3^-	1.77	11.5	9.3
SO_4^{2-}	3300	89	20
Cl^-	2.03	0.14	0.05

The leachate from crushed barite ore do not contains hazardous contaminants, as heavy metals with concentration higher than the regulated limit (Zn = 1.2 mg/l, Mo = 0.2 mg/l, Cu = 0.6 mg/l, for L/S = 0.1) [26].

Table 2

The concentrations of released chemical species from lead slag / *Concentrațiile speciilor chimice emise eliberate de zgura de plumb*

Chemical composition/ *Compoziție chimică*	Concentration/*Concentrație* (mg/l)		
	L/S=0.1	L/S=1	L/S=2
Zn	0.07	1.58	0.52
Mo	6.9	6.8	0
Mn	26.4	22.3	25.4
Cr	0	0	0.007
Ca	45.6	36.4	96.8
SiO_2	33	24	23
NH_3	24.07	3.04	7.66
Cu	1.80	2.41	0.12
Mg	29.889	31.59	0.056
PO_4^{3-}	1.1	0.5	0.3
NO_2^-	0	11	11

The lead slag was used as substitute of the river sand in concrete. The lead slag contains lead as free metal and various mineralogical components (e.g. $Ca_2PbSi_3O_9$, $Ca_2Al(AlSiO_7)$) (Fig. 1).

The leachate from crushed lead slag contains hazardous contaminants, namely Mo and Cu upper the regulated limit (Mo = 0.2 mg/l, Cu = 0.6 mg/l, for L/S = 0.1) [26]. These heavy metals may be immobilized into cementitious matrix of the concrete. Also, must be noted the low content of the lead slag in concrete.

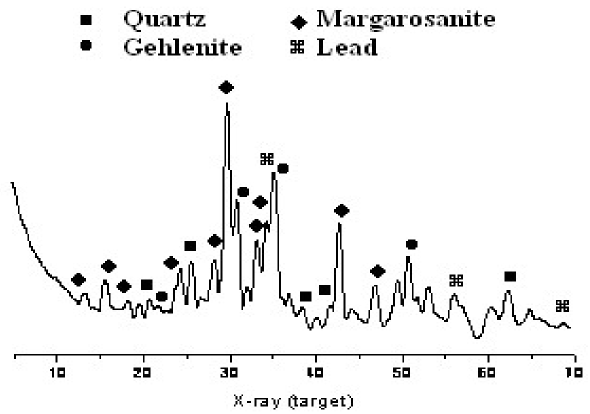

Fig. 1 - X-ray diffraction of lead slag / *Difracția de raze X a zgurii de plumb.*

The recycled cathode ray tube (CRT) glass was utilized as a fine aggregate for concrete. The CRT glass wastes have an increasing importance as a pozzolanic addition. Glass chemical composition and average fineness are important parameters in view of development of performance in binders and concretes [27].

By using granular glass wastes in mortars/concretes involve a threat due to its high potential of expansion because of reaction between alkali contained in the pore solution, reactive amorphous or poorly crystallized silica present in certain aggregates, and water. Consequently, we tested the potential reactivity of CRT glass using chemical test, according to Romanian standard SR 5440-2009 [28].

The glass is crushed and sieved (0.15…0.30 mm). This material is then reacted with an alkaline solution (1N NaOH) at 80°C. At 24 hours, the amount of dissolved silica from the aggregate and the reduction in alkalinity of the solution are measured. These data are plotted against a provided curve. The aggregate falls into one of three ranges: innocuous, deleterious, or potentially deleterious. The values highlight the innocuous behavior of CRT in the ASR chemical test (domain A) [29].

The river sand, hematite and steel shot particle size distribution is presented in Figure 2.

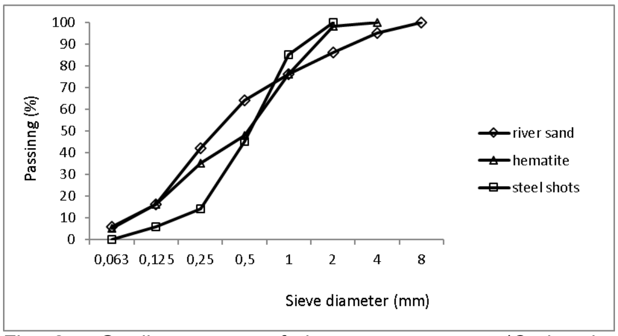

Fig. 2 - Grading curve of the some aggregate/*Curba de granulozitate a unor agregate.*

The aggregates used into concrete are characterized by the physical properties presented in Table 3.

Table 3

Physical characteristics of aggregates/*Caracteristici fizice ale agregatelor*

Aggregates/*Agregate*	Characteristics/*Caracteristici*		
	Specific gravity / *Densitate absolută* (kg/m^3)	Bulk density / *Densitate în grămadă* (kg/m^3)	Water absorbtion/ *Absorbţia de apă* (%)
River sand/ *Nisip de râu*	2600	1550	1.2
Barite/*Barită*	3500	-	0.25
CRT glass/*Sticlă CRT*	2850	1450	-
Steel shot/*Alice de oţel*	7300	-	0.1
Lead slag/*Zgură de plumb*	3300	1880	0.4
Hematite/*Hematit*	4165	1470	-

Table 4

Mix proportion of concrete series /*Compoziția betoanelor studiate*

Codes/*Cod*			B	AHB	ANB	AGB	ZNB
Components/*Componenți*							
Cement /Ciment CEM II AV 42.5 (dm^3/m^3)			133.3	133.3	133.3	133.3	133.3
Filler/*Filer* (dm^3/m^3)	Barite/*Barită*		25.2	25.2	-	-	-
	Limestone/*Calcar*		-	11.8	36.2	35.1	36.2
	Total volume		25.2	37	36.2	35.1	36.2
Aggregates/ *Agregate* (dm^3/m^3)	0/4	River sand /*Nisip de râu*	-	73	201.2	-	201.1
		Steel shot /*Alice de oţel*	-	66.5	174.3	174.6	-
		Hematite /*Hematit*	-	66.5	-	-	-
		Barite/*Barită*	495.5	113	-	-	-
		CRT glass/*Sticlă CRT*	-	-	-	201.5	-
		Lead slag/*Zgură de plumb*	-	-	-	-	174.3
		Total volume/Volum total	495.5	319	375.5	376.1	375.4
	4/16	Barite/*Barită*	303.9	345.8	295.1	291.5	93.9
		Lead slag/*Zgură de plumb*	-	-	-	-	201.1
		Total volume/Volum total	303.9	345.8	295.1	291.5	295
Superplaticizer admixture (SP)/*Aditiv superplastifiant* (%)			1.6	1.6	1.6	1.6	1.6
w/c // *a/c*			0.66	0.58	0.38	0.38	0.44

2.2. Concrete mixes

The special concrete series were designed with the same type and a constant dosage of cement and different water/binder (w/b) ratios. A superplasticizer (SP), included in the carboxilated polyethers class, was used at a constant ratio of 1.6 wt.% comparative to the amount of cement for all the heavyweight concrete mixtures.

The mixtures details are given in Table 4.

A good mixture design and an efficient mixing procedure are two of the key issues to obtain desired performance in hardened state of HWC. When the mixing procedure is not well stablished, a poor workability and heterogeneity in the resulting product inevitably occurs. The materials were placed in the mixer in the following sequence: aggregate, followed by mixture of cement and filler (separately homogenized for 2 min.) and mixed for 2 min. Approximately, 70% of the water was added and mixed for 2 min; the superplasticizer with the rest of the water were added in the mixer and mixed for 2-3 min.

Concrete mixes were casted into 150x150x150 mm cubic molds and vibrated. After 24 h the samples were demolded and kept in water of 20 ± 2^0C until the time of testing.

2.3. Methods

The fresh and hardened concretes were monitored by: density, workability, compressive strength and ultrasonic pulse velocity (US). All these characteristics were measured after standard procedures [30-36]. The compressive strength and ultrasonic pulse velocity of concrete samples were tested after 28, 90, 180 and 360 days of curing in water.

The linear attenuation coefficients of studied concretes at different gamma energies (Co-60:1.25 MeV, Cs-137: 0.662 MeV, Ir-192: 0.37 MeV) in narrow beam conditions have been measured. To investigate the effect of heavy aggregates in concrete composition on gamma radiation shielding properties, the photon attenuation was determined by measuring the ratio of transmission of the penetrating radiation through concrete samples (equivalent dose rate ratio). Plotting the logarithmic form of equation 1 versus different depth photons penetrating (x), a straight line was obtained. The linear attenuation coefficient, μ, was obtained using the value of the slope. The radioactive source was shielded by pin-hole collimators to achieve the narrow beam condition. The measurements were repeated 10 time in order to decrease the statistical errors.

Gamma ray attenuation coefficients, μ (cm^{-1}) have been evaluated according to the exponential attenuation low.

$$I(x) = I_0\, e^{-\mu x} \qquad (1)$$

where: I_0 is the incident intensity of photons penetrating a layer of material of thickness x and I is the emerging intensity of photons.

3. Results

3.1. The characteristics of the fresh concretes

Concrete mixes were cohesive, without showing the phenomenon of segregation and separation of water.

The density and workability are controlled by the mix parameters namely filler types, aggregate types and water/binders ratios. The concretes density was varied from 2890 to 3390 kg/m^3.

The workability of concretes measured as slump was varied with concrete mix design from 0 to 45 mm.

3.2. The characteristics of the hardened concretes

The apparent density reflects the concrete mixes components. The apparent density values of concretes with barite as coarse aggregates and steel shot, CRT glass and lead slag as substitutes of the river sand or filler in concrete were into range from 2870 to 3430 kg/m^3. The concrete density values are showed in Figure 3. The concrete density is decisive determined by the aggregate density, since the concrete contains 75-80 vol.% aggregates.

The compressive strengths were tested. The obtained results are graphically showed in Figure 4.

The following aspects can be found:

- The compressive strength of the concrete has an evolution significantly influenced by the type of aggregate with high density.
- The compressive strength had an increasing trend for all studied concrete, regardless type of aggregate (barite, CRT glass waste, lead slag, hematite and steel shot).
- The long-term compressive strength of the concrete seems to be favored by the presence of CRT glass waste (AGB concrete). This is the

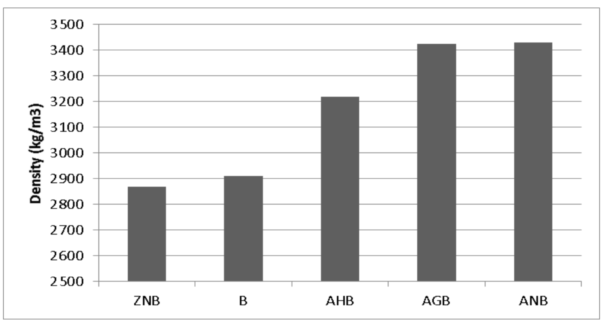

Fig. 3 - The apparent density of the concretes with various aggregate types/ *Densitatea aparentă a betoanelor cu agregate diferite.*

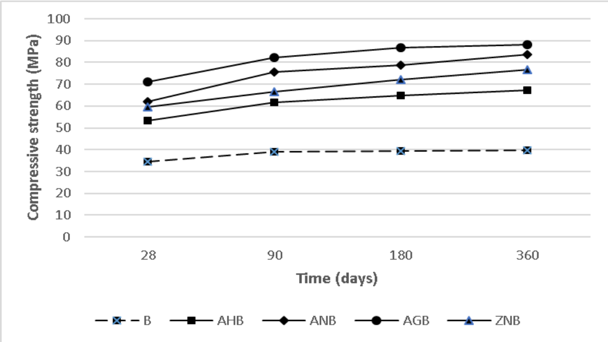

Fig. 4 - The compressive strengths evolution of the concretes in time/ *Evoluția rezistenței la compresiune a betoanelor în timp.*

effect of the pozzolana character of the CRT glass microparticles with contribution to formation of new hydrosilicate phases by the pozzolanic reaction.

Ultrasonic pulse velocity (US) in concrete assesses its homogeneity, the incidence or absence of internal defects, cracks and segregation. The obtained results are graphically presented in Figure 5. The ultrasonic pulse velocity increases with the density and the compactity of the concrete. Since the concrete density is directly influenced by the aggregate density, this means that the ultrasonic pulse velocity increases together with the aggregate density.

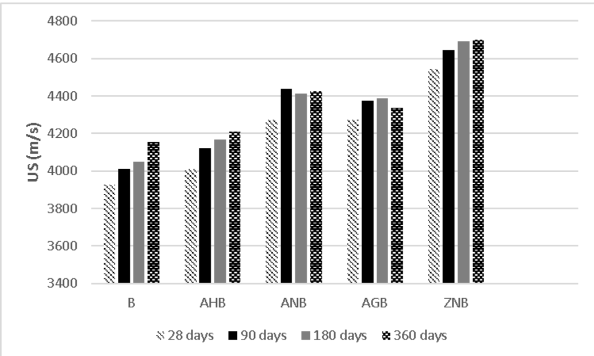

Fig. 5 - The ultrasonic pulse velocity evolution of the concretes/ *Evoluția vitezei de propagare a ultrasunetelor.*

The linear attenuation coefficients decrease with the energy photon emission (Fig. 6). It appears that concrete AGB with barite, CRT glass and steel shot aggregates had the highest linear attenuation coefficient for all energy gamma radiation (Co-60, Cs-137, Ir-192). These results reflect the contribution of the elements with great atomic number, Z as Ba (Z = 56). Also, the high density of the concrete was significantly influenced by the steel shot aggregate as partially substitute of the siliceous sand.

The results showed that concrete with complex aggregate based on barite, steel shot and CRT glass waste have had the highest linear attenuation coefficient of 0.192 cm^{-1} for Co-60, higher by 18.2% than concrete with barite as total aggregate.

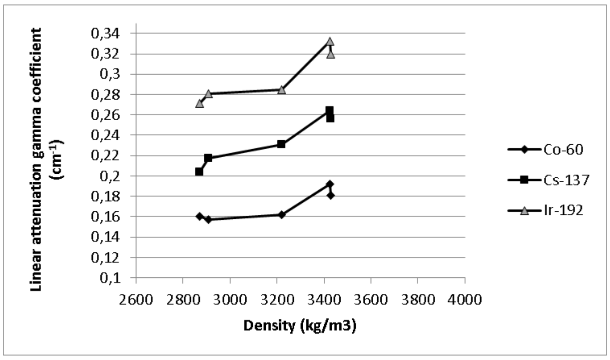

Fig. 6 - Linear attenuation coefficient vs concrete density/*Coeficientul linear de atenuare în corelație cu densitatea.*

The shielding properties of studied concrete, concretized by the linear attenuation coefficients, are favored firstly by the high density. The experimental data showed that shielding properties are influenced by the concrete homogeneity (existing gaps, cracks) as according of the ultrasonic pulse velocity results (Fig. 5).

In Figure 7 is showed the correlation between velocity pulse values and the gamma linear attenuation coefficient of the concretes, for 28 days. The concrete with lead slag aggregate is characterized by highest velocity pulse value, but this is irrelevant for the linear attenuation coefficient.

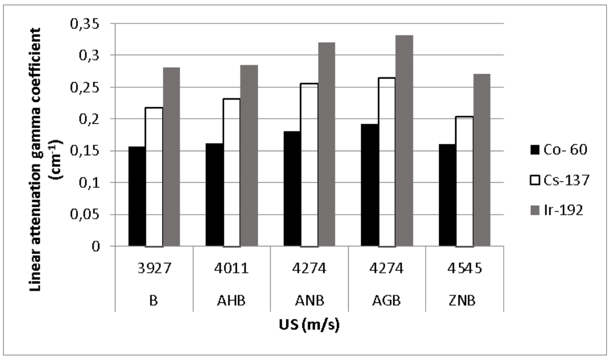

Fig. 7 - Linear attenuation coefficient vs concrete ultrasonic velocity pulse /*Coeficientul linear de atenuare în corelație cu viteza de propagare a ultrasunetelor.*

Generally, there is a good correlation of the gamma attenuation coefficient increase with the ultrasonic velocity of the concrete with the barite aggregate, with the exception of the concrete with lead slag as a substitute for the coarse barite.

4. Conclusions

This study evaluated the combined effects of different variables such as aggregate type (barite, CRT glass, lead slag, hematite and steel shot), the filler type (limestone, barite) and the different water to binder ratios on the characteristics of HWC. Based on the above discussions, the following conclusions were drawn:

-The density of the concrete was influenced by the aggregate type. Thus, the total or partial substitution of natural sand with steel shot, barite, hematite and lead slag leads to an increase in the concrete density at over 2870 kg/m^3.

- The compressive strength of the concrete had an evolution significantly influenced by the type of aggregate with high density. The compressive strength of the heavyweight concrete with barite, CRT glass, lead slag, hematite and steel shot is continuously ascending. The compressive strength of the concrete with CRT glass as sand is significantly advantageous by the presence of the pozzolanic addition.
- The ultrasonic pulse velocity in concrete assesses its homogeneity, the incidence or absence of internal defects, cracks and segregation.
- The linear attenuation coefficients decrease with the energy photon emission. The results showed that concrete with complex aggregate based on barite, steel shot and CRT glass waste have had the highest gamma linear attenuation coefficient of 0.192 cm^{-1} for Co-60, higher by 18.2 % than concrete with barite as total aggregate.

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