

## RECYCLING OF RADIOACTIVE CONCRETE RESULTED FROM DECOMMISSIONING OF VVR-S NUCLEAR RESEARCH REACTOR, ROMANIA

RADU DEJU<sup>1</sup>, CLAUDIU MAZILU<sup>2</sup>, DANIELA GURAU<sup>1,3,\*</sup>, MONICA MINCU<sup>1</sup>

<sup>1</sup>“Horia Hulubei” National Institute for Physics and Nuclear Engineering, POB MG-6,  
Bucharest-Magurele, 077125, Romania

<sup>2</sup>Technical University of Civil Engineering, Bucharest, 020396, Romania

<sup>3</sup>Academy of Romanian Scientists, Bucharest, 030167, Romania

\*Corresponding author: daniela.gurau@nipne.ro

*Received June 23, 2017*

*Abstract.* The presented study is part of a research project that has as main purpose recycling of coarse and fine aggregates from radioactive concrete, as rubble pre-placed and mortar for filling containers and the stabilization of the radioactive waste. The leaching behaviour of <sup>137</sup>Cs, <sup>60</sup>Co and <sup>152</sup>Eu radionuclides was studied in aqueous medium, for the radioactive mortar samples made from recycled aggregates compared to samples made of natural aggregates. Recycling of the radioactive concrete allows the minimization of the low level radioactive concrete volume that should be disposed, in accordance with all the applied legal requirements.

*Keywords:* crushed concrete, leaching behaviour, concrete aggregates, reuse and recycling, radioactive waste.

### 1. INTRODUCTION

During the operation of the nuclear installations, the concrete from their structure can become radioactive through contamination or activation. The decommissioning of the nuclear installations can produce important quantities of radioactive waste, from which, the majority is represented by the concrete waste. At present, in Europe, a large amount of concrete need to be recycled annually, following the 2030 year, this amount will grow. From the published data [1], it can be observed that in the European Union many decommissioning projects will be developed in 2025–2040 period. The final disposal of these radioactive waste will decrease the storage capacity and will increase the costs, following a lower economic feasibility. Until now, in Romania, the low level radioactive waste (LLW) are preplaced as blocks in containers and solidified with mortar obtained with natural fine aggregates. This method provides a degree of filling of the LLW per container of approximately 50 vol.%.

Ishikura and the co-authors [2, 3] showed that the recycling of the radioactive concrete can be a major solution for reducing the volume of radioactive waste, aiming to save natural resources and environmental protection. Deju and the co-authors [4–7] have elaborated in details an overview of the methods applicable in the recycling of the radioactive concrete.

Starting with 1957, IFIN-HH from Magurele, own a VVR-S nuclear research reactor that used distilled light water as moderator, coolant and reflector, having 2 MW reactor thermal power. The reactor has functioned using EK-10 (10%  $^{235}\text{U}$  enrichment) and S-36 (35%  $^{235}\text{U}$  enrichment) fuel assemblies. The reactor was in operation until 1997, when was stopped and passed in conservation until 2001 when the shut-down was decided. In 2010, the decommissioning project started, planned to be finalized in 2020. The radioactive concrete waste that will be generated from the dismantling of the reactor structure (*e.g.*: biological shield of the reactor core and hot cells) represents an estimated amount of about 70 tons. In addition, 1000 tons of non-radioactive concrete will be generated during the decommissioning project. This will create a lower economic feasibility and will increase also the final disposal capacity.

The authors present an innovative method for the immobilization/ recycling of the low level radioactive concrete (contaminated or activated) in the form of pre-placed rubble (sort of 16/50 mm) and fine aggregates (sort of 0/5 mm) in the mortar needed to fill the voids in containers with radioactive waste. It is expected that the designed process will provide a substantial increase of filling ratio for radioactive waste containers, up to 75 vol.%. In this way, the final volume of disposed waste can be significantly reduced producing economic effects. To achieve the objectives of the proposed project, the fine recycled aggregates and the mortar that use these aggregates must meet the technical requirements applicable for the low level radioactive waste. These are: (i) diameter of fine aggregates  $\leq 5$  mm; (ii) the mortar fluidity need to meet ASTM C939 standards [8] regarding the flowing time of 16÷50 sec through a standardized flow cone; (iii) the maintaining the fluidity mortar >60 minutes with limitation until the mortar bleeding stop; (iv) the degree of filling of the container with mortar  $\geq 95$  vol.%; (v) the proportion of recycled fine aggregates used in mortar  $\geq 900$  kg/m<sup>3</sup>; (vi) compressive strength of cured mortar  $\geq 30$  N/mm<sup>2</sup> (300 kgf/cm<sup>2</sup>) after 28 days; (vii) the leaching index of the cured mortar  $\geq 6$ .

During the presented research activities, many laboratory testing were carried out [5–7, 9–13]. Various tests were analysed in order to identify which recipe meet the proposed specification for fluidity and allow the obtaining of the highest fill ratio for mortar. The parameters that influence the increasing of pre-placed rubble fill ratio and mortar fill ratio were analysed by optimal properties identification of mortar. Couple of tested are presented to confirm results obtained in the laboratory. There is verified the influence of the recycled concrete characteristics for fresh and cured mortar and of the modality of filling the container taking into account the

degree of filling with rubble. Also, it is determined the filling degree of mortar in containers of 100 liters, the compressive strength of composite cubes and the leaching index of radionuclides  $^{137}\text{Cs}$ ,  $^{60}\text{Co}$  and  $^{152}\text{Eu}$  of cured mortar.

## 2. EXPERIMENTAL

### 2.1. MATERIALS

For the experimental preparation of the filling mortar, the recycling aggregates with sort of 0/2.5 and 0/5 mm were used, obtained from C16/20, C25/30, and C35/45 reference concrete. Their recipes and the properties for fresh and cured mortar are detailed in the paper publish by Deju and co-authors [5, 6]. The Portland composite cement type CEM II/B-M 42.5 (Lafarge Romania) was used for normal concrete. BASF MasterGlenium 27 high range water reducing admixture was used for concrete. This additive has as the main benefit, the ability to work with a very low water/cement ratio and still obtain extended workability retention. Another additive that was used in the tests was the BASF MasterMatrix SDC 150 which is a high performance viscosity modifying agent (VMA) for fluid concrete. Is important to specify that the initial tests [6] were made with CEM V A(S-V) 42.5 cement (Lafarge Romania), BASF MasterGlenium 27 and BASF RheoMATRIX 150. Now, these materials were taken out of production. The pre-placed rubble used in the tests was kept 24 hours completely immersed in water and drained 5 minutes before use. In Table 1 are presented the grain size distribution for pre-placed rubble with sort 16/50 mm and 0/5 mm and for mortar prepared with fine aggregates with sort 0/5 mm, both obtained from C25/30 reference concrete.

Table 1

Grain size distribution

Grain size distribution					
pre-placed rubble		fine aggregates			
sort (mm)	% wt.	sort (mm)	% wt.	sort (mm)	% wt.
45/50	18.6	2.5/5	18.31	0.3/0.6	7.71
31.5/45	50.5	1.25/2.5	17.75	0.15/0.3	19.48
16/31.5	30.9	0.6/1.25	18.73	0/0.15	18.02

### 2.2. CHARACTERISTICS INFLUENCE OF RECYCLED REFERENCE CONCRETE ON FRESH AND CURED MORTARS

In any process of demolition and recycling of concrete structures, the material characteristics are not the same in all the volume. To check the easiness

with which can be determined the optimal flow of mortars obtained with recycled fine fraction, the properties of fresh mortars and those resistance to compression were determined using the mortars recipes type presented in Table 2.

*Table 2*

Percentage by weight for mortar recipe type

Recipe group	Aggregate type	Sort (mm)	Percentage by weight
F	recycled	0/5	100% C25/30 concrete
G	recycled	0/2.5	100% C25/30 concrete
H	recycled	0/2.5	66.6% C25/30 and 33.4% C35/45 concrete
I	recycled	0/2.5	33.4% C25/30 and 66.6% C35/45 concrete
J	recycled	0/2.5	33.3% C16/20, 33.3% C25/30 and 33.4% C35/45 concrete
K	recycled	0/2.5	33.3% C16/20 and 66.6% C25/30 concrete
L	recycled	0/2.5	50% C16/20, 25% C25/30 and 25% C35/45 concrete
M	recycled	0/2.5	25% C16/20, 25% C25/30 and 50% C35/45 concrete
O	recycled	0/2.5	66.6% C16/20 and 33.4% C25/30 concrete
S	recycled	0/2.5	100% C35/45 concrete
T	recycled	0/2.5	100% C16/20 concrete
U	recycled	0/2.5	20% C16/20, 60% C25/30 and 20% C35/45 concrete

### 2.3. FILLING MODALITY INFLUENCE ON THE FILLING DEGREE OF CONTAINER WITH RUBBLE

There have been made four pre-placed tests for each manual and mechanized procedures, for rubble with sort 16/50 mm that was obtained from recycling of C25/30 concrete. The tests were made using as container a bucket of 22 liters. For the mechanical procedure, a rectangular funnel with  $31 \times 41.5$  cm top opening and  $10.5 \times 20$  cm bottom opening. To check the filling modality influence, the volume of voids was determined by replacing the voids with water. 2000 ml, 1000 ml and 100 ml graded cylinders with divisions of 20 ml, 10 ml and 1 ml were used to evaluate the exact quantity of water that replace the void from the specific container.

### 2.4. COMPRESSIVE STRENGTH OF COMPOSITE CUBES

In order to evaluate the compression strength of the C25/30 concrete samples made from coarse aggregate with sort 16/50 mm and mortar that contain fine fraction with sort 0/5 mm, six cubs with dimensions of  $15 \times 15 \times 15$  cm were made. Three sampled were prepared with vibrant concrete and three with non-vibrant. The frequency for vibration was of 50 Hz with a 30 sec duration time. The vibration procedure determine the concrete mixture to liquefy, reducing the internal friction of the components of cement, aggregate, and water. This allows the

concrete mixture to move around more easily in the specific container used. This procedure is a useful technique to eliminate the voids. Moreover, the vibrations and liquefaction allow the air bubbles to escape from the mixture. The air bubbles decrease the concrete mixture density.

For the preparation of the samples, it was used a steel device layer type in order that the mortar will envelope the rubble preventing it from reaching the surface of the container used. In this way, the cube is coated with a layer of mortar of at least 5 mm (in order to avoid the wall effect). All the cast concrete samples were removed from the molds after 1 day and kept in water at  $20 \pm 2$  °C, until the compression tests were started in the 28 day using a 3000 kN hydraulic press. The preparation and the preservation of specimens and of compression request was made in accordance with the current standards.

#### 2.5. CONTAINER FILLING DEGREE WITH MORTAR

Couple of samples were used for each mortar recipe used in tests regarding the container filling degree with mortar. In Table 3 are presented the composition of the fresh mortar mixtures.

*Table 3*  
Composition of the fresh mortar mixtures

Recipe group	Recipe	Quantity (kg)			Ratio water to cement	Ratio sand to cement	Super plasticizing additive (%)	Viscosity modifier additive (%)
		Water	Cement	Sand				
F	F1	0.65	1	1.3	0.65	1.3	1	0.5
	F2	0.60	1	1.3	0.60	1.3	1	0.5
	F3	0.55	1	1.3	0.55	1.3	1	0.5
	F4	0.50	1	1.3	0.50	1.3	1	0.5
	F5	0.53	1	1.3	0.53	1.3	1	0.5
	F6	0.65	1	1.7	0.65	1.7	1	0.5
	F7	0.60	1	1.7	0.60	1.7	1.5	1.0
	F8	0.54	1	1.3	0.54	1.3	1	0.5
G	G1	0.60	1	1.3	0.60	1.3	1	0.5
	G2	0.60	1	1.3	0.60	1.3	1	0.5
	G3	0.55	1	1.3	0.55	1.3	1	0.5
H	H1	0.60	1	1.3	0.60	1.3	1	0.5
	H2	0.55	1	1.3	0.60	1.3	1	0.5
I	I1	0.60	1	1.3	0.60	1.3	1	0.5
	I2	0.55	1	1.3	0.55	1.3	1	0.5
J	J1	0.55	1	1.3	0.55	1.3	1	0.5
	J2	0.60	1	1.3	0.60	1.3	1	0.5
K	K1	0.55	1	1.3	0.55	1.3	1	0.5
L	L1	0.55	1	1.3	0.55	1.3	1	0.5
M	M1	0.55	1	1.3	0.55	1.3	1	0.5
O	O1	0.55	1	1.3	0.55	1.3	1	0.5

Table 3  
(continued)

S	S1	0.55	1	1.3	0.55	1.3	1	0.5
	S2	0.55	1	1.3	0.55	1.3	1	0.5
T	T1	0.55	1	1.3	0.55	1.3	1	0.5
	T2	0.60	1	1.3	0.60	1.3	1.5	1
	T3	0.58	1	1.3	0.58	1.3	1	0.5
	T4	0.57	1	1.3	0.57	1.3	1	0.5
U	U1	0.55	1	1.3	0.55	1.3	1	0.5

The Marsh cone test was done to study the rheological properties of cement mixtures. The flow time of the cement mixture through Marsh cone is an indicator of viscosity which can be measured using a Marsh funnel, by observing the time it takes for a known volume of cement mixture to flow from a cone through a short tube. 1 litre of cement mixture was made to flow through the March cone after 0, 15, 30 and 60 minutes of mixing, and time in seconds was measured using a stopwatch. In Table 4 are presented the results obtained during this test.

Table 4  
Properties of the fresh mortar mixtures

Recipe group	Recipe	Marsh flow time (s)				Density (kg/m <sup>3</sup> )	Observation
		min					
		0	15	30	60		
F	F1	15	16	-	-	1905	Homogeneous, too fluid
	F2	20	21	-	-	1934	Homogeneous, fluid
	F3	25	30	-	-	1959	Homogeneous, fluid
	F4	45	-	-	-	2007	Homogeneous, fluid
	F5	33	-	-	-	2004	Homogeneous, fluid
	F6	21	21	23	24	1991	Homogeneous, fluid
	F7	35	-	38	-	1937	Homogeneous, fluid, $T_s = 0.019\text{cm}$
	F8	28	28	29	31	2040	Homogeneous, fluid, $T_s = 0.013\text{cm}$
G	G1	28	-	29	-	1969	Homogeneous, fluid
	G2	20	-	-	-	1974	Homogeneous, fluid
	G3	29	31	30	33	2040	Homogeneous, fluid, $T_s = 0.013\text{cm}$
H	H1	25	-	29	-	1975	Homogeneous, fluid
	H2	27	28	28	30	1975	Homogeneous, fluid, $T_s = 0.016\text{cm}$
I	I1	20	-	-	-	2001	Homogeneous, fluid
	I2	30	30	32	33	2040	Homogeneous, fluid, $T_s = 0.009\text{cm}$
J	J1	26	26	28	29	2034	Homogeneous, fluid, $T_s = 0.022\text{cm}$
	J2	18	-	-	-	1993	Homogeneous, fluid
K	K1	29	29	29	30	2027	Homogeneous, fluid, $T_s = 0.013\text{cm}$
L	L1	28	29	31	32	2024	Homogeneous, fluid, $T_s = 0.013\text{cm}$
M	M1	28	30	31	32	2028	Homogeneous, fluid, $T_s = 0.016\text{cm}$
O	O1	27	28	27	29	2060	Homogeneous, fluid, $T_s = 0.022\text{cm}$
S	S1	29	30	31	32	2026	Homogeneous, fluid, $T_s = 0.016\text{cm}$
	S2	30	31	31	32	2032	Homogeneous, fluid
T	T1	70	79	-	-	2116	Homogeneous, fluid

Table 4  
(continued)

	T2	17	-	-	-	1989	Homogeneous, fluid
	T3	23	25	26	28	2023	Homogeneous, fluid, $T_s = 0.025\text{cm}$
	T4	26	27	27	30	2006	Homogeneous, fluid, $T_s = 0.016\text{cm}$
U	U1	25	25	26	28	2023	Homogeneous, fluid, $T_s = 0.013\text{cm}$

The amount of water accumulated at the surface, named bleeding, is normal and expected on freshly placed concrete. This does not necessarily have or negative effect on the quality of the plastic or hardened concrete. However, excessive bleeding can lead to some performance problems. The bleeding rate ( $T_s$ ) was analysed and expressed in terms of cubic of water bleed after 3 hours, per square centimetre of sample surface. The values obtained are presented in Table 4. The rate of bleeding is controlled by the permeability of the plastic paste and the flow of mater is controlled by the permeable space or capillaries between particles, after the solid particles are settled.

The attempts of filling with mortar using the recipe F8 were performed on three types of containers named CB, CP and CPR type. The mortar was poured out in containers with a speed of 25 l/min. For the CB case, a 100 litter container made from steel ( $D = 43.5$  cm and  $h = 68$  cm) was used. Three such container were used for working at a filling height of 63 cm (effective volume of 93.58 litters). The tests made on those containers were done using a mixture of recycled rubble with sort 16/50 mm, C25/30 concrete block (dimensions of  $19 \times 19 \times 30$  cm) and mortar with fine fraction. On the bottom of each container, a stand (dimensions of  $10 \times 10 \times 5$  cm) was placed for supporting the concrete block. Then, rubble with sort 16/50 mm was placed inside up to the superior part of stand. A concrete block was placed on the stand and rubble was placed in the space between the block and the container wall and up to the superior part of the block. For the CP case, was used the same 100 litter container type like in the CB case. The mixture used included rubble with sort 16/50 mm and mortar with recycled fine fraction (recipe F8). The CPR container was made from PVC with 8 litter volume (dimensions of  $D = 18.5$  cm,  $h = 28.5$  cm). The filling height was of 28 cm and an effective volume of 8 litters. The mixture used in this case was made from rubble with sort 16/50 mm, recycled from C25/30 concrete and mortar with recycled fine fraction prepared with the addition of  $^{137}\text{Cs}$ ,  $^{60}\text{Co}$  and  $^{152}\text{Eu}$  chlorides solutions. The mortar prepared in a 20 l bucket using an AEG PN 3000 SUPER X2 equipment (with a speed of 50 rot/min and a power of 1010 W) was poured out up to the superior part of the container. Solid mortar constituents (fine aggregates and cement) have been mixed without liquids for 3 min. 2/3 from the quantity of water and  $^{137}\text{Cs}$ ,  $^{60}\text{Co}$  and  $^{152}\text{Eu}$  chlorides solutions have been added and mixed again for other 3 min. After that, 1/3 from the quantity of water and super plasticizing and viscosity modifier additives have been added and continuously mixed for 6 min.

## 2.6. LEACHING TESTS

The parameter used to categorize the effectiveness of a matrix material named leachability index was studied in this paper. The leaching test was performed evaluating the cumulative fraction leached (CLF), the diffusion coefficient ( $D$ ) and the leachability index ( $L$ ). The cumulative fraction leached (CLF) was evaluated using eq. (1):

$$CLF = \frac{\sum \Delta_n / \Delta_i}{V/S} \quad (1)$$

where:  $\Delta_i$  is the initial sample activity at zero time (Bq);  $\Delta_n$  is the activity leached out of sample after the leaching time, (Bq);  $V$  is the sample volume (cm<sup>3</sup>) and  $S$  is the sample surface (cm<sup>2</sup>);  $t$  is the renewal period of leaching agent (15, 22, 30, 37, 45, 52, 60, 90 days).

The Long-term behaviour of the radionuclides in the leaching process from the waste matrix was evaluated determining the diffusion coefficient, calculated based on the Fick's second law which predicts how diffusion causes the concentration to change with time. The diffusion coefficient for radionuclides is calculated using the return slope,  $m$ , of the linear regression line through data points in  $\sum \Delta_n / \Delta_i$  and  $\sqrt{t}$ . The diffusion coefficient ( $D$ ) was evaluated using eq. 2:

$$D = \frac{\pi}{4} m^2 \frac{V^2}{S^2} \quad (2)$$

where:  $D$  is the diffusion coefficient (cm<sup>2</sup>/day);  $m$  is the slope ( $\sum \Delta_n / \Delta_i, \sqrt{t}$ ) ( $d^{-1/2}$ );  $V$  is the sample volume (cm<sup>3</sup>);  $S$  is the sample surface (cm<sup>2</sup>).

Based on the diffusion coefficient, the leachability index ( $L$ ) was evaluated using eq.3:

$$L = \log(D). \quad (3)$$

For studying the leachability, four samples were artificially contaminated with <sup>137</sup>Cs, <sup>60</sup>Co and <sup>152</sup>Eu radionuclides, according to the methodology presented in detail by Deju and co-authors [7]. The characteristics of the mortar composition used for samples are presented in Table 5.

Table 5

Mortar composition and leachability index for test samples

Type of aggregates	Quantity (kg)			Leachability index ( $L$ )		Curing time (days)
	water	cement	sand	<sup>137</sup> Cs	<sup>60</sup> Co	
natural	0.45	1	0.8	10.2	12.4	28
fine recycled	0.54	1	1.3	10.3	12.2	28
natural	0.45	1	0.8	9.64	12.2	56
fine recycled	0.54	1	1.3	9.84	12.3	56



For the samples preparation, a quantity of 44.5 ml of distilled water and 61.5 ml of  $^{137}\text{Cs}$ ,  $^{60}\text{Co}$  and  $^{152}\text{Eu}$  chlorides solutions was used for samples with natural aggregates, and 48 ml of distilled water and 59.82 ml of  $^{137}\text{Cs}$ ,  $^{60}\text{Co}$  and  $^{152}\text{Eu}$  chlorides solutions was used for samples made from recycled aggregates. The samples have a weight of approximately 150 g, a diameter of 8 cm, a thickness of 1.5 cm and a medium density of  $1.9 \text{ g/cm}^3$ . After the specific curing time, each sample was immersed in 400 ml of distilled water. The activity of the leaching agent (distilled water) was measured after, 15, 22, 30, 37, 45, 52, 60, 90 days with a laboratory gamma-ray system from Ortec which consist of a high purity germanium detector (model GEM60P4-95), a DSPEC jr. 2.0 Digital gamma-ray spectrometer and a lead castle. The main performance specifications of the detector warranted by the producer are: relative efficiency 60%, resolution (FWHM) 1.95 keV, peak-to-Compton ratio 70:1, peak shape (FWFM/FWHM) 3.0, all evaluated at 1.33 MeV peak of  $^{60}\text{Co}$ .

### 3. RESULTS AND DISCUSSIONS

The influence of recycled reference concrete characteristics on fresh and cured mortars was studied. In Table 6 are presented the mass, density and flexural strength characteristics for the mortar test specimens (ts) with dimensions of  $4 \times 4 \times 16 \text{ cm}$ .

Table 6

The mass, density and flexural strength

Recipe group	Recipe	Mass (g)			Average density ( $\text{kg/m}^3$ )	Flexural strength		
		ts1	ts2	ts3		Force (daN)		
						ts1	ts2	ts3
F	F1	518.0	516.2	512.0	2013	500	436	444
	F2	530.4	521.4	531.4	2061	448	462	452
	F3	530.2	527.6	536.6	2076	492	464	418
	F4	559.0	558.8	572.0	2200	520	570	526
	F5	550.0	559.2	542.2	2150	400	512	390
	F6	532.4	514.3	522.0	2043	452	463	411
	F7	554.0	552.4	542.2	2147	460	460	428
	F8	497.4	549.2	546.4	2074	528	400	484
G	G1	534.8	531.2	534.8	2084	404	396	424
	G2	523.1	536.2	530.8	2070	421	399	416
	G3	500.8	550.4	549.6	2084	528	464	472
H	H1	545.2	538.6	541.4	2116	426	462	420
	H2	550.3	546.3	533.6	2123	452	436	440
I	I1	551.2	557.4	555.2	2166	486	506	502
	I2	543.4	563.7	561.5	2173	512	523	489
J	J1	547.2	534.4	554.6	2130	468	446	430
	J2	507.9	528.4	543.2	2057	456	421	432
K	K1	544.6	542.0	543.8	2123	436	490	484

Table 6

(continued)

L	L1	537.2	542.6	548.0	2119	486	490	504
M	M1	549.4	538.4	552.2	2135	470	458	456
O	O1	540.8	542.8	541.4	2116	450	416	474
S	S1	555.0	558.8	549.8	2166	448	448	484
	S2	493.0	546.4	542.0	2059	506	460	464
T	T1	576.0	567.4	571.2	2232	610	550	624
	T2	543.8	547.7	532.8	2115	589	560	575
	T3	546.2	548.0	553.2	2145	530	542	472
	T4	487.0	539.4	524.8	2019	560	450	454
U	U1	543.4	543.2	556.2	2139	528	520	518

The test specimens were the subject of flexure. In the upper side, under the direct charge, the specimen was the subject of the compression and in the lower part shows the efforts in stretch. In Fig. 1 are presented the values obtained for the average flexural strength and the average compression strength, in  $\text{N/mm}^2$ . The mean value was around  $11 \text{ N/mm}^2$  for flexural strength and  $35 \text{ N/mm}^2$  for compression strength.

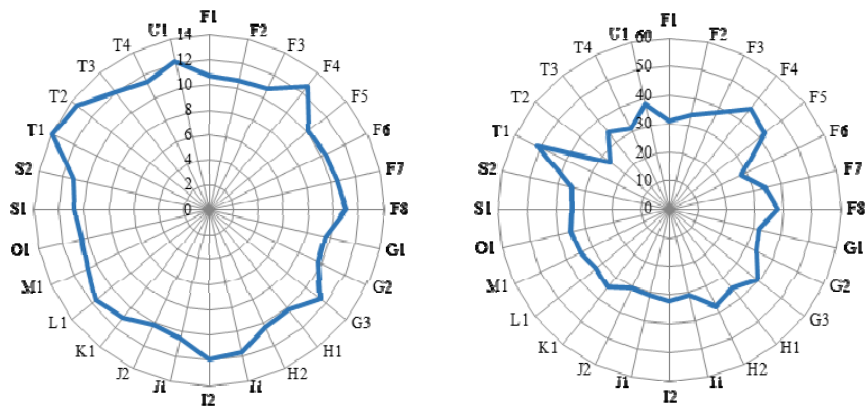


Fig. 1 – The average flexural strength (left); the average compression strength (right).

In Table 7 are presented the values obtained for the compression strength for the two prism scraps (noted as 1<sup>st</sup> and 2<sup>nd</sup> sp) obtained for each prismatic test specimens (ts) after the flexure tests.

Table 7

## The compression strength

Recipe group	Recipe	Compression strength					
		Force (daN)					
		ts1		ts2		ts3	
		1 <sup>st</sup> sp	2 <sup>nd</sup> sp	1 <sup>st</sup> sp	2 <sup>nd</sup> sp	1 <sup>st</sup> sp	2 <sup>nd</sup> sp
F	F1	4550	4800	5050	5300	5100	5200
	F2	5300	5400	5650	5600	5450	5500
	F3	6250	6050	5850	5520	6200	6525
	F4	6100	6800	6900	7600	8100	7925
	F5	7100	6400	6900	5700	7400	7225
	F6	4950	4625	4300	4600	3950	4200
	F7	5425	6200	5325	5250	5250	5300
	F8	6450	5800	5575	6750	4900	5900
G	G1	4550	4700	5450	5600	5825	4700
	G2	4950	5200	5450	5350	5700	5550
	G3	6150	6350	6500	6400	6425	6300
H	H1	5650	5950	5625	5700	5550	5600
	H2	6150	5900	6150	6025	5750	5950
I	I1	5000	5000	4975	5100	5000	4500
	I2	5200	5050	5350	5250	4850	5050
J	J1	5375	5000	4825	4875	4600	4925
	J2	4900	5150	4900	4850	4550	4825
K	K1	5700	5350	5200	5800	5800	5500
L	L1	4825	5125	5400	5500	5625	5125
M	M1	5025	5450	5550	5725	5675	5600
O	O1	5550	5450	5850	5950	5250	5925
S	S1	4475	5450	6200	5625	5375	5750
	S2	5450	5650	5750	5700	5825	5325
T	T1	8725	7850	8875	8925	7750	7225
	T2	4350	4200	4500	4150	4100	4250
	T3	5725	5475	5925	5200	5500	5450
	T4	4600	4950	5300	4700	5500	4900
U	U1	5950	6050	5750	6050	6175	6275

From the analysis performed, no essential changes were observed for fresh mortars properties. Instead, for different compositions of samples, the optimum flow can be obtained relatively easily (F8, G3, H2, I2, J1, K1, L1, M1, O1, S1, T4, and U1). A different behaviour shows the fine aggregate with sort 0/5 mm obtained from C25/30 concrete and that with sort 0/2.5 mm obtained from C16/20 concrete. These two type of concrete are more powdery and requires more water. The data obtained lead to the idea that large lots made from recycled aggregated can be made requiring a good homogeneity, for which the mortar recipes can be determined relatively easily, ensuring the optimal flow. It can be observed that to all optimal flow values of mortars, strong compression strength were corresponding to the prismatic test specimens involved in the study. The

compression strength obtained meet the requirement of the technical specification, such as resistance to compression  $\geq 30 \text{ N/mm}^2$  of cured mortar after 28 days. It should be noted that when the cement with valid exceeded is used, this has a negative influence on the flowing of the fresh mortar due to the particle agglomeration and the decreasing of the specific surface.

Studying the influence of the filling method of the container on the degree of filling with rubble, was found that the volume of voids in volume are from 10.72 up to 10.90 liters of water for manual filling tests and from 11.12 up to 11.22 liters of water for mechanized filling tests. From the analysis of the data obtained was revealed that the mechanized filling leads in decreasing the rubble filling level of the bucket. Also, was observed that both filling methods displays good reproducibility of results. The outcomes obtained for testing the resistance to compression of the composite cubs indicate that the average resistance of compression was 32.5 MPa for the vibrant mortar and 33.6 MPa for non-vibrant mortar. Was observed that the breaking of the test specimen at compression test was realized in principle *via* mortar reinforced injection, the value of the compression resistance of the concrete being in both cases over the minimum of 30 MPa compression resistance limit imposed for mortar.

After the tests regarding the container filling degree with mortar were performed, the densities values (expressed in  $\text{kg/dm}^3$ ) and the percentage by weigh (%wt.) were obtained for fresh mortar. The results for densities were from 2.050 up to 2.075 for CB, from 2.050 up to 2.075 for CP and 2.050 for CPR containers type. The results obtained for %wt. were from 98.9 to 99.6 for CB, from 99.2 up to 99.7 for CP and 99.2 for CPR containers type. For all cases, the degree of container filling has fulfil the specific condition. For the container with 17.65 kg, gamma-ray spectrometry measurement were performed using a special gamma-ray spectrometry system named ISOCART, used especially for radioactive waste drums assay. The results expressed in Bq, obtained for  $^{137}\text{Cs}$ ,  $^{60}\text{Co}$  and  $^{152}\text{Eu}$  radionuclides were 4.19E+04, 4.10E+04 and 1.24E+04 respectively. Dosimetry measurements were also performed to ensure the radiological security using a dose rate equipment form Berthold, composed from LB 123 monitor and LB 1236 probe. The mean value was of  $2\mu\text{Sv/h}$ . Based on the average apparent diffusion coefficient, the leachability index for the diffusing species was calculated for  $^{137}\text{Cs}$  and  $^{60}\text{Co}$  radionuclides and the data are presented in Table 5.  $^{152}\text{Eu}$  radionuclide display a limited solubility, being kept in the cement matrix.

The leaching index value is not influenced by the duration curing time. The leachability index values are bigger for  $^{60}\text{Co}$  radionuclide than  $^{137}\text{Cs}$  radionuclide, both for samples obtained with natural aggregates and recycled aggregates. The samples obtained with recycled aggregates displays for  $^{137}\text{Cs}$  radionuclide values of the leachability index smaller than those obtained with natural aggregates.

Following the values obtained for the leaching index ( $L$ ) for all samples, was observed that the cement matrix used for mortar preparation present hydraulic and

chemical properties (leachability index  $> 6$ ). It means that it is proper to be a matrix material for wastes cementation. As Ochs and co-authors observed [14],  $^{152}\text{Eu}$  radionuclide has a lower solubility in the cementation systems. The experimental results have revealed that the europium ions are retained almost 100% in cement by the calcium silicate hydrate. This explains the lack of activity in solution.

#### 4. CONCLUSIONS

After the tests were conducted, was observed that recycled concrete radioactive waste can be used as rubble and fine powder aggregates. It has been found that for different composition of the fine recycled aggregate, the mortars that use this type of aggregate can be easily optimised from the point of view of the flow characteristics and resistance to compression. The volume of voids is grater for the mechanized filling than the manual filling. The breaking of the composite cubes made from rubble with sort 16/50 mm and mortar made following the F8 recipe, was done during the compression tests. The value of the compression resistance of the concrete was above the minimum acceptable value for concrete of 30 MPa. The densities obtained for fresh mortar were from 2.050 up to 2.075 kg/dm<sup>3</sup> and the filling degree from 98.9 up to 99.7 %wt.

During the tests made, the leachability index had higher value than 6, for all the samples made both with natural aggregates and fine recycled. This demonstrates that the composition used is proper to be a matrix material for waste cementation. The europium ions are kept almost 100% by the calcium silicate hydrate indicating the lack of activity in solution.

*Acknowledgements.* This work was supported by the Romanian National Authority for Scientific Research, CNDI-UEFISCDI, in the framework of 145/02.07.2012 project.

#### REFERENCES

1. European Commission, *Nuclear Safety and the Environment, 1998. Recycling and Reuse of radioactive Material in the Controlled Nuclear Sector*, Report EUR 18041.
2. T. Ishikura, D. Oguri H, S. Abe, K. Ohnishi, *Development of Recycling Techniques for Nuclear Power Plant Decommissioning Waste*, 11<sup>th</sup> International Conference on Nuclear Engineering, Tokyo, Japan, 20–23 April 2003; doi: 10.1080/18811248.2004.9715541.
3. T. Ishikura, H. Ueki, K. Ohnishi, D. Oguri, *Utilization of Crushed Radioactive Concrete for Mortar to Fill Waste Container Void Space*, Journal of Nuclear Science and Technology **41**(7), 741–750 (2004); doi: 10.1080/18811248.2004.9715541.
4. R. Deju, M. Dragusin, I. Robu, C. Mazilu, C. Tuca, *Review on Radioactive Concrete Recycling Methods*, Romanian Reports in Physics **65**(4), 1485–1504 (2013).
5. R. Deju, I. Robu, M. Dragusin, C. Mazilu, C. Tuca, *Selection tests for recycled radioactive sand obtaining method*, Romanian Reports in Physics **67**(2), 673–692 (2015).

6. R. Deju, I. Robu, M. Dragusin, C. Mazilu, C. Tuca, *Tests regarding filling performance of the mortars obtained by radioactive recycled sand*, Romanian Reports in Physics **67**(3), 1159–1175 (2015).
7. R. Deju, D. Gurau, L. Done, C. Mazilu, I. Robu, *The Study of Radionuclides Leaching from Mortar Made with Natural Aggregates and Recycled Aggregates Arising from Decommissioning of RN VVR-S*, Romanian Reports in Physics **68**(4), 1466–1481 (2016).
8. ASTM International, ASTM C939/C939M-16a, *Standard Test Method for Flow of Grout for Preplaced-Aggregate Concrete (Flow Cone Method)*, West Conshohocken, PA, 2016, [www.astm.org](http://www.astm.org).
9. C. Mazilu, I. Robu, R. Deju, *Characteristics of the mortars obtained by radioactive recycled sand*, 16<sup>th</sup> International Multidisciplinary Scientific Geo-conference SGEM (indexed in Thomson Reuters ISI, Web of Knowledge, SCOPUS, etc.), Book 4: Energy and Clean Technologies, Nuclear Technologies **1**, 3–10, Albena, 2016.
10. I. Robu, C. Mazilu, R. Deju, *Performance tests on filling with mortar the cementing containers of radioactive concrete waste*, 3<sup>th</sup> International Congress Water, Waste and Energy Management, Extended Abstracts book, Rome (Italy), July 2016.
11. I. Robu, C. Mazilu, R. Deju, *Aspects concerning the use of recycled concrete aggregates*, 20<sup>th</sup> Innovative Manufacturing Engineering & Energy International Conference (ImanEE 2016), IOPscience doi:10.1088/1757-899X/161/012072, Kallithea Chalkidiki, Greece, September 2016.
12. I. Robu, C. Mazilu, R. Deju, *Study concerning characterization of some recycled concrete aggregates*, Mathematical Modelling in Civil Engineering **12**(1), 1–12 (2016); doi: 10.1515/mmce2016 0001.
13. I. Robu, C. Mazilu, R. Deju, *Studiu privind caracterizarea unor agregate reciclate din beton*, 2<sup>nd</sup> National Conference on Building Engineering, 16 October 2015, Bucharest (Romania).
14. M. Ochs, D. Hager, S. Helfer, B. Lotenbach, *Solubility of Radionuclides in Fresh and Leached Cementitious Systems at 220C and 500C*, Mat. Res. Soc. Symp. Proc. **506**, 773–780 (1998).