BABEŞ-BOLYAI UNIVERSITY CLUJ NAPOCA

PhD thesis

RADON TRANSPORT THROUGH BUILDING MATERIALS

Summary

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2011

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Key words: radioactivity, addition, cement, concrete, admixtures, exhalation, difussion

1. INTRODUCTION

The purpose of this thesis is to highlight the features of building materials used in the radon transport. Considering that, of all building materials, concrete seems to contribute best to the increase of contained radon concentration, this thesis presents the influence of concrete features on the exhalation and increase of radon concentration. Different concrete mixtures were created using various types of cement and the influence of cement additions on the radon exhalation was surveyed.

2. SCOPE OF RESEARCH

The main scope of this thesis is to determine the influence cement additions, composition factors and strength and durability features of concretes made with various cement types on radon diffusion, on the exhalation rate from concretes made with various cement types and respectively on the indoor radon concentration.

I have designed three research programs, the first program consisting in the determination of influence of cement additions, cements and respectively aggregates on concrete's radioactivity. This was made by testing additions included in cement composition, by testing cements made with various additions and respectively by preparing and testing concretes made with these cements to assess the influence of additions and ultimately of cements on concrete's radioactivity.

The second research program consisted in the correlation of strength (compressive strength) and durability (porosity, water permeability and air) features of concretes achieved with cements including various additions and admixtures to the radon exhalation rate and respectively contained radon concentrations.

In the third program I have prepared concrete mixtures using two types of added cements which I tested for efficiency as barriers against radon.

Measurements were made using a radon measurement system Pylon AB-5, comprising a radon monitor type Pylon AB-5, LUCAS cells, passive counter type CPRD together with a special box determining the radon exhalation, a system that was acquired under the project "Technical solutions for the protection of current and new buildings in Romania exposed to radioactive emissions, an essential requirement in providing hygiene and health of population, in consistency with European legislation", funded by the Ministry of Education and Research within the national program for research-development and innovation AMTRANS – Territory Arrangement and Transports [3].

3. THESIS STRUCTURE

This thesis is structured in seven sections, briefly described below.

Section 1 shortly presents the radon sources. The factors influencing radon migration from source towards indoor air, as well as the factors limiting the gas migration from soil to the building, are presented.

Also the factors influencing radon emissions from building materials are underlined.

Section 2 shows the features and particulars of radon, entry paths, as well as factors favoring transport of radon indoor. Factors influencing indoor and outdoor radon rate variations are also illustrated.

Section 3 determines the features and properties of ingredient materials of concrete elements and concretes. Particulars of concrete microstructures, specific features (porosity, permeability, diffusion), as well as factors affecting the above are approached. Also, a brief description is given of the cement hydration mechanism, of the manufacturing process as well as of particulars and features of additions used in cement manufacturing process (slag, puzzolana, fly fly-ash, limestone).

The other ingredients of concrete, aggregates, water and admixtures are shortly described, highlighting their respective roles in the concrete mix.

This section outlines the main concrete features that are of significance for radon emissions: water / cement ratio, gel/ space ratio, pores volume, pores system, porosity, permeability, diffusion and relations between the above.

Section 4 illustrates the features of radon emissions from concrete, as well as the factors influencing the radon exhalation from concrete. Thus, a synthesis of surveys, research works and regulations present on international level is presented and, *in extenso*, the results of experimental surveys destined to determine the influence of various additions used in cements on concrete's radioactivity; of a correlation between the cement dose, water / cement ratio, strength and durability features of concrete and the radon exhalation rate; on the influence of water / cement ratio, concrete density and permeability on the radon diffusion.

Section 5 illustrates the contribution of concrete and other construction to the increase of indoor radon concentration. In this section, results achieved for the indoor radon concentration are presented with reference to the radon exhalation rate for concrete with various types of blended cement.

Section 6 underlines radon reduction methods for existent buildings, as well as technical - constructive methods for new buildings exposed in areas with radioactive potential, highlighting the particulars and efficiency of each method.

Also, results achieved in testing the efficiency of radon barriers are presented, which are applied on concretes made with two types of blended cement.

Section 7 is reserved for the overall conclusions resulted following the survey of specialized literature, as well as conclusions based on experimental research performed.

Hereinafter I shall illustrate briefly the results of experimental research performed.

4. EXPERIMENTAL RESEARCH

4.1. Influence of composition factors and strength and durability features of concretes on radon emission

4.1.1. Influence of additions in cements and of Romanian cements on concrete's radioactivity

The research program consisted of tests on additions included in the composition of cements present on Romanian market, of various blended cements and respectively the preparation of concretes using such cements to assess the influence of additions, implicitly cements, on concrete's radioactivity.

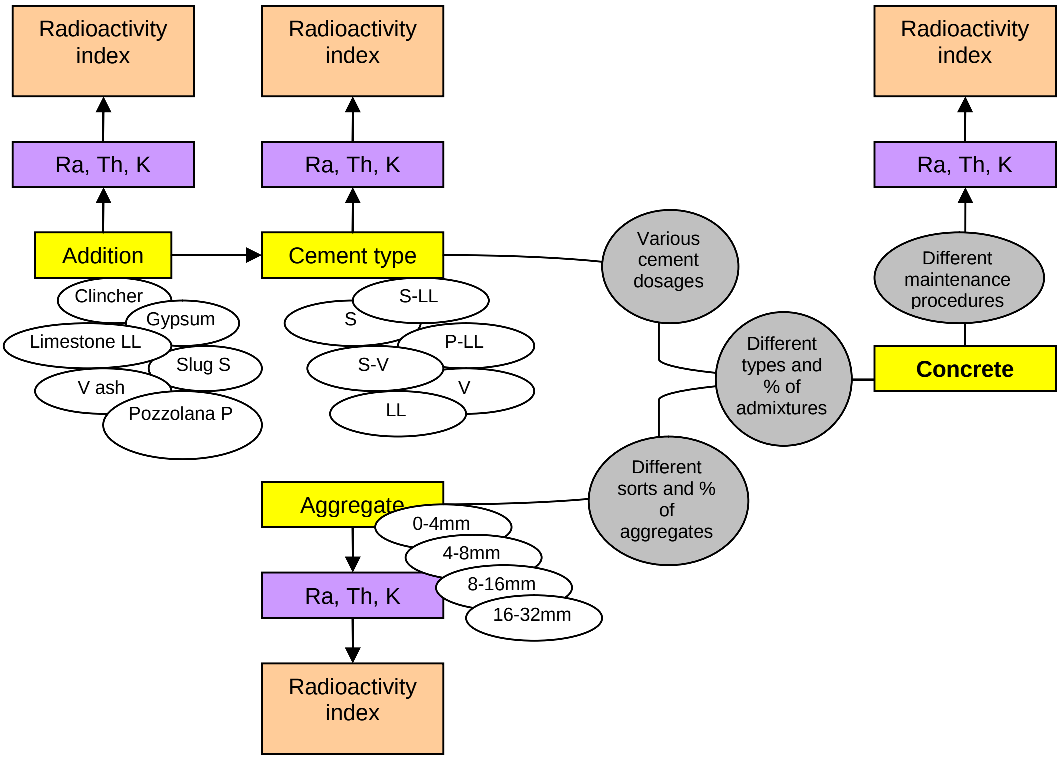


Fig. 4.1.1 – Flow chart of the research program on the determination of influence of cement additions, cements and aggregates on concrete's reactivity

The results achieved in the determination of concentrations of the main radioactive elements (Ra-226, Th-232, K-40) and of the radioactivity index are presented below for various additions used in cements, blended cements and concretes prepared therewith.

1. Additions

In the assessment of the radioactivity of additions used in cements, measurements were taken through gamma spectroscopy with HP detector (Ge). Thus measurements were taken for the main radioactive elements in various compounds used in cements.

Results achieved for the concentrations of the main radioactive elements for various ingredients used in cements manufacturing are presented herein and synthesized in figure 4.1.2.

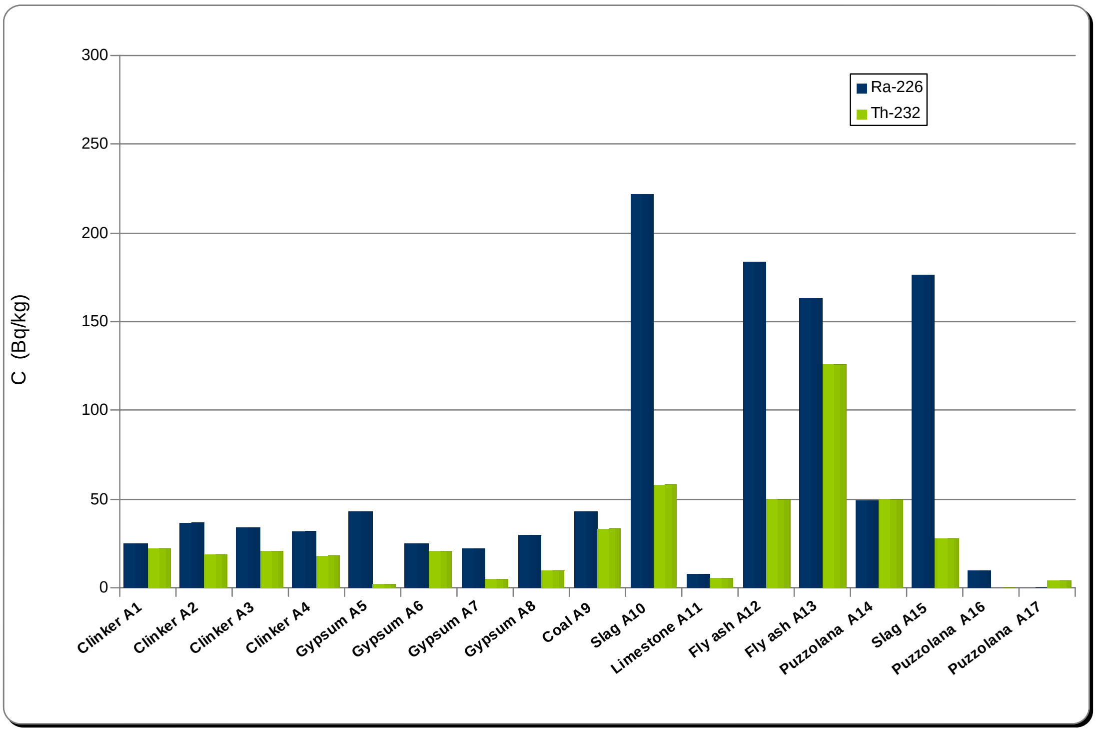


Fig.4.1.2 – Concentrations of radium and thorium activities

For the same type of component, results achieved vary based on the different generating sources. Also, figure 4.1.2 displays the high values of radionuclides for slag (A10, A15) and fly-ash (A12, A13) compared to those obtained for limestone (A11).

2. Cements

The contribution of tested additions was assessed by adding them in the manufacturing process of different cement types. Thus, tests were performed on no-addition cements (CEM I 42,5 R) and cements with additions of slag, fly-ash, limestone, slag + limestone, puzzolana + limestone, fly-ash + slag. Tested cements are manufactured in Romania, by different manufacturing plants, and the addition rate used for the same cement type vary from one plant to another.

Therefore, in assessing the radioactivity of cements that is due to additions included therein, concentrations of radionuclides - Ra, Th and K – were measured and radioactivity indexes of various cement types were determined.

Values achieved for the concentration of radionuclides activities are displayed graphically in figure 4.1.3, compared to the maximum permitted value of radium in cement, regulated by Ministry of Health Order no. 51 / 1983 [97].

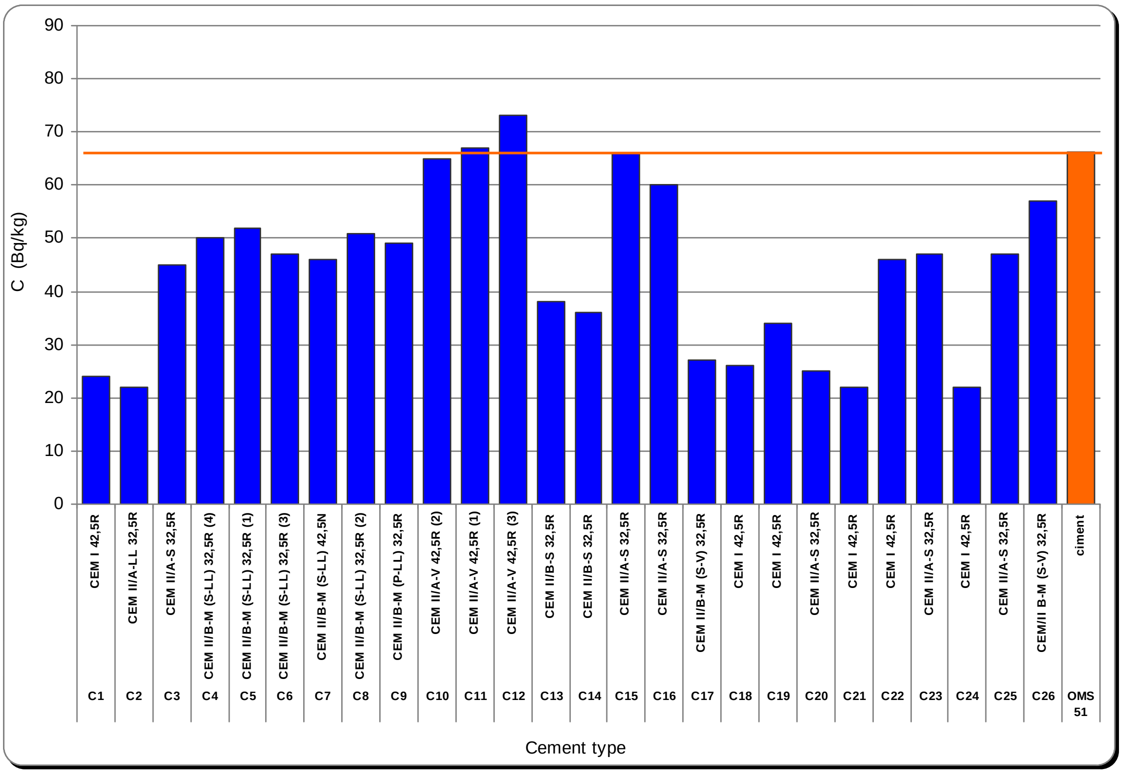


Fig.4.1.3 – Concentrations of radium activities for various cement types

Cements with fly-ash additions (II/A-V 42,5R) display radium concentration values closed to the maximum value permitted in Romania. In case of II/A-S 32,5 R cement, values achieved are observed to be dependent on the slag source and respectively on the percentage used in cement manufacturing processes (6-20%).

In case of Th-232 and K-40 radionuclides, all tested cement types display values far inferior to the maximum value permitted in Romania.

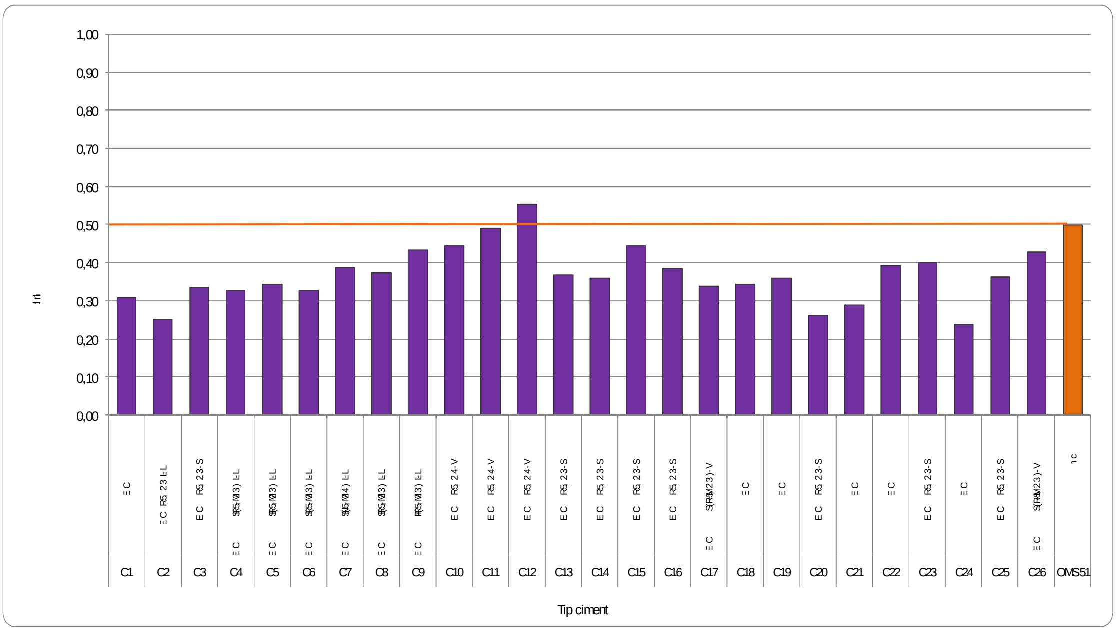


Fig. 4.1.4 – Values of radioactivity index for various cement types

Cement C12, with fly-ash additions, presents a radioactivity index 10% higher than the maximum value permitted in Romania. Fly-ash-added cements, C10 and C11, present values (0,45 and respectively 0,49) that are close to the maximum value permitted for the radioactivity index.

The other tested cement types displayed radioactivity index values below the maximum permitted value.

3. Aggregates

In the preparation of concretes, aggregates with sorts of 0-4 mm, 4-8 mm, 8-16 mm and 16-32 mm were used; the shares of aggregates used, as well as results of radioactive isotopes measurements performed on aggregates quantities between 157 and 177 g are shown in table 4.1.1.

Table 4.1.1 – Results of radioactive isotopes measurements for various aggregate sizes used in the preparation of concretes

Aggregate size (mm)	Aggregate percentages used in the preparation of concretes		Concentration of radionuclides activity (Bq/kg)			Radioactivity index I
	Concrete code B1-B4, B14-B20	Concrete code B5-B13	Ra-226	Th-232	K-40	
0-4	35%	45%	10,4	11,5	506	0,26
4-8	15%	23%	9,2	10,0	528	0,26
8-16	21%	32%	10,0	13,0	555	0,28
16-32	29%	-	6,8	10,0	292	0,17
Maximum permitted value as per MHO no. 51/1983			**29,97**	**91,39**	**869,5**	

In case of aggregates, values achieved for the 4 sorts, for Ra-226, Th-232 and K-40 radionuclides are observed to be far inferior than the maximum values permitted in Romania.

Based on the percentages of aggregates used in the preparation of concretes and respectively of cements used in each tested concrete type, the concentration of radionuclides activity due to concrete ingredients can be assessed.

4. Concretes

In the preparation of concretes, the folowing were used:

- Cements with additions of puzzolana (P), slag (S), limestone (LL), fly-ash (V);
- River aggregates, sorts 0-4 mm, 4-8 mm, 8-16 mm, 16-32 mm, granularity area 3, favorable, in accordance with NE 012-1/2007 [4];
- Plasticizing / superplasticizing admixtures or air entrainment, depending on the case.

The research program consisted in the determination of concentrations of Ra-226, Th-232 and K-40 radionuclides in concretes prepared with some cement types with additions.

Among the cement types surveyed, those having the highest values for radium activity concentration and respectively for radioactivity index were selected in order to enable the assessment of concretes prepared therewith. Thus, the following cement types were used: C10, C11, C12 (CEM II/A-V 42,5 R, with various fly-ash percentages and sources), C15 (CEM II/A-S 32,5 R, with slag additions), C4, C5, C8 (CEM/II B-M (S-LL) 32,5 R with various slag and limestone concentrations) and C9 (CEM/II B-M (P-LL) 32,5 R, with puzzolana and limestone additions).

So, concerts were prepared with blended cements (Portland cements with additions that vary between 21-35%: CEM II/B-M (S-LL) 32,5 R, CEM II/B-M (P-LL) 32,5 R, Portland cements with additions between 6-20%: CEM II/A-V 42,5 R si CEM II/A-S 32,5 R), in various cement dosages, using different types and concentrations of admixtures (superplasticizing / plasticizing, air entrainment) and aggregates with maximum granulation of 16 mm and respectively 32 mm. The cement types, cement dosages and admixtures used in concrete preparation, the overall quantity of aggregates used as well as the water / cement ratio are illustrated in table 4.1.2.

Table 4.1.2 – Compositions of concretes prepared with various cement types

Concrete code	Cement type	Cement dosage (kg/m^3)	Admixtures (% of the cement quantity)	Water (l)	Aggregate (kg)	Maximum aggregate granulation (mm)	Water / cement ratio
B1	CEM II/B-M (P-LL) 32,5R	350	0,4% plasticizer	161,0	1718,0	32	0,46
B2	CEM II/B-M (P-LL) 32,5R	350	0,4% plasticizer	161,0	1718,0	32	0,46
B3	CEM II/B-M (S-LL) 32,5R (1)	320	1% superplasticizer	160,0	1752,0	32	0,50
B4	CEM II/B-M (S-LL) 32,5R (1)	400	1% superplasticizer	160,0	1646,0	32	0,40
B5	CEM II/A-V 42,5R (1)	470	0,7% superplasticizer	211,5	1589,0	16	0,45
B6	CEM II/A-V 42,5R (2)	350	0,7% superplasticizer	192,5	1733,0	16	0,55
B7	CEM II/A-V 42,5R (1)	350	0,7% superplasticizer	189,0	1733,0	16	0,54
B8	CEM II/A-V 42,5R (2)	470	0,7% superplasticizer	211,5	1589,0	16	0,45
B9	CEM II/A-V 42,5R (3)	350	0,7% superplasticizer	189,0	1733,0	16	0,54
B10	CEM II/A-V 42,5R (3)	470	0,7% superplasticizer	206,8	1589,0	16	0,44
B11	CEM II/A-V 42,5R (2)	400	0,05% entrainer+ plasticizer	204,0	1666,0	16	0,51
B12	CEM II/A-V 42,5R (1)	400	0,05% entrainer+ plasticizer	200,0	1666,0	16	0,50
B13	CEM II A-V 42,5R (1)	400	0,05% entrainer+ plasticizer	200,0	1666,0	16	0,50
B14	CEM II B-M (S-LL) 32,5R (2)	340	0,4% plasticizer	159,8	1739,0	32	0,47
B15	CEM II/B-M (S-LL) 32,5R (2)	400	0,4% plasticizer	168,0	1651,0	32	0,42
B16	CEM II/B-M (S-LL) 32,5R (2)	300	0,4% plasticizer	168,0	1773,0	32	0,56
B17	CEM II/B-M (S-LL) 32,5R (3)	320	1% superplasticizer	150,4	1752,0	32	0,47
B18	CEM II/A-S 32,5R	320	1% superplasticizer	160,0	1752,0	32	0,50
B19	CEM II/A-S 32,5R	400	1% superplasticizer	160,0	1646,0	32	0,40
B20	CEM II/B-M (S-LL) 32,5R (3)	400	1% superplasticizer	144,0	1646,0	32	0,36

Concrete samples with sizes of 150x150x150 mm were executed in compliance with the norms in force upon the execution date [4]. They were emerged in water for 2 or 7 days, and then cured until the test age (28 days, 180 days and one year) in air with 20 ^{0}C temperature and 65% humidity.

The activity concentration of Ra-226 varies between 15-25 Bq/kg. The values achieved for Th-232 range between 10-22 Bq/kg, and for K-40 the values range between 250-770 Bq/kg. The values achieved for Ra and Th in concretes are similar to those obtained in different international experimental research [35], being ranked in the lower levels of the range quoted in specialized literature.

Radioactivity indexes for concretes surveyed are presented in figure no. 4.1.5.

Fig.4.1.5 – Values of radioactivity indexes achieved for concretes manufactured with various blended cement types

Concretes prepared with the same cement type, submerged in water for 2 days (B1) display values of the radioactivity index 30% higher compared to concretes cured for 7 days (B2).

For concretes prepared with the same cement type in class 32,5 cured in similar conditions, the radioactivity index values raise with the increase of cement quantity.

In case of concretes prepared with cement in class 42,5 small differences may be noted between the radioactivity index values for different cement dosages.

For the same cement dosage, values achieved for concretes submerged in water for 7 days and then held in air with controlated conditions of temperature and humidity do not display significant differences, as below:

- For cement dosages of 320 kg/m^3, radioactivity index values were achieved between 0,25 and 0,30.
- For cement dosages of 350 kg/m^3, radioactivity index values were achieved between 0,32 and 0,36.
- For cement dosages of 400 kg/m^3, radioactivity index values were achieved between 0,26 and 0,38.

The results acquired during this research program may be synthesized in the chart displayed in figure 4.1.6.

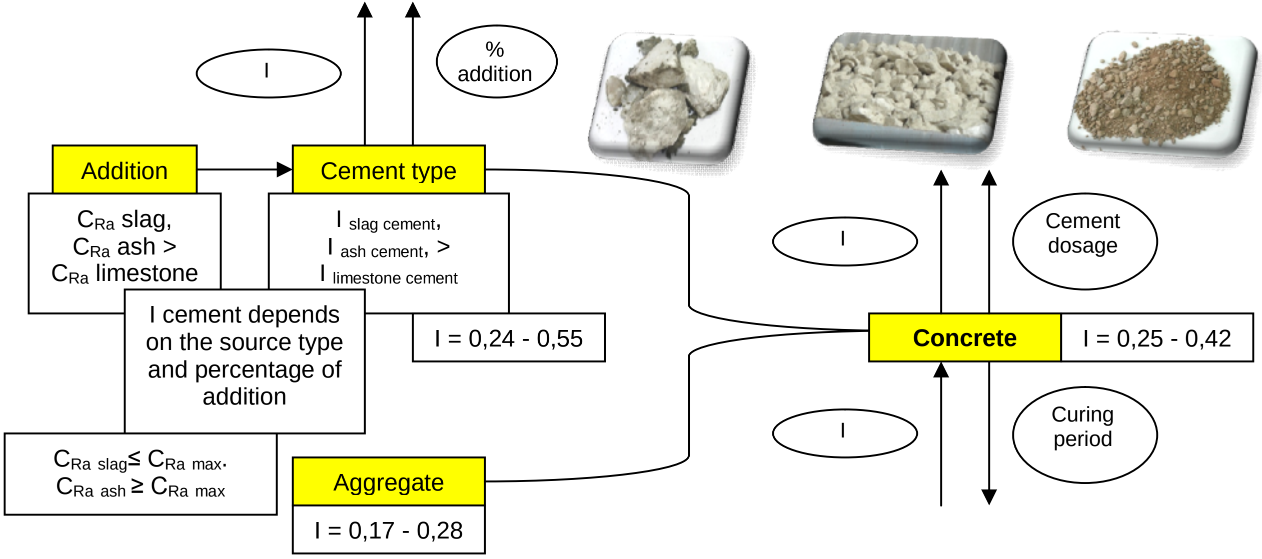


Fig. 4.1.6 – Chart of results acquired for cement additions, cements, aggregates and concretes

4.1.2. Experimental research for the determination of strength and durability features for concretes prepared with various cement types

4.1.2.1. Research program

The research program consisted in the determination of certain strength and durability features (compressive strength, porosity, water and air permeability, etc.) of concretes prepared with cements with various additions and admixtures, in order to cross-reference results achieved with the radon exhalation rates and respectively the values acquired for the indoor radon concentration (Ill.4.1.7).

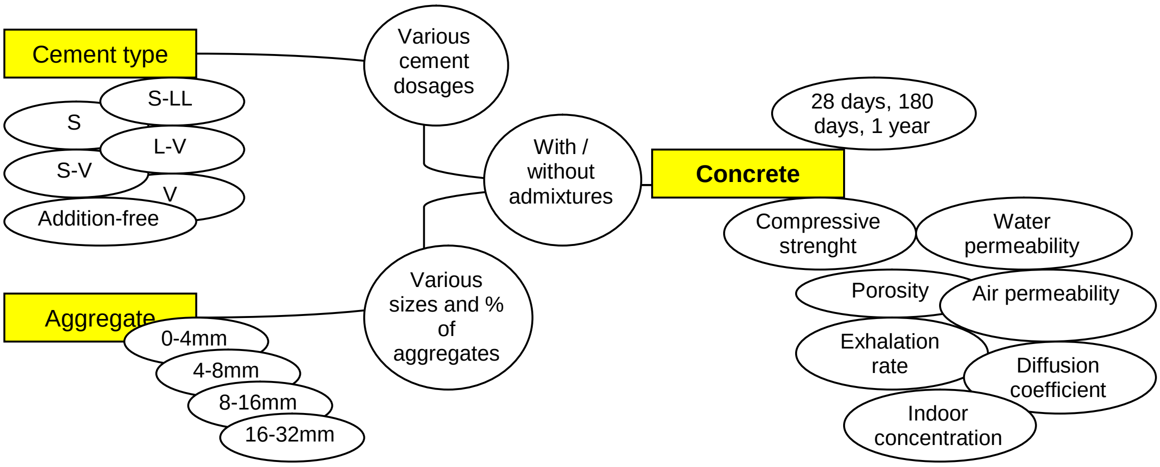


Fig. 4.1.7 – Flow chart for the research program covering the cross-reference of strength and durability features of concretes prepared with cements with various additions and admixtures with the radon exhalation rates and respectively indoor radon concentrations

During the experimental program, concretes in classes C12/15….C20/25 were prepared with cements having various additions of slag, slag + limestone, limestone + fly-ash, no admixtures and respectively concretes in strength classes C18/22,5….C30/37 with cements having various additions of slag, slag + limestone, limestone + fly-ash, slag + fly-ash, fly- ash and various dosages of superplasticizing admixture (S). Also test specimens were prepared from concrete in classes C25/30 and C30/37 with cement added with slag, fly- ash or with no addition and various dosages of air entrainers (A). The entrained air rate was of approximately 3,5%, for concretes prepared with added cement and respectively 4,9%, for concretes prepared with addition-free cement, for the same water / cement ratio of approximately 0,36.

After preparation, the concrete samples were submerged in water for 7 days, and then held in indoor environment at a temperature of 20 ^{0}C and a humidity of approximately 65% until the test date.

The compressive strengths, air and water permeability features were determined for concretes of various compositions after 28 days, 180 days and 1 year after pouring. Also, porosity values and respectively radon exhalation rates were determined from the concrete types surveyed from long-term strength and durability standpoints.

4.1.2.2. Results acquired

Compressive strength

During the research program, the strength features, the air and water permeability were determined on concretes having various compositions, in order to enable cross-reference with radon exhalation rates.

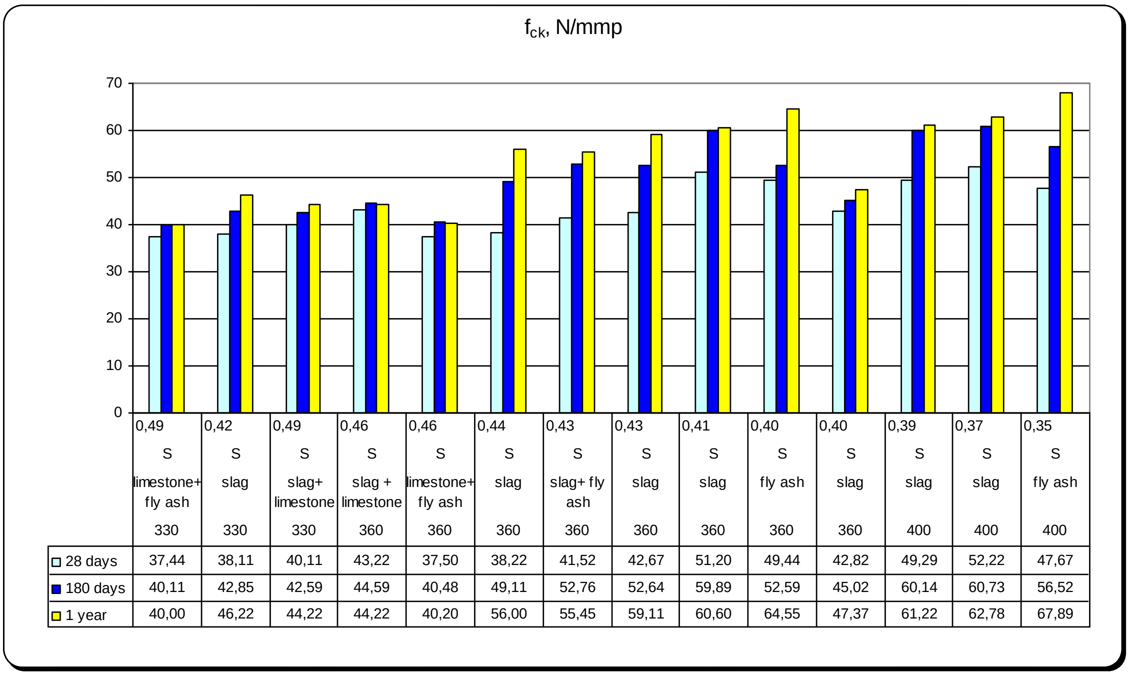
Table 4.1.3 presents the composition and features of fresh concretes (water / cement ratio, entrained air content).

Table 4.1.3 – Composition and features of fresh concretes

No.	Cement addition type	Prescribed concrete class	Cement dosage (kg/m^3)	Admixture	W/C ratio	Entrained air (%)
1	slag	C 12/15	300	-	0,55	-
2	slag + limestone	C 12/15	300	-	0,57	-
3	limestone + fly-ash	C 12/15	300	-	0,58	-
4	slag +limestone	C 16/20	320	-	0,53	-
5	slag	C 18/22,5	340	-	0,49	-
6	slag+ limestone	C 18/22,5	340	-	0,52	-
7	limestone + fly-ash	C 18/22,5	340	-	0,52	-
8	slag	C 20/25	380	-	0,43	-
9	slag+ limestone	C 20/25	380	-	0,48	-
10	limestone + fly-ash	C 18/22,5	330	Superplasticizer	0,49	-
11	slag	C 18/22,5	330	Superplasticizer	0,42	-
12	slag+ limestone	C 18/22,5	330	Superplasticizer	0,49	-
13	slag	C 20/25	360	Superplasticizer	0,44	-
14	fly-ash	C 20/25	360	Superplasticizer	0,40	-
15	slag+ fly-ash	C 20/25	360	Superplasticizer	0,43	-
16	slag	C 20/25	360	Superplasticizer	0,43	-
17	slag	C 20/25	360	Superplasticizer	0,41	-
18	slag	C 20/25	360	Superplasticizer	0,40	-
19	slag+ limestone	C 20/25	360	Superplasticizer	0,46	-
20	limestone + fly-ash	C 20/25	360	Superplasticizer	0,46	-
21	slag	C 30/37	400	Superplasticizer	0,39	-
22	slag	C 30/37	400	Superplasticizer	0,37	-
23	fly-ash	C 30/37	400	Superplasticizer	0,35	-
24	-	C 25/30	450	Air entrainer	0,35	4,9
25	slag	C 25/30	450	Air entrainer	0,36	3,6
26	slag	C 30/37	510	Air entrainer	0,35	3,3
27	fly-ash	C 30/37	510	Air entrainer	0,37	3,8

W/C ratio, cement dosages and curing conditions were consistent with the regulations in force upon the concrete preparation date.

Compressive strengths achieved for concretes prepared with superplasticizing admixture are presented in figure 4.1.8, and the time evolution of compressive strength of concretes prepared with no admixture and respectively with air entrainer is illustrated *in extenso* herein.



	0,49 S limestone+ fly ash 330	0,42 S slag 330	0,49 S slag+ limestone 330	0,46 S slag + limestone 360	0,46 S limestone+ fly ash 360	0,44 S slag 360	0,43 S slag+ fly ash 360	0,43 S slag 360	0,41 S slag 360	0,40 S fly ash 360	0,40 S slag 360	0,39 S slag 400	0,37 S slag 400	0,35 S fly ash 400
□ 28 days	37,44	38,11	40,11	43,22	37,50	38,22	41,52	42,67	51,20	49,44	42,82	49,29	52,22	47,67
■ 180 days	40,11	42,85	42,59	44,59	40,48	49,11	52,76	52,64	59,89	52,59	45,02	60,14	60,73	56,52
□ 1 year	40,00	46,22	44,22	44,22	40,20	56,00	55,45	59,11	60,60	64,55	47,37	61,22	62,78	67,89

Fig. 4.1.8 – Time evolution of compressive strengths of concretes prepared with superplasticizing admixture

From the standpoint of strength features, all concretes assessed correspond to their prescribed classes, displaying higher compressive strengths than those provided for classification.

The most significant increases in compressive strengths between 28 and 180 days are noted in concretes prepared with cement with slag, and the slowest evolution is seen in concretes prepared with cement added with limestone and fly-ash.

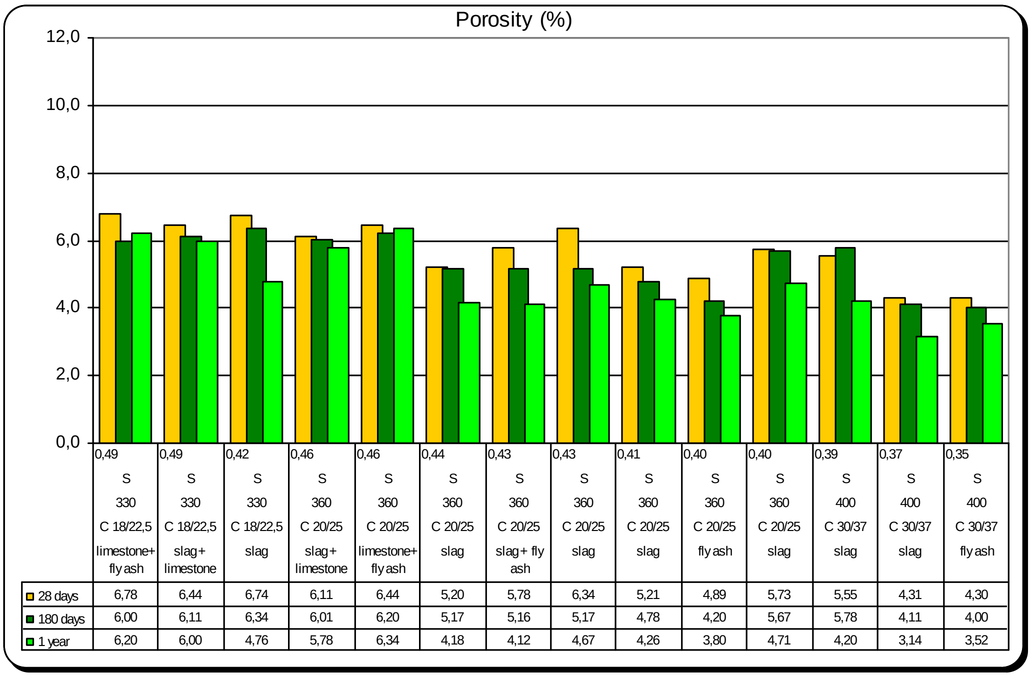
Between 180 days and 1 year, a significant strength increase was achieved in concretes prepared with admixtures and cement with slag and respectively fly-ash additions. Generally, in concretes prepared with cement with limestone and fly-ash additions, a leveling of strengths is seen between 180 days and 1 year.

For the same concrete class, concerts prepared with cements added with slag are noted to have a higher compressive strength compared to other concretes, disregarding the presence / absence of admixtures. Lower compressive strengths were revealed for concretes prepared with cements added with limestone and fly-ash. Certainly, a crucial factor is the cement strength value.

Porosity

The values achieved for concrete porosity at 28 days, 180 days and 1 year are presented in figure 4.1.9. The samples were kept indoor, at a temperature of 20 ^{0}C and humidity of approximately 65% prior the start of porosity tests.

Porosity reduces in time, more significantly in concretes prepared with cements added with slag and respectively fly-ash additions comparative with concretes prepared with cements added with slag and limestone.



	0,49 S 330 C 18/22,5 limestone+ fly ash	0,49 S 330 C 18/22,5 slag+ limestone	0,42 S 330 C 18/22,5 slag	0,46 S 360 C 20/25 slag+ limestone	0,46 S 360 C 20/25 limestone+ fly ash	0,44 S 360 C 20/25 slag	0,43 S 360 C 20/25 slag+ fly ash	0,43 S 360 C 20/25 slag	0,41 S 360 C 20/25 slag	0,40 S 360 C 20/25 fly ash	0,40 S 360 C 20/25 slag	0,39 S 400 C 30/37 slag	0,37 S 400 C 30/37 slag	0,35 S 400 C 30/37 fly ash
🟨 28 days	6,78	6,44	6,74	6,11	6,44	5,20	5,78	6,34	5,21	4,89	5,73	5,55	4,31	4,30
🟩 180 days	6,00	6,11	6,34	6,01	6,20	5,17	5,16	5,17	4,78	4,20	5,67	5,78	4,11	4,00
🟩 1 year	6,20	6,00	4,76	5,78	6,34	4,18	4,12	4,67	4,26	3,80	4,71	4,20	3,14	3,52

Fig. 4.1.9 – Time evolution of porosity of concretes prepared with superplasticizing admixtures

Air and water permeability

Concrete samples were also tested for permeability, measuring the water infiltration depth at pressures of 4, 8 or 12 bar (permeability class P4, P8 or P12) respectively the air flow depth (air permeability coefficient).

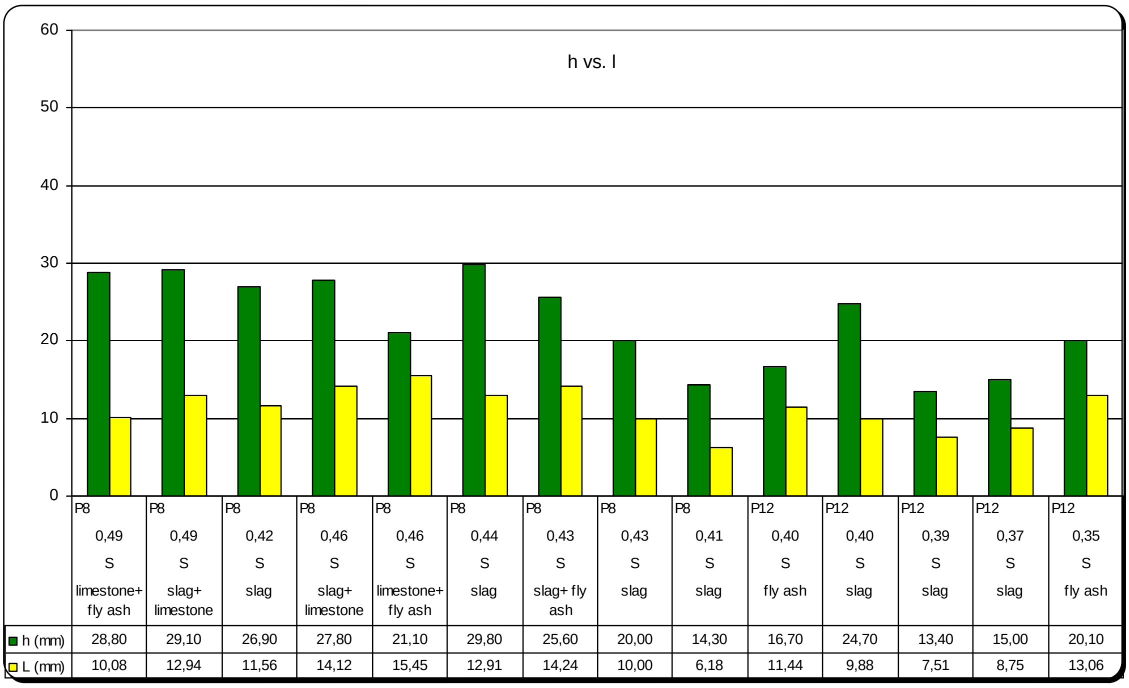
The determination of air permeability was achieved using the Torrent method.

Air flow is unidirectional and perpendicular on the exposed surface, the same way aggressive agents are transported to the inner sections of concrete. Humidity within concrete structure may have a significant influence on the water or air permeability.

The purpose was to carry out a comparative assessment between the air flow depth in concrete, determined by Torrent method (L) and the water infiltration depth (h) at pressures of 4, 8 or 12 bar and correlating such values with radon emissions. The results achieved are displayed in figure 4.1.9 and presented *in extenso* herein.

Following experimental research works performed, the air permeability values were noted to increase with time, so the summary shall present results achieved on 1-year-old concretes. The thesis shall also present the results achieved at 28 and respectively 180 days.

Laboratory research has revealed that concrete permeability reduces with the increase of hydrated cement materials and with the reduction of W/C ratio.



	P8 0,49 S limestone+ fly ash	P8 0,49 S slag+ limestone	P8 0,42 S slag	P8 0,46 S slag+ limestone	P8 0,46 S limestone+ fly ash	P8 0,44 S slag	P8 0,43 S slag+ fly ash	P8 0,43 S slag	P8 0,41 S slag	P12 0,40 S fly ash	P12 0,40 S slag	P12 0,39 S slag	P12 0,37 S slag	P12 0,35 S fly ash
🟩 h (mm)	28,80	29,10	26,90	27,80	21,10	29,80	25,60	20,00	14,30	16,70	24,70	13,40	15,00	20,10
🟨 L (mm)	10,08	12,94	11,56	14,12	15,45	12,91	14,24	10,00	6,18	11,44	9,88	7,51	8,75	13,06

Fig. 4.1.10 – Water infiltration depth (P8 and P12) and air flow depth for concretes prepared with superplasticizing admixture, at the age of 1 year

The air flow depth for concretes prepared with superplasticizing admixture generally account for 40-60% of the air flow depth for 8 and 12 bar.

For concretes prepared with air entrainer, values achieved for the air flow depth account for, in average, 50-55%, the highest value being achieved on concretes with cements added with fly-ash (56%), and the lowest, 50%, being measured on concretes with cements added with slag.

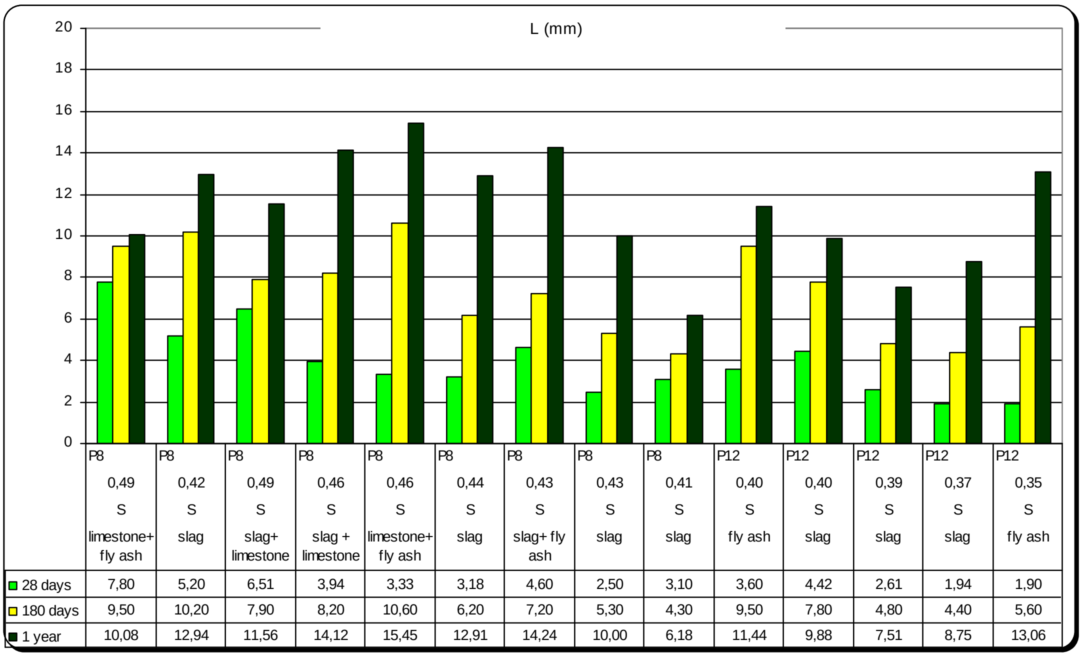
When comparing concretes with similar cement dosages, concretes prepared with cements added with slag display air flow depths lower than other types of concretes surveyed, disregarding the presence / absence of admixtures. Generally, high air flow depths were achieved for concretes prepared with cements added with limestone and fly-ash.

Analyzing the results achieved, the air flow depth L (mm) determined using Torrent method is found to vary based on the cement, admixture type and concrete age.

Water infiltration depth and respectively the air flow depth decrease with the reduction of W / C ratio.

In case of concretes prepared with superplasticizing admixtures and with low water / cement ratios, the air flow depth measured was lower than in case of concretes prepared with high W / C ratios, especially for concretes prepared with cement added slag. The same conclusions may be drawn from admixture-free concretes.

The air flow depth in concretes increases in time, disregarding the type of admixture used.

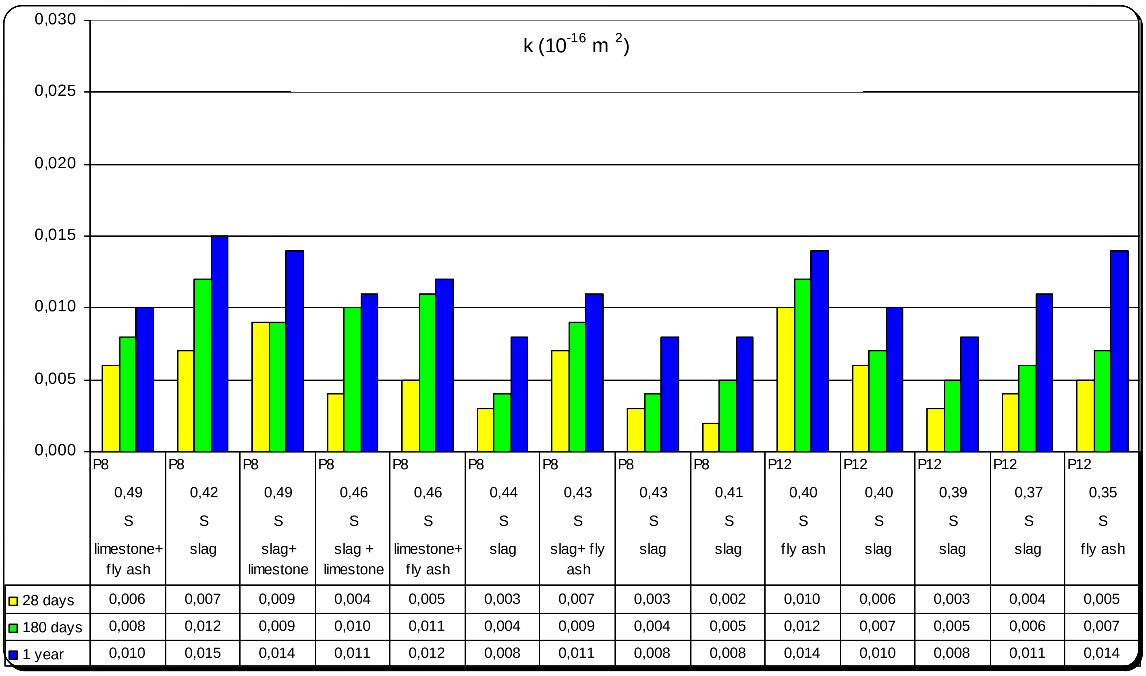


	P8 0,49 S limestone+ fly ash	P8 0,42 S slag	P8 0,49 S slag+ limestone	P8 0,46 S slag + limestone	P8 0,46 S limestone+ fly ash	P8 0,44 S slag	P8 0,43 S slag+ fly ash	P8 0,43 S slag	P8 0,41 S slag	P12 0,40 S fly ash	P12 0,40 S slag	P12 0,39 S slag	P12 0,37 S slag	P12 0,35 S fly ash
🟩 28 days	7,80	5,20	6,51	3,94	3,33	3,18	4,60	2,50	3,10	3,60	4,42	2,61	1,94	1,90
🟨 180 days	9,50	10,20	7,90	8,20	10,60	6,20	7,20	5,30	4,30	9,50	7,80	4,80	4,40	5,60
■ 1 year	10,08	12,94	11,56	14,12	15,45	12,91	14,24	10,00	6,18	11,44	9,88	7,51	8,75	13,06

Fig. 4.1.11 – Variation in time of the air flow depth for concretes prepared with superplasticizing admixtures

Also, the air permeability coefficient was determined for concretes prepared with various cement types, the results achieved being displayed in figure 4.1.12.

The permeability coefficient follows the same trend with the air flow depth, with lower values for concretes with cements added with slag, for the same cement dosage and respectively for a W / C ratio, disregarding the presence / absence of admixtures.



	P8 0,49 S limestone+ fly ash	P8 0,42 S slag	P8 0,49 S slag+ limestone	P8 0,46 S slag + limestone	P8 0,46 S limestone+ fly ash	P8 0,44 S slag	P8 0,43 S slag+ fly ash	P8 0,43 S slag	P8 0,41 S slag	P12 0,40 S fly ash	P12 0,40 S slag	P12 0,39 S slag	P12 0,37 S slag	P12 0,35 S fly ash
🟨 28 days	0,006	0,007	0,009	0,004	0,005	0,003	0,007	0,003	0,002	0,010	0,006	0,003	0,004	0,005
🟩 180 days	0,008	0,012	0,009	0,010	0,011	0,004	0,009	0,004	0,005	0,012	0,007	0,005	0,006	0,007
🟦 1 year	0,010	0,015	0,014	0,011	0,012	0,008	0,011	0,008	0,008	0,014	0,010	0,008	0,011	0,014

Fig. 4.1.12 - Variation in time of the air permeability coefficient for concretes prepared with superplasticizing admixtures

For concretes prepared with superplasticizing admixtures and respectively air entrainers, the values achieved for permeability coefficient are certain grades lower than those achieved for admixture-free concretes, at the same W/C ratio.

The permeability coefficient progresses in time, between 28 days and 1 year the increase varying between 25%-60%, slower evolutions being seen for concretes prepared with cements added slag and respectively for concretes prepared with air entrainers and cements added with fly-ash.

The influence of density and porosity on the air permeability coefficient is shown below.

The air permeability coefficient for concretes prepared with the same cement type increases with the concrete's porosity.

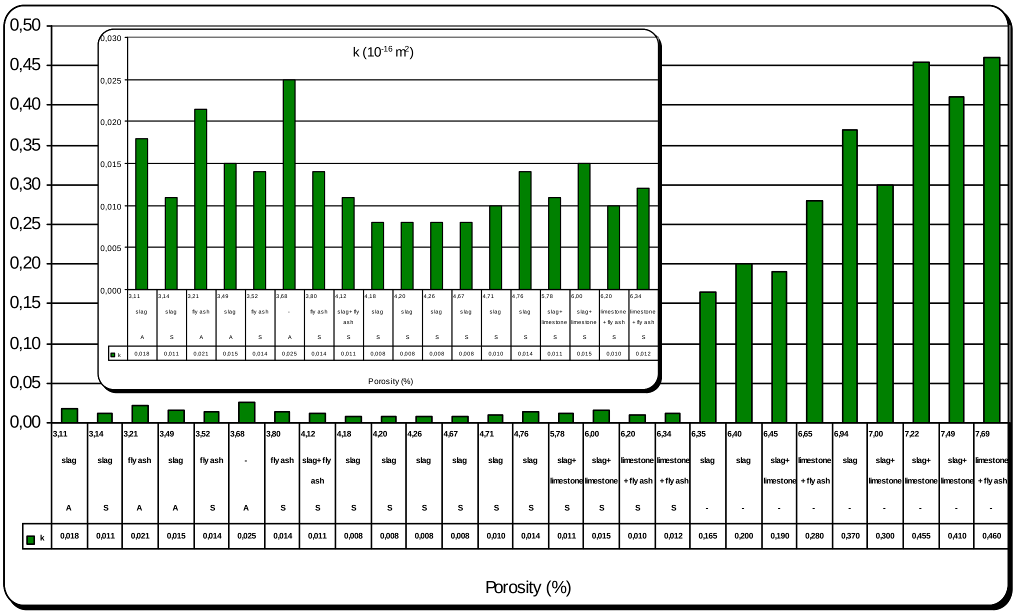


Fig. 4.1.13 - Variation of air permeability coefficient based on concrete's porosity, at the age of 1 year

At the age of 1 year, values achieved for the permeability coefficient of concretes prepared with admixtures are 15-20 times lower than values achieved for admixture-free concretes, at the same W/C ratio. This case also illustrates the role of admixtures used in concrete preparation.

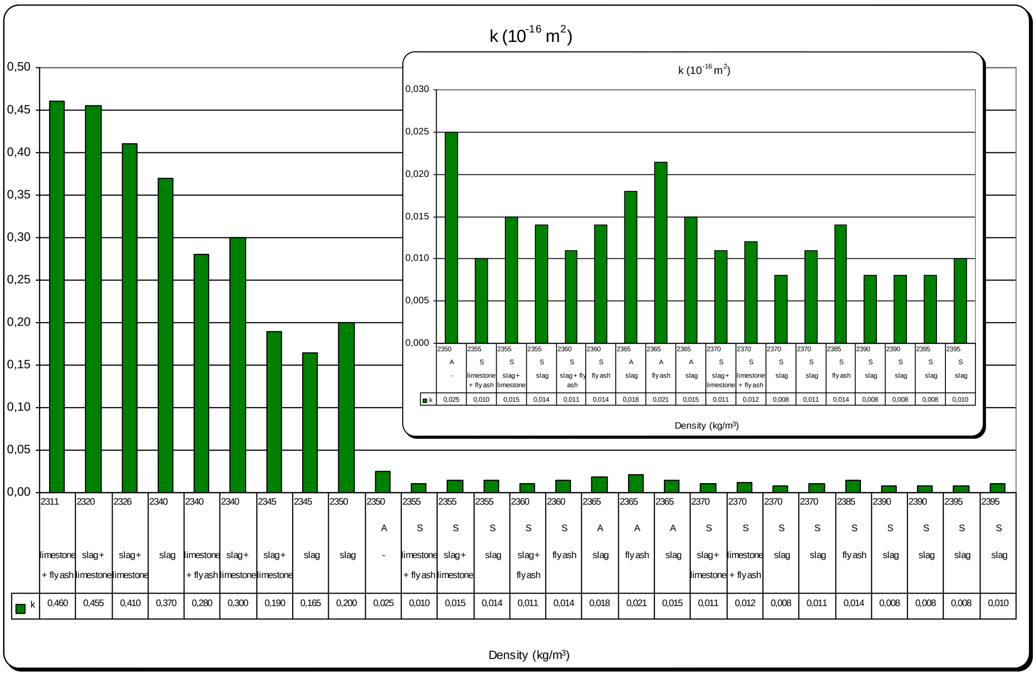


Fig. 4.1.14 - Variation of air permeability coefficient based on concrete's density, at the age of 1 year

For concretes prepared with the same cement type, the air permeability coefficient decreases with the increase of concrete's density.

For a given compressive strength, the lowest permeability coefficient values were achieved in concretes prepared with cement added with slag.

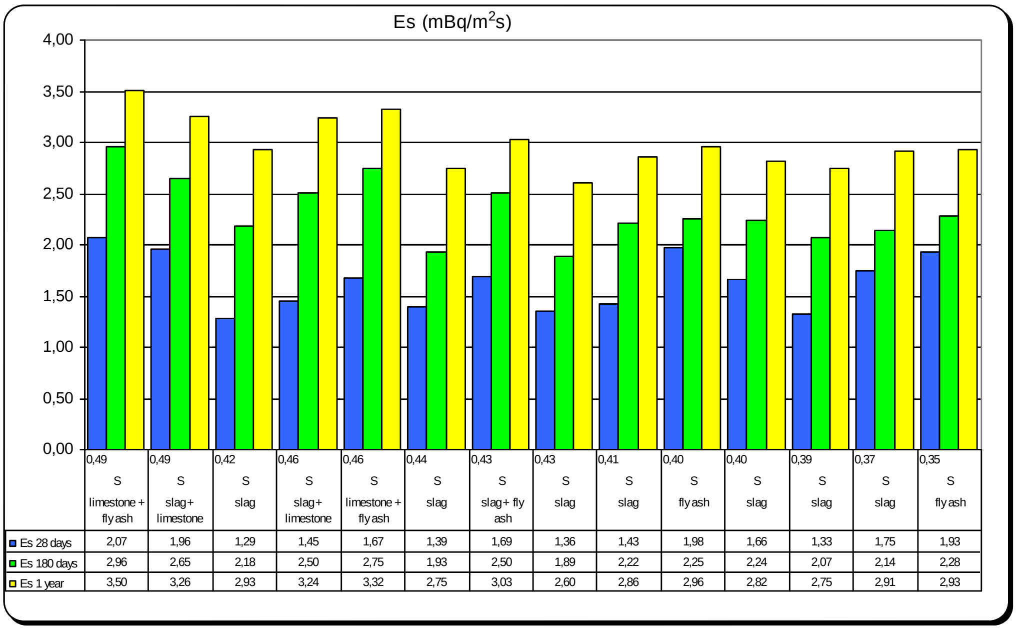
The air permeability coefficient decreases with the increase of compressive strength, higher coefficient values being achieved in admixture-free concretes.

Out of all concretes assessed, concretes prepared with cement added with slag display a denser structure, are less permeable and less porous than other concrete types, this fact being proved by results achieved for water infiltration depth, air flow depth and respectively for the air permeability coefficient.

The strength and durability features illustrated confirm the quality of concretes surveyed, especially of concretes prepared with superplasticizing admixture and respectively air entrainer.

4.1.3. Results achieved for the radon exhalation rate prepared with various cement types

The concrete samples prepared with various cement types were also tested for radon exhalation rate at 28 days, 180 days and 1 year. Results achieved for the radon exhalation rate on the concrete surface unit are displayed in figure 4.1.15. I state that for exhalation measurement the radon assessment system Pylon AB-5 was used, comprising radon monitor type Pylon AB-5 together with a special box used to deposit the concrete samples.



	0,49 S limestone + fly ash	0,49 S slag+ limestone	0,42 S slag	0,46 S slag+ limestone	0,46 S limestone + fly ash	0,44 S slag	0,43 S slag+ fly ash	0,43 S slag	0,41 S slag	0,40 S fly ash	0,40 S slag	0,39 S slag	0,37 S slag	0,35 S fly ash
🟦 Es 28 days	2,07	1,96	1,29	1,45	1,67	1,39	1,69	1,36	1,43	1,98	1,66	1,33	1,75	1,93
🟩 Es 180 days	2,96	2,65	2,18	2,50	2,75	1,93	2,50	1,89	2,22	2,25	2,24	2,07	2,14	2,28
🟨 Es 1 year	3,50	3,26	2,93	3,24	3,32	2,75	3,03	2,60	2,86	2,96	2,82	2,75	2,91	2,93

Fig. 4.1.15 – Variation in time of radon exhalation rate per surface unit, for concretes prepared with superplasticizing admixture

The exhalation rate values increase in time, as the values achieved at 28 days after pouring range between (1,29-2,44) mBq/m^{2}s, and 1 year after pouring the same values range between (2,33-3,79) mBq/m^{2}s.

Radon exhalation rate per concrete mass unit are displayed in figure 4.1.6

The exhalation rate values Em vary between (7-12,5) mBq/kgh for concretes at 28 days, (11-16,5) mBq/kgh for concretes at 180 days and (14,5-20,5) mBq/kgh for concretes at 1 year, the values in the upper section of such intervals being achieved for admixture-free concretes.

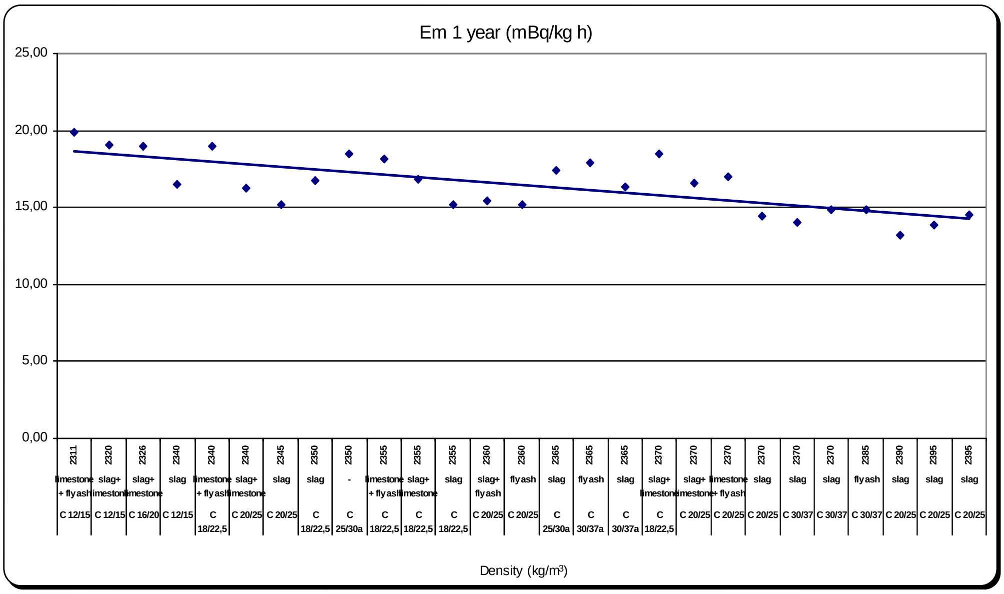
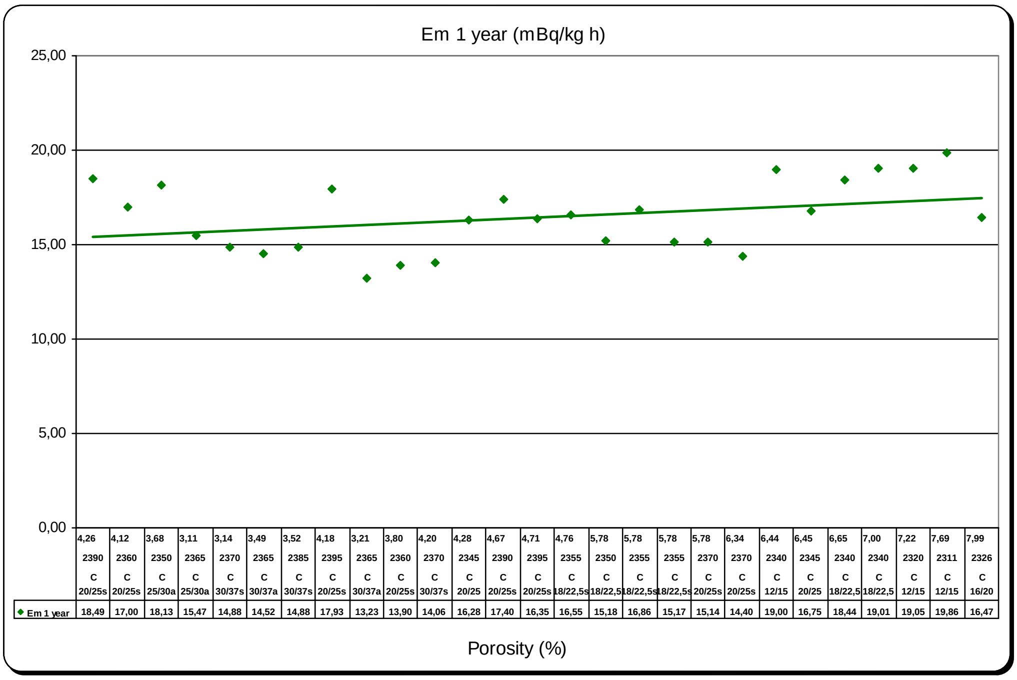


Fig. 4.1.16 – Variation of radon exhalation rate value per mass unit, at 1 year, based on concrete density

The radon exhalation rate value per mass unit is noted to decrease with the increase of concrete density, disregarding concrete's age.

The variation of radon exhalation rate value per mass unit based on concrete porosity at various ages is displayed in figure 4.1.17.



	4,26	4,12	3,68	3,11	3,14	3,49	3,52	4,18	3,21	3,80	4,20	4,28	4,67	4,71
	2390	2360	2350	2365	2370	2365	2385	2395	2365	2360	2370	2345	2390	2395
	C 20/25s	C 20/25s	C 25/30a	C 25/30a	C 30/37s	C 30/37a	C 30/37s	C 20/25s	C 30/37a	C 20/25s	C 30/37s	C 20/25	C 20/25s	C 20/25s
◆ Em 1 year	18,49	17,00	18,13	15,47	14,88	14,52	14,88	17,93	13,23	13,90	14,06	16,28	17,40	16,35

	4,76	5,78	5,78	5,78	5,78	6,34	6,44	6,45	6,65	7,00	7,22	7,69	7,99
	2355	2350	2355	2355	2370	2370	2340	2345	2340	2340	2320	2311	2326
	C 18/22,5s	C 18/22,5	C 18/22,5s	C 18/22,5s	C 20/25s	C 20/25s	C 12/15	C 20/25	C 18/22,5	C 18/22,5	C 12/15	C 12/15	C 16/20
◆ Em 1 year	16,55	15,18	16,86	15,17	15,14	14,40	19,00	16,75	18,44	19,01	19,05	19,86	16,47

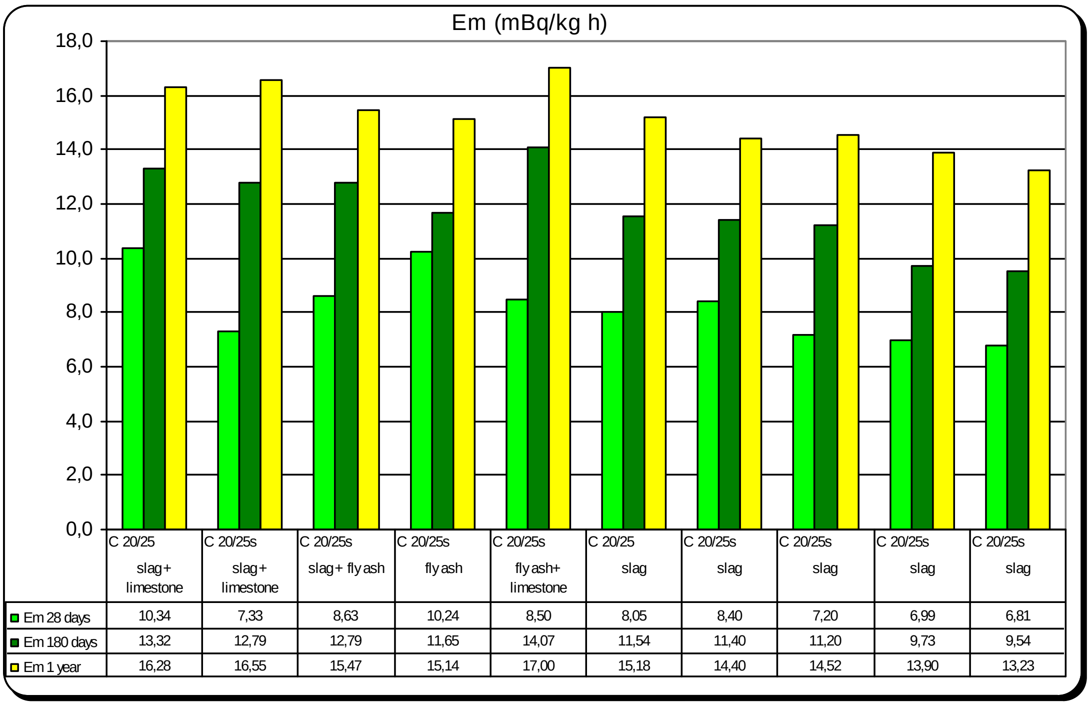
Fig. 4.1.17 - Variation of radon exhalation rate value per mass unit, at 1 year, based on concrete porosity

The radon exhalation rate value per mass unit increase with the increase of concrete density, disregarding concrete's age.

Concrete porosity decreases in time, more significantly for concretes prepared with cement added with slag and respectively fly-ash.

High porosity values were achieved, especially for admixture-free concretes.

For the same concrete class, the variation of exhalation rate values Em based on concrete composition is shown in figure 4.1.18.



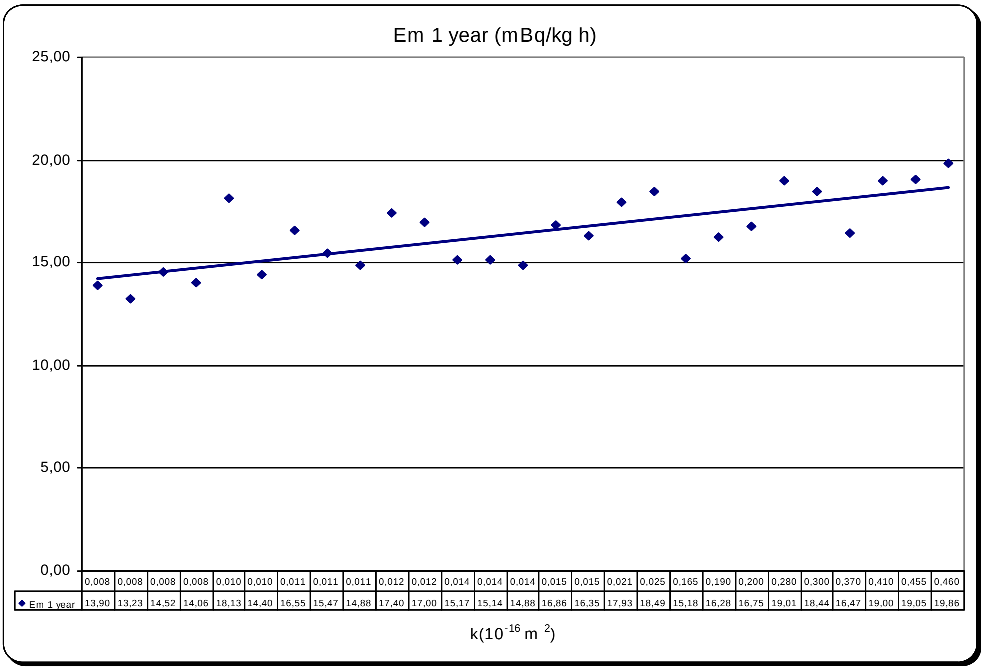
	C 20/25 slag+ limestone	C 20/25s slag+ limestone	C 20/25s slag+ fly ash	C 20/25s fly ash	C 20/25s fly ash+ limestone	C 20/25 slag	C 20/25s slag	C 20/25s slag	C 20/25s slag	C 20/25s slag
🟩 Em 28 days	10,34	7,33	8,63	10,24	8,50	8,05	8,40	7,20	6,99	6,81
🟩 Em 180 days	13,32	12,79	12,79	11,65	14,07	11,54	11,40	11,20	9,73	9,54
🟨 Em 1 year	16,28	16,55	15,47	15,14	17,00	15,18	14,40	14,52	13,90	13,23

Fig. 4.1.18 - Variation of radon exhalation rate values for concretes in class C20/25, prepared with and without superplasticizing admixture (S)

For the same concrete class, the radon exhalation rate values of concretes prepared with cement added with slag are lower than those of concretes prepared with other cement types, disregarding the admixture used.

The radon exhalation rate values Em range between (7-12,5) mBq/kgh for concretes at 28 days, (11-16,5) mBq/kgh for concretes at 180 days and (14,5-20,5) mBq/kgh for concretes at 1 year, the values in the upper section of such intervals being achieved for admixture-free concretes.

The variation of radon exhalation rate value per mass unit based on air permeability coefficient of concrete at 1 year after pouring is shown in figure 4.1.19.



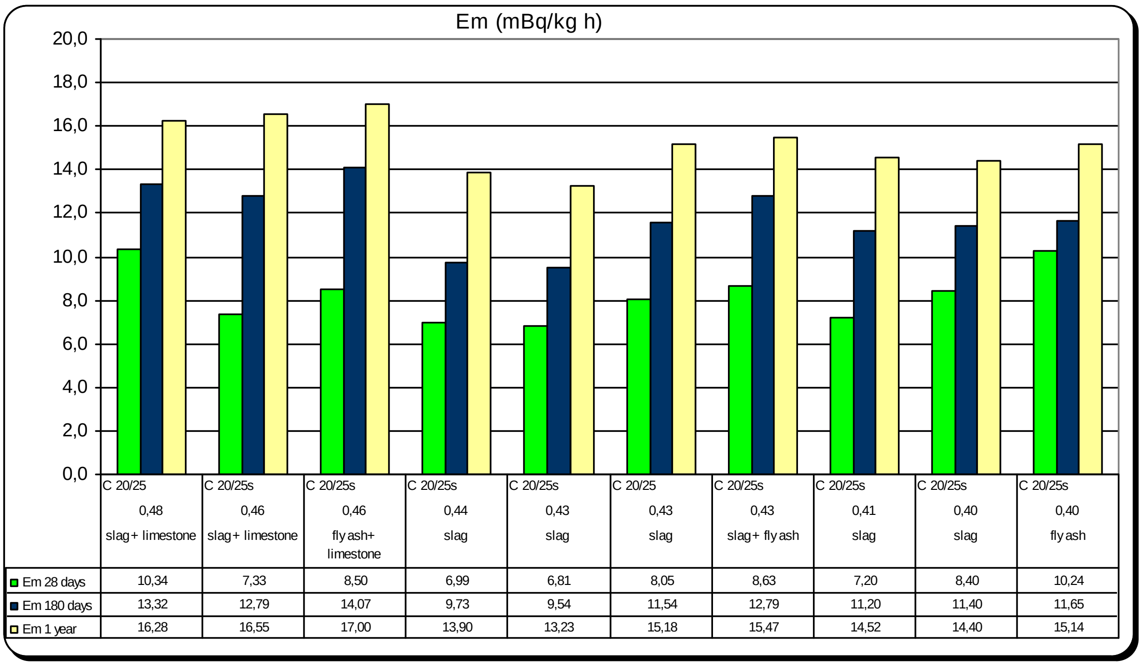
	0,008	0,008	0,008	0,008	0,010	0,010	0,011	0,011	0,011	0,012	0,012	0,014	0,014	0,014	0,015	0,015	0,021	0,025	0,165	0,190	0,200	0,280	0,300	0,370	0,410	0,455	0,460
◆ Em 1 year	13,90	13,23	14,52	14,06	18,13	14,40	16,55	15,47	14,88	17,40	17,00	15,17	15,14	14,88	16,86	16,35	17,93	18,49	15,18	16,28	16,75	19,01	18,44	16,47	19,00	19,05	19,86

Fig. 4.1.19 - Variation of radon exhalation rate value per mass unit,
at 1 year, based on air permeability coefficient

The radon exhalation rate value per mass unit increase with the air permeability coefficient, disregarding concrete’s age.

The radon exhalation rate per mass unit shows a linear variation with the air permeability coefficient, achieving high Em values for high values of the air permeability coefficient, consequently for concretes with low density and high porosity.

Based on the W/C ratio, concretes display different exhalation rates, as they are generally higher for high W/C ratios.



	C 20/25 0,48 slag+ limestone	C 20/25s 0,46 slag+ limestone	C 20/25s 0,46 fly ash+ limestone	C 20/25s 0,44 slag	C 20/25s 0,43 slag	C 20/25 0,43 slag	C 20/25s 0,43 slag+ fly ash	C 20/25s 0,41 slag	C 20/25s 0,40 slag	C 20/25s 0,40 fly ash
🟩 Em 28 days	10,34	7,33	8,50	6,99	6,81	8,05	8,63	7,20	8,40	10,24
🟦 Em 180 days	13,32	12,79	14,07	9,73	9,54	11,54	12,79	11,20	11,40	11,65
🟨 Em 1 year	16,28	16,55	17,00	13,90	13,23	15,18	15,47	14,52	14,40	15,14

Fig. 4.1.20 - Variation of radon exhalation rate value per mass unit based on W/C ratio, for concrete class C20/25

The findings of this research program are synthesized in charts displayed in figures 4.1.21 - 4.1.23.

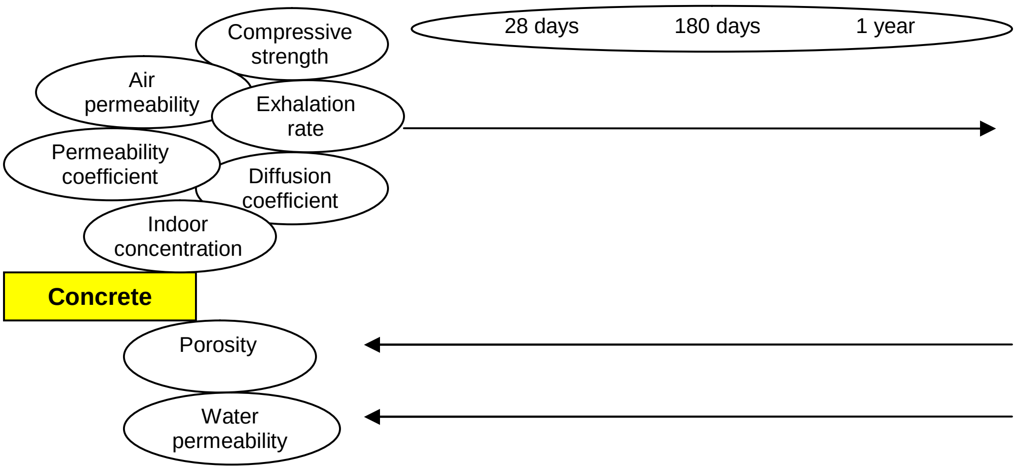


Fig.4.1.21 – Variation in time of features of concretes prepared with various cement types

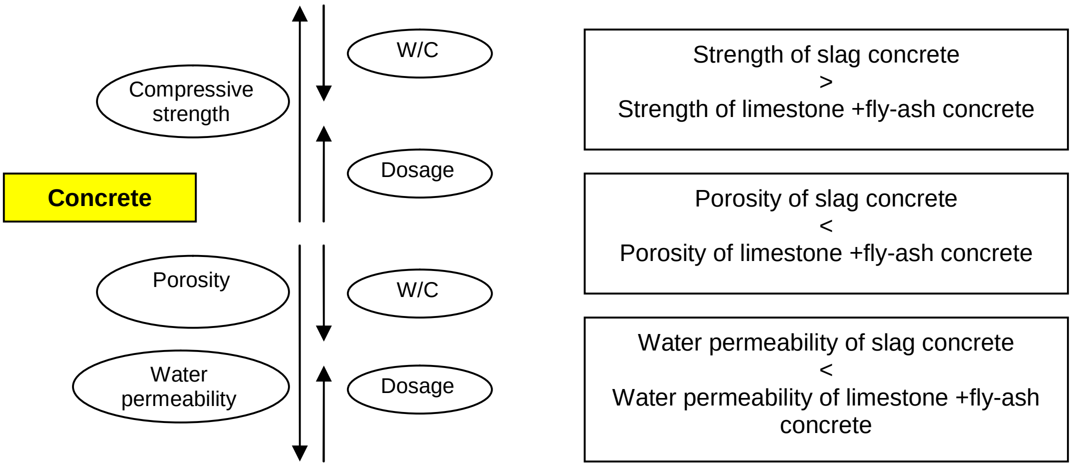


Fig.4.1.22 – Influence of W/C ratio, dement dosage and additions on the features of concretes prepared with various cement types

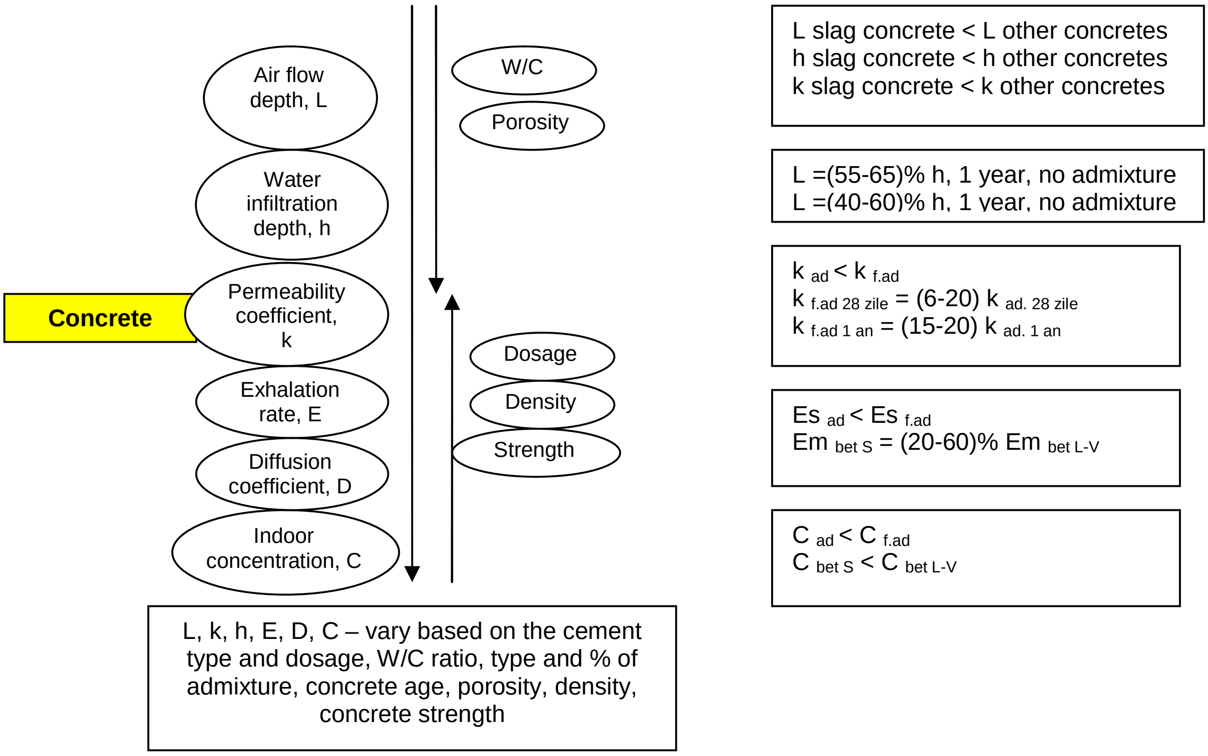


Fig. 4.1.23 – Chart of results achieved when cross-referencing concrete features with radon exhalation rate and indoor concentration

4.2. Influence of concrete features on radon transport

Radon may be transported due to concentration gaps (diffusive transport). The diffusive transport is considered to be the main process causing radon exhalation from building materials and also it is considered to play a major role in radon transport from soil to building's basement.

In order to achieve a fair description of radon transport through concrete certain information is needed on the concrete structure, porosity and permeability, on processes causing the transport of radon, on radon's interaction with environment, factors favoring the generation of radon, etc.

The concrete features (that depend on its composition and especially on the W/C ratio) the influence the transport of radon through concrete are, mainly, porosity, permeability, diffusion, humidity and density.

For concretes cured at controlated temperature and humidity conditions, the overall concrete porosity (Rogers, [111]) may be calculated with the formula:

$P_t = 1 - d/G$

where:
d = concrete density, (g/cm^3)
G = solid density (2,6x10^{3}kg/m^3).

4.2.1. Influence of W/C ratio and concrete density on apparent and overall porosity of concrete

Using the results achieved experimentally as per section 4.1.2, the apparent and overall porosity values are shown below. The concrete samples used in these experiments were prepared as per the norms in force upon research date. Remember that samples were, after pouring and unmolding, cured for seven days in water and then, until the test date (28 days and respectively 1 year) in air at 20 ^{0}C temperature and 65% humidity.

The overall porosity $P_t = P_{ap} + P_2$, where P_2 is the volume of closed pores. The overall porosity values are 0,01-0,04 higher than apparent porosity values, P_2 = 0,01 - 0,04. The volume of closed spores increases with the reduction of W/C ratio.

Figure 4.2.1 displays the values achieved for concrete's overall porosity using the Rogers formula and the values achieved experimentally for apparent porosity, 1 year after concrete pouring.

The volume of pores closed at one year after pouring is higher than the volume achieved at 28 days, P_2 = 0,03-0,05, for W/C>0,39 and respectively P_2 = 0,06, for A/C<0,37.

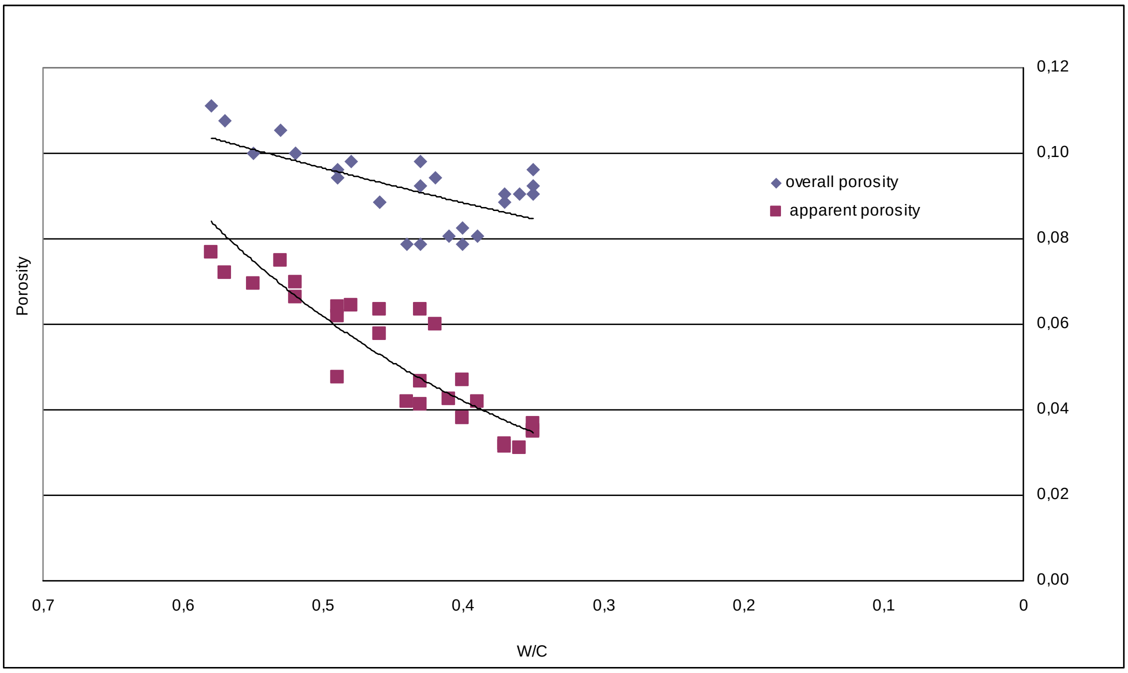


Fig. 4.2.1 – The values achieved for the overall porosity of concrete and values achieved experimentally for the apparent porosity, at the age of one year

The overall porosity values decrease slower with the reduction of W/C ratio than those of concrete apparent porosity. The ratio between the concrete apparent porosity determined experimentally and the overall porosity decreases with the reduction of the W/C ratio.

4.2.2. Influence of W/C ratio and of concrete density on the permeability coefficient

Using the results achieved in section 4.1.2, a comparison shall be made below between values acquired experimentally and values achieved by calculation for the permeability coefficient using values determined experimentally for the concrete density and W/C ratio.

The permeability coefficient at 28 days and respectively 1 year was calculated with the Rogers formula [109] using values determined experimentally for the concrete density:

$k= 0,22 \exp(-12,4\ d)$

where d = concrete density, (g/cm^3).

Compared to concretes prepared with blended cements, those prepared with addition-free cements show higher permeability coefficient values.

At 1 year after pouring, admixture-free concretes show much higher permeability coefficient values than calculated, disregarding the W/C ratio and the cement type use.

In case of concretes with admixtures, values determined experimentally for the permeability coefficient are lower than theoretical values, disregarding the W/C ratio, the admixture type and respectively the cement type used in the preparation of concretes.

Figure 4.2.2 shows the variation of permeability coefficient, values determined experimentally vs. values determined theoretically based on concrete density determined at 28 days.

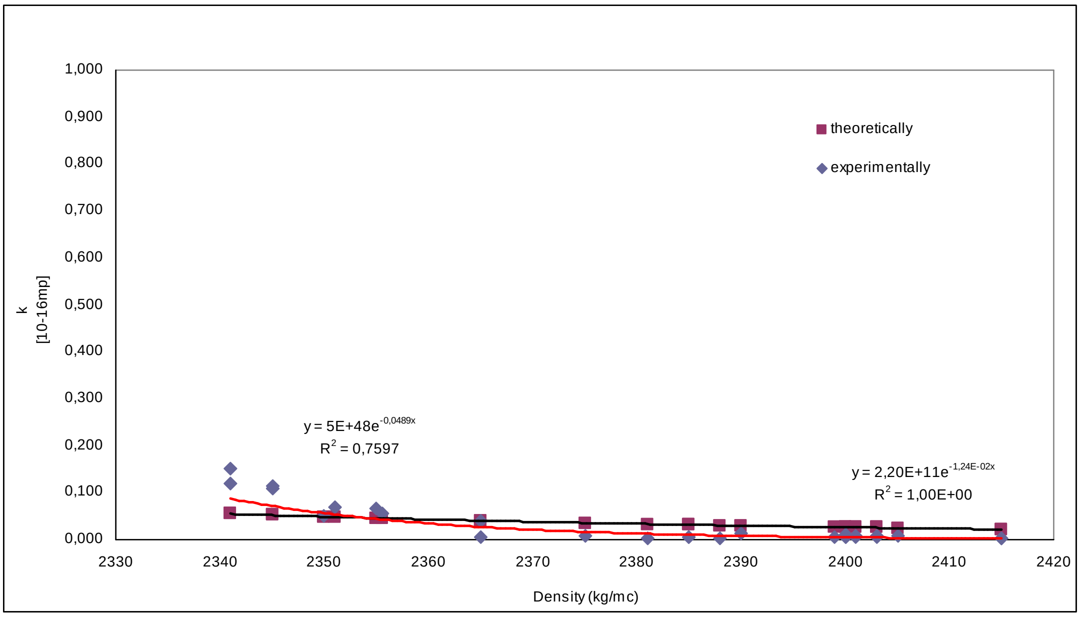


Fig. 4.2.2 – Permeability coefficient values, determined experimentally and respectively theoretically, based on concrete density measured at 28 days

For concretes at 28 days, values achieved experimentally for the permeability coefficient are similar to those determined theoretically.

Figure 4.2.3 shows the variation of permeability coefficient, values determined experimentally vs. values determined theoretically based on concrete density measured at 1 year.

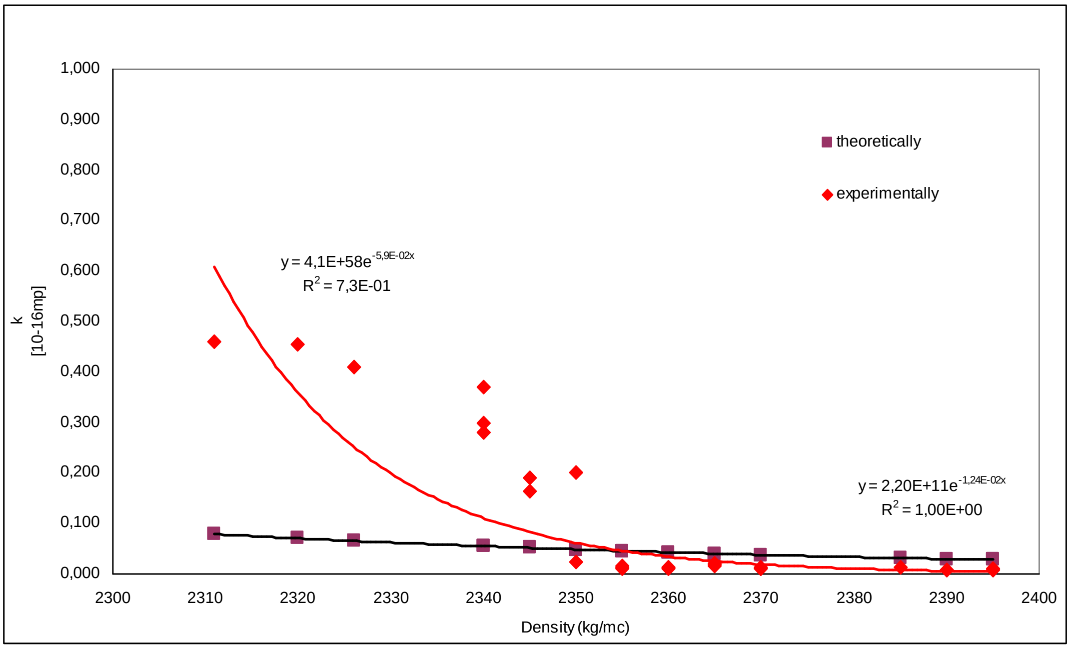


Fig. 4.2.3 - Permeability coefficient values, determined experimentally and respectively theoretically, based on concrete density measured at 1 year

For concretes with densities exceeding 2350 kg/m^3 (values achieved for concretes prepared with admixtures), values determined experimentally for permeability coefficient are similar to those achieved theoretically. Thus, Rogers formula is verified for concretes with densities exceeding 2350 kg/m^3.

Figures 4.2.4 and 4.2.5 shows the variation of permeability coefficient, values determined experimentally vs. values determined theoretically based on the W/C ratio used in preparation of concretes.

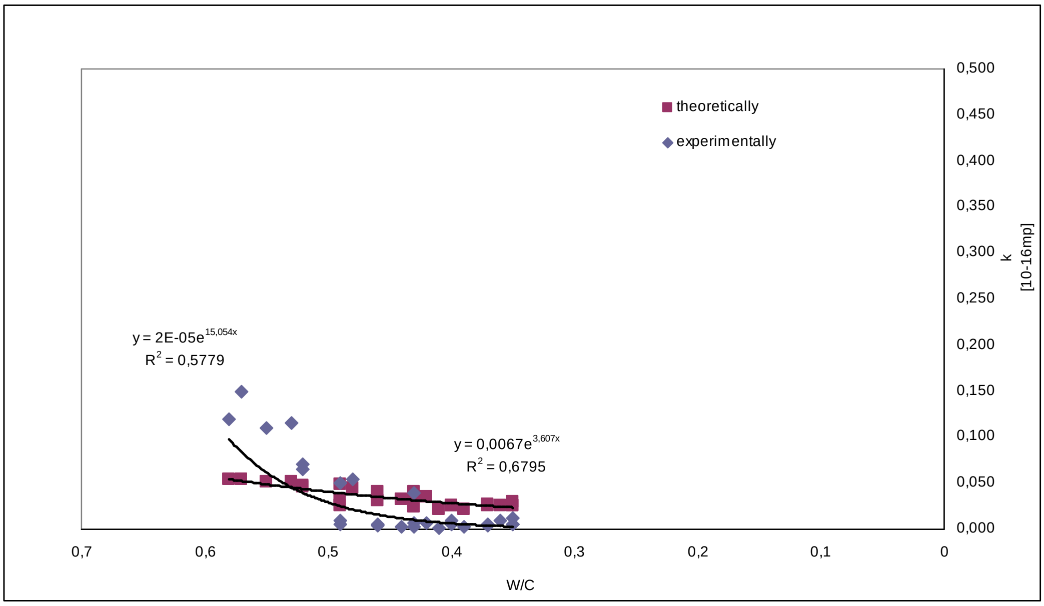


Fig. 4.2.4 - Permeability coefficient values, determined experimentally and respectively theoretically, based on the W/C ratio, 28 after sample pouring

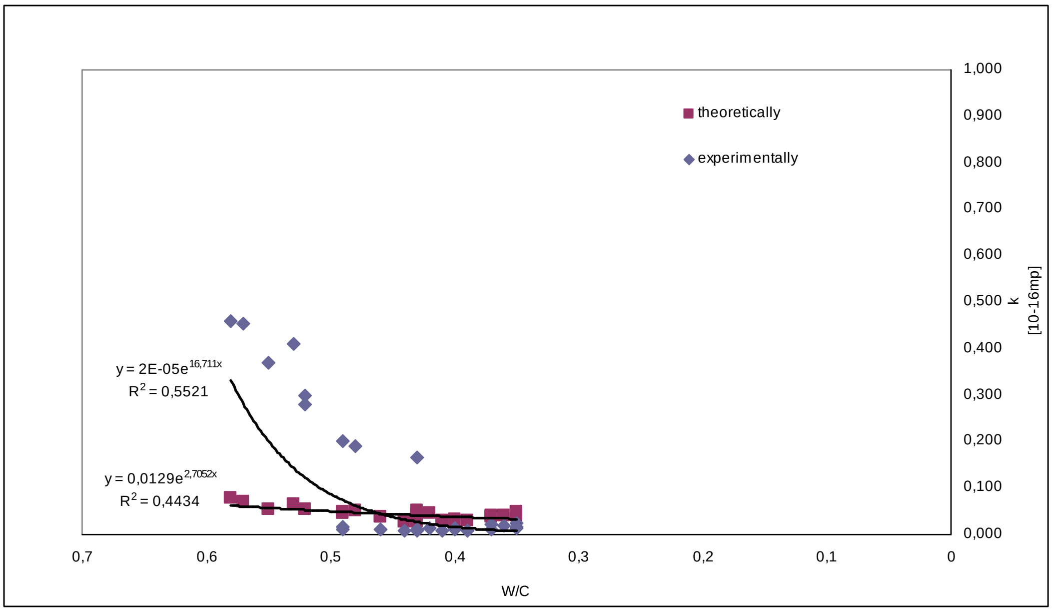


Fig. 4.2.5 - Permeability coefficient values, determined experimentally and respectively theoretically, based on the W/C ratio, 1 year after sample pouring

For concretes prepared with admixtures, at 1 year and W/C ratios lower than 0,45, the values achieved experimentally for the permeability coefficient are similar to those calculated with Rogers formula.

The permeability coefficient decreases with the increase of cement dosage and implicitly with the reduction of W/C ratio.

4.2.3. Influence of W/C ratio and concrete density on the diffusion coefficient

4.2.3.1. Influence of W/C ratio on the diffusion coefficient

The dependence of diffusion coefficient on the W/C ratio was determined by various authors. E.g., Rogers [109] found a correlation between the two parameters (RAETRAD model), namely:

$D = 1{,}5\ x10^{-10}\ \exp\ (11{,}4\ A/C)$

For W/C ratios ranging between 0,52 and 0,67, Rogers determined values of the diffusion coefficient varying between 1,8 $x10^{-8}$ and 4,6 $x10^{-7}$ m^2/s. Nielson and Rogers [111] found a relation between the diffusion coefficient and the W/C ratio, namely:

$D = 7{,}7\ x10^{-10}\ \exp\ (8{,}7\ A/C)$.

Figure 4.2.6 displays calculated values, using the above formulas, of radon diffusion coefficient for W/C ratios achieved for concretes prepared with various cement types.

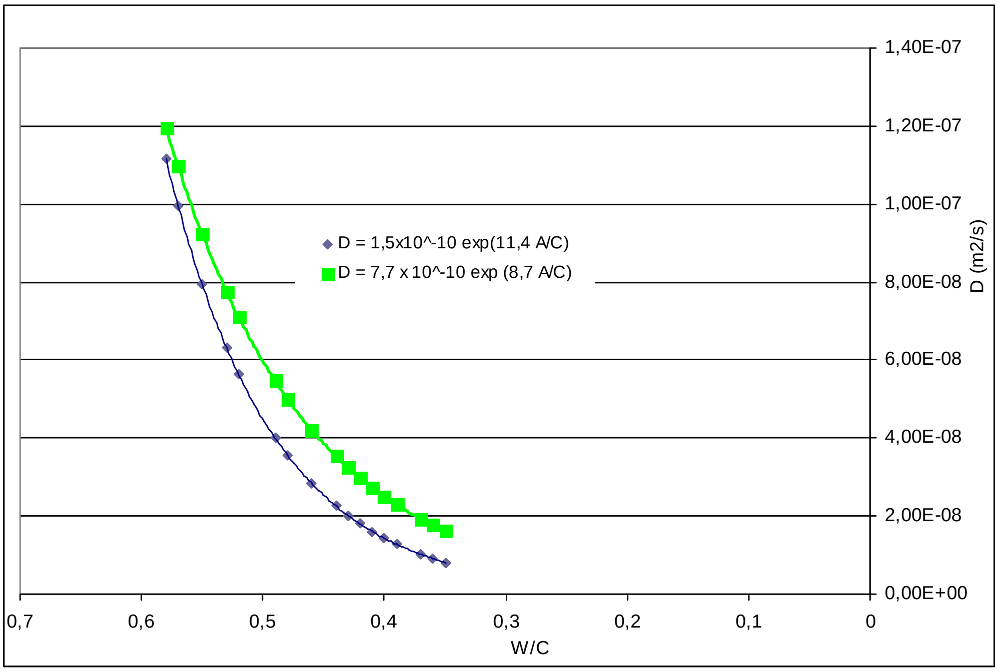


Fig. 4.2.6 – Variation of diffusion coefficient based on W/C ratio

The diffusion coefficient decreases with the increase of cement dosage and implicitly with the reduction of W/C ratio.

For higher W/C ratios, results achieved using the two correlation curves are close. Values determined using Rogers correlation formula are lower than those determined based on the Nielson & Rogers formula.

4.2.3.2. Influence of concrete density on the diffusion coefficient

The dependence of diffusion coefficient on the concrete density is much more useful as concrete density concrete density is much more easier to determine for placed concretes. Nielson and Rogers [111] found a dependence relation:

$D = 0{,}084\ \exp\ (-0{,}0064d)$.

Figure 4.2.7 displays the values of radon diffusion coefficients in concretes prepared with various cement types, based on their density measured at 1 year. The radon diffusion coefficient decreases with the increase of concrete density.

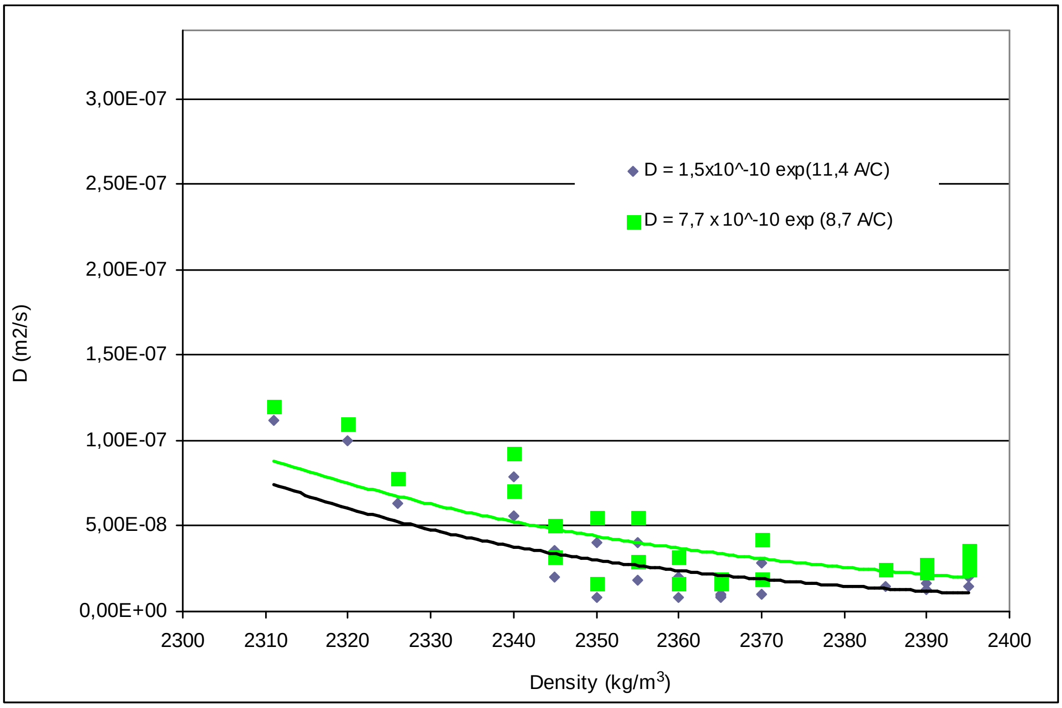


Fig.4.2.7- Variation of diffusion coefficient based on concrete density, at 1 an

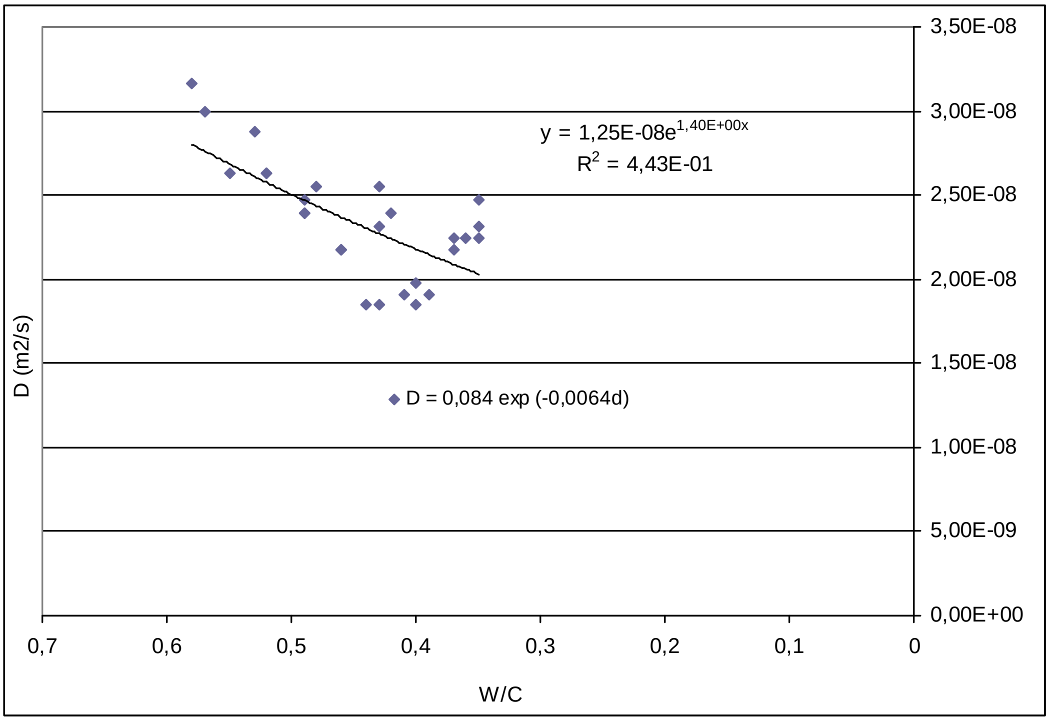


Fig.4.2.8 – Variation of diffusion coefficient based on W/C ratio, at 1 year

The diffusion coefficient decreases with the reduction of W/C ratio, disregarding the concrete age.

4.2.4. Influence of concrete permeability on the diffusion coefficient

Figure 4.2.9 shows the variations of diffusion coefficient based on the values achieved for permeability coefficient measured at 1 year.

The diffusion coefficient increases linearly with the permeability coefficient, disregarding the concrete age.

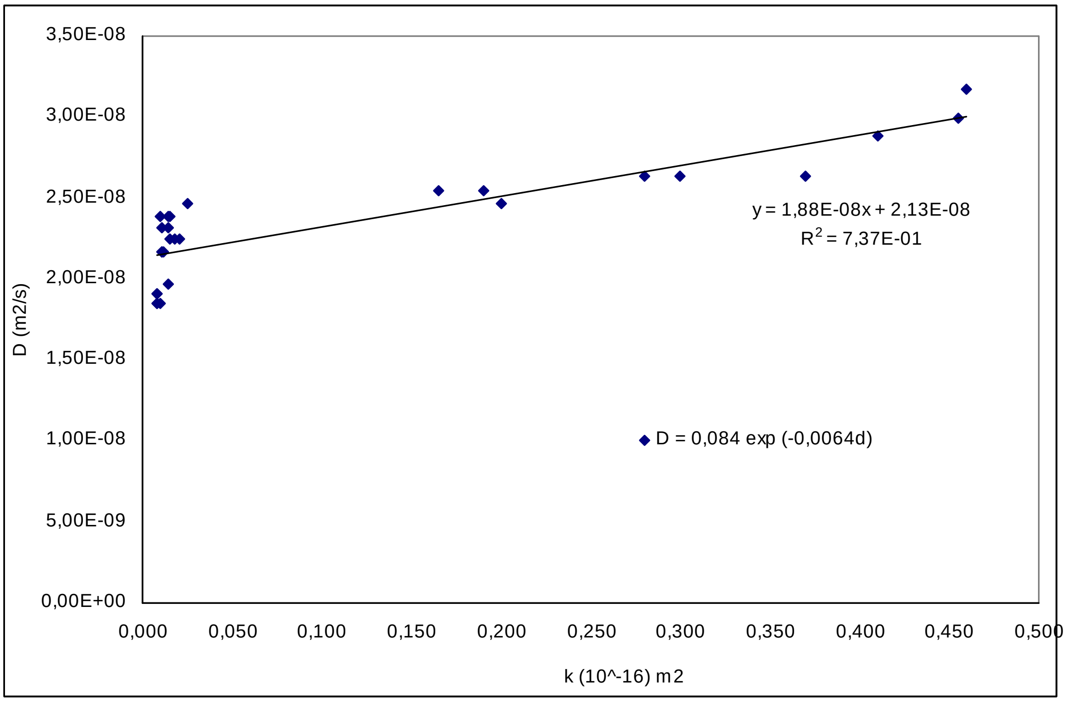


Fig. 4.2.9 – Variations of diffusion coefficient based on the values achieved for permeability coefficient measured at 1 year

The radon diffusion coefficients through concrete vary between 1,62x10^{-8} and 1,1x10^{-7} m^2/s (data achieved using the Rogers formula) or, based on the data acquired using Nielson & Rogers formula (1991), between 8 x10^{-9} and 1,1x10^{-7} m^2/s.

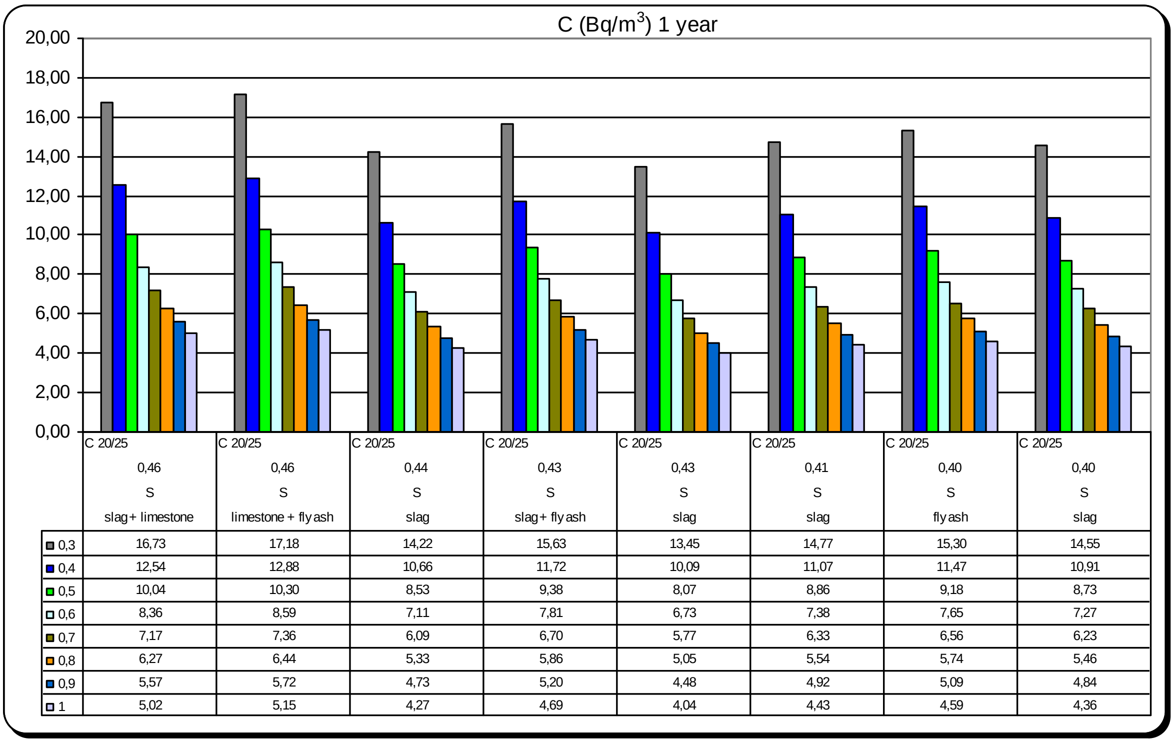
At 28 days, admixture-free concretes, with W/C ratios ranging between 0,5 and 0,4, show permeability coefficients determined experimentally that are similar to those obtained theoretically. In case of admixture-free concretes, with W/C ratios higher than 0,5, the experimentally determined values for the permeability coefficient are higher than theoretical values. Concretes prepared with admixtures have experimentally determined permeability coefficients lower than the coefficients calculated using Rogers formula, disregarding the W/C ratio or the cement type used in the preparation thereof.

5. CONTRIBUTION OF CONCRETES TO THE INDOOR RADON CONCENTRATION

5.1. Determination of the indoor radon concentration, using the radon exhalation rate in concretes prepared with various cement types

Using the results achieved in measurements of radon exhalation rates in concrete, presented in section 5.2 herein, the radon concentrations within the template room were calculated, being displayed as example in figure 5.1.1 for concretes at 1 year. We note that results achieved for the radon concentration in indoor air, due to exhalation at 28 days and respectively 180 days after concrete pouring of concretes are presented *in extensor* herein.

When calculating the indoor radon concentration, the formula C = E x V / λ_v was used, where λ_v is the ventilation rate and V is building's volume.



	C 20/25 0,46 S slag+ limestone	C 20/25 0,46 S limestone + fly ash	C 20/25 0,44 S slag	C 20/25 0,43 S slag+ fly ash	C 20/25 0,43 S slag	C 20/25 0,41 S slag	C 20/25 0,40 S fly ash	C 20/25 0,40 S slag
■ 0,3	16,73	17,18	14,22	15,63	13,45	14,77	15,30	14,55
■ 0,4	12,54	12,88	10,66	11,72	10,09	11,07	11,47	10,91
■ 0,5	10,04	10,30	8,53	9,38	8,07	8,86	9,18	8,73
□ 0,6	8,36	8,59	7,11	7,81	6,73	7,38	7,65	7,27
■ 0,7	7,17	7,36	6,09	6,70	5,77	6,33	6,56	6,23
■ 0,8	6,27	6,44	5,33	5,86	5,05	5,54	5,74	5,46
■ 0,9	5,57	5,72	4,73	5,20	4,48	4,92	5,09	4,84
□ 1	5,02	5,15	4,27	4,69	4,04	4,43	4,59	4,36

Fig. 5.1.1 – Evolution of the indoor radon concentration based on the ventilation rate, walls made of concrete in class C20/25 with superplasticizing admixture

We note that, in case of a room made of concrete prepared with cement with limestone and fly-ash additions, radon concentrations are higher than for other concretes prepared with cement with slag and limestone and respectively slag additions, disregarding the age of concretes assessed.

For a room made of concrete prepared with cement with slag additions and superplasticizing additions, after the age of one year after pouring, the lowest radon concentration was achieved, approximately 14 Bq/m^3, at a room ventilation rate of 0,3 h^{-1} .

The maximum value achieved for the indoor radon concentration is of 20 Bq/m^3, value achieved for a room made of admixture-free concrete prepared with cement with limestone and fly-ash additions, at 1 year after pouring, at a room ventilation rate of 0,3 h^{-1}.

If there are no means of infiltration for radon in soil and in case the indoor air has a concentration of 8 Bq/m^3 (which is a regular value for outdoor), the building may determine an increase of the radon concentration of up to 28 Bq/m^3 .

The values achieved for the indoor radon concentrations for the concretes assessed are not significant.

6. MITIGATION MEASURES FOR THE INDOOR RADON CONCENTRATION

6.1. Determinations made on various types of membranes with resistance against radon infiltration

In order to test the efficiency of various barriers against radon implemented in concretes, concrete cubes were cast with the edge of 100 mm, their characteristics being displayed in table 6.2.1. Remember that these determinations were made by the author during the research-innovation program AMTRANS funded by the Ministry of Research and Education [3].

Table 6.1.1 – Features of concretes A and B

A: Concrete prepared with cement II/A-S 32,5R (slag addition)	
B: Concrete prepared with cement II/B-M (S-V) 32,5R (slag and limestone addition)	
Cement dosage	420 Kg/m^3
W/C ratio	0,5
Water	210 liters/m^3
Aggregate: sort 0-3 mm sort 3-7 mm sort 7-16 mm	650 Kg/m^3 425 Kg/m^3 610 Kg/m^3

Samples were submerged in water for 7 days. After the age of 28 days, the features of concretes were determined, the result achieved being presented in table 6.1.2.

Table 6.1.2 – Features of concretes

Sample	Air permeability, k ($10^{-16}m^2$)	Air flow depth, L (mm)	Water infiltration depth at 6 bar, h (mm)
A	0,031	12,20	21,00
	0,044	14,70	9,00
	0,013	7,80	15,00
Average	**0,029**	**11,57**	**15,00**
B	0,071	18,60	15,00
	0,082	19,90	23,00
	0,017	9,10	28,00
Average	**0,056**	**15,87**	**22,00**

Two cubes of each series were covered with two types of barriers. The surface of concretes were not specially treated prior the application of barriers, the application of such barriers and curing being achieved at room temperature.

All tests were carried out in similar humidity and temperature conditions. Pylon AB 5 system was used.

The two barriers used in this experiment are:

1) Epoxy product T – bi-component based on epoxy resins, used as basic layer (primer) and / or intermediate / final protection layer;

2) Acrylstyrene paint N – mono-component anti-corrosion product based on polymers in aqueous dispersion, used as protection layer and finishing for concrete, brick, ceramic, surface.

Barriers were applied in two layers, on each cube side, as per manufacturer specifications.

Considering that the structure of concrete in cubes presents irregularities, we take as reference the radon exhalation rate from an uncovered concrete cube. Exhalation rates were measured for the uncovered cubes (E_o) and for the cubes covered with the two barriers (E), the results being displayed in table 6.1.3.

Table 6.1.3 – Radon exhalation rate, sample A

Concrete sample A	Exhalation rate (Bq/s)	Average value (Bq/s)	Reduction (%)
Uncovered concrete cube	1,83E-05	1,83E-05	-
	1,46E-05		
	1,83E-05		
	2,19E-05		
	1,46E-05		
	1,83E-05		
	2,19E-05		
	1,83E-05		

Concrete sample A	Exhalation rate (Bq/s)	Average value (Bq/s)	Reduction (%)
Concrete cube covered with 2 layers of epoxy product	1,46E-05	1,52E-05	16,67
	1,46E-05		
	1,83E-05		
	1,83E-05		
	1,46E-05		
	1,10E-05		
Concrete cube covered with 2 layers of polymer-based product	1,83E-05	1,77E-05	3,33
	1,83E-05		
	1,46E-05		
	2,19E-05		
	1,46E-05		
	1,83E-05		

Table 6.1.4 – Radon exhalation rates, sample B

Concrete sample B	Exhalation rate (Bq/s)	Average value (Bq/s)	Reduction (%)
Uncovered concrete cube	1,83E-05	2,28E-05	-
	2,56E-05		
	1,83E-05		
	2,19E-05		
	2,19E-05		
	2,56E-05		
	2,19E-05		
	2,92E-05		
Concrete cube covered with 2 layers of epoxy product	2,19E-05	1,83E-05	20,00
	1,83E-05		
	1,83E-05		
	1,83E-05		
	1,83E-05		
	1,46E-05		
Concrete cube covered with 2 layers of polymer-based product	2,56E-05	2,13E-05	6,67
	2,19E-05		
	1,83E-05		
	1,83E-05		
	2,19E-05		
	2,19E-05		

The radon exhalation rate from the uncovered cube prepared with CEM II/B–M 32,5R is 24% higher compared to the rate measured for the uncovered cube prepared with CEM II/A–S 32,5R. This fact may be explained be the higher porosity of the concrete made with CEM II/B-M 32,5 R and respectively air permeability of approximately 2 times higher than in the case of concrete prepared with CEM II/A-S 32,5 R.

In case of cubes covered with epoxy product, the reduction of the exhalation rate was approximately 20 % higher in cube B compared to cube A.

This is also seen in the case of polymer-based product, yet the reduction achieved in cube B is doubled compared to cube A.

The most important issue to note is that for both samples (A and B), the reduction achieved through the application of the 2 components of the epoxy product is approximately 5 times higher than the reduction achieved using the polymer-based product in aqueous dispersion (for A series) and respectively 3 times higher (for B series).

These results are similar to those achieved by the specialists in KVI laboratory in Netherlands.

Conclusions of this survey are displayed in figure 6.1.1.

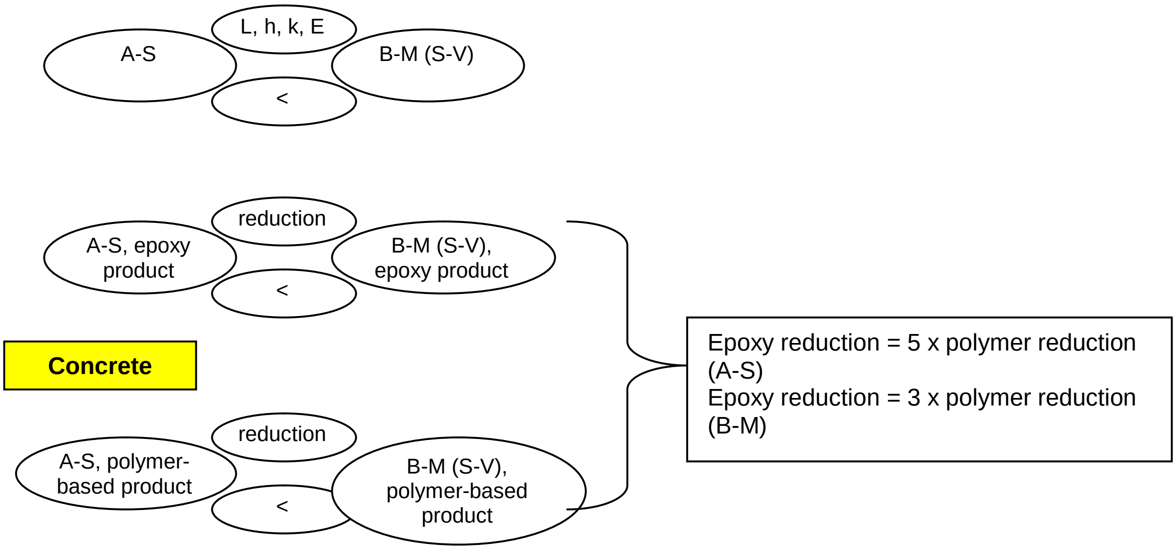


Fig. 6.1.1 – Determinations carried out on 2 types of membranes resistant against radon infiltration

7. CONCLUSIONS AND PERSONAL CONTRIBUTIONS

This paper covers issues related to the presence of radon in building materials, especially in concrete, highlighting the influence of features of concrete ingredients (various cement types, aggregates), as well as the influence of concrete's features on radon exhalation and respectively on radon concentrations inside buildings. Also, the role of environment factors on radon emissions from building materials is approached and, briefly, the main radon reduction methods for existent and new buildings.

7.1. General conclusions resulted from the survey of specialized literature

The physical characteristics of concrete that influence the transport of radon through concrete are porosity, permeability, radon diffusion through concrete, concrete density, water/cement ratio, gel/space ratio.

The main principles that may be noted from the survey of specialized literature are:

- In concrete, the permeability coefficient value decreases significantly with the reduction of water / cement ratio and it is highly dependent on concrete humidity. The shift from an almost saturated state to the dry concrete state determines an increase of air permeability coefficient with approximately two level orders.
- The diffusion coefficient changes with concrete age, as the porous structure thereof changes in time, especially during concrete hydration.
- Gas diffusion through concrete is strongly affected be wet environment, a prolonged treatment of concrete reducing the diffusion coefficient approximately six times.
- Radon diffusion through concrete may be considered an important mechanism for radon infiltration in buildings. The radon diffusion coefficient through pores increases with the concrete's water/cement ratio.
- The reduction of diffusion coefficient is fast in high humidity, which means that increase of water contents in pores causes the block of diffusion transport.
- The diffusion coefficient assessed changes rapidly for emanation coefficients between 0,1 and 0,3 and a thickness of the concrete panel between 4 and 15 cm.
- The values of radon concentration vary based on the ventilation rate of indoor air.
- Radon emissions form concrete in a building depends on the indoor relative humidity, being maximum for a humidity between 30 and 75%.
- Indoor radon concentration depends on the location and sealing faults of buildings, on the soil permeability and humidity, on the indoor ventilation rate, etc.

Radon exhalation from concrete is a combination between radon generation, which is dependent on radium concentration, the concrete emanation coefficient and the type of materials used in preparation of concrete and radon transportation through concrete's microstructure, a process that is dependent on concrete humidity.

Thus, radon exhalation from concrete is influenced by the concentration of radium activity in concrete, by the emanation coefficient, by the radon diffusion coefficient, by the structure and finishing of walls, of concrete panels, by the room ventilation capacity, by the humidity and age of concrete, etc.

For the radon exhalation rates from concrete the following conclusions may be noted:

- Radon exhalation rates from concrete are dependent on concrete age. The values of exhalation rates on the mass unit increase during the first 180-720 after pouring. After this age, exhalation rates decrease with time.
- Air humidity has a significant influence on the exhalation rate. Radon exhalation rates increase with humidity up to 70-75%.
- Fly-ash used in concrete preparation reduces the radon exhalation rate from concrete.
- The exhalation rate may be reduced in a given type of concrete even if its radium contents are high.
- Time dependence of radon exhalation rate is a combination of various factors, most important factor being the concrete humidity. Water pays an important role in the stimulation or delay of radon emission based on material's saturation degree.
- The exhalation rate values decrease with the increase of concrete humidity, especially for humidity between 50-80%.
- Radon exhalation rates from concrete are strongly dependable on temperature, at 50 ^{0}C being of approximately 3,5 times the exhalation rate at 20 ^{0}C; generally, at temperatures exceeding 50 ^{0}C the exhalation rate remains constant.

This paper presents the main methods for reducing radon concentrations in existent and new buildings. Recommendations presented shortly for the application of radon-resistant construction techniques are based on information gathered from numerous research projects, applied in Europe and largely in US.

The construction techniques presented may be applied for individual buildings with one or two stories, with / without basement, that are specific to rural areas or to city outskirts.

For buildings that are to be built, polyethylene – tar board may be applied as barrier against radon.

The efficiency of such radon reduction systems range between 50-99%, based on the method used, on building and soil features, on weather conditions, etc.

The experimental research and risk-benefit assessments made for each proposed method grounded the conclusion that the implementation of depressurizing system underneath the concrete panel is the most efficient radon-reducing method.

7.2. Conclusions generated by experimental research

The main ideas that may be issued based on experimental research carried out on several types of additions used in cement preparation, on cements with/ without additions and respectively on concretes prepared with various concrete types are shown below.

Additions

For the same type of addition, results achieved vary based on the different sources of such addition. Also, high values are noted for radionuclides in fly-ash and slag, compared to those measured for limestone.

Cements

- Contribution of additions tested was assessed by the application thereof in various types of cements. Thus, addition-free cements and cements with additions of slag, fly-ash, limestone, slag+limestone, puzzolana+limestone, slag+fly-ash were tested. The cements tested are manufactured in Romania, at various plants, the percentages of additions used for the same cement type varying from one plant to another.
- Cement with fly-ash addition, CEM II/A-V 42,5 R as a radium activity concentration equal with or higher than the maximum permitted value, based on the source and respectively on the fly-ash percentages used.

- As for the cement with slag addition, CEM II/A-S 32,5 R it has radium concentrations between 45 and 66 Bq/kg, differences being due to sources and slag percentages used in cement manufacturing.
- All cement types tested have lower values of thorium and respectively potassium concentrations compared to the maximum permitted limits in Romania.
- The lowest radioactivity index was achieved for the cement with limestone addition, CEM II/A-LL 32,5 R, a value accounting for 50% of the maximum permitted limit.
- Addition-free cements, CEM I 42,5 R, present radioactivity indexes between 0,31 and 0,34, values that are higher compared to those measured for cement with additions of limestone, results correlated with those achieved for clincher and respectively limestone.
- In case of cements with slag and limestone additions, the values achieved for the reactivity index range between 0,33 - 0,39 due to different slag percentages used in cement manufacturing.
- Cements with slag addition, CEM II/B-S 32,5 R, have radioactivity indexes of 0,36 and respectively 0,37, sensitively higher than the value (0,34) achieved for CEM II/A-S 32,5R, a difference that is due to the higher slag percentage in CEM II/B-S 32,5R.
- In case of CEM II/A-S 32,5 R, values achieved for the radioactivity indexes are 0,45 and respectively 0,39, differences that are due to the various slag sources used.
- The cement with puzzolana and limestone additions, CEM II/B-M (P-LL) 32,5 R, have a radioactivity index of 0,43, a value that accounts for 86% of the maximum permitted value.
- In case of cements with fly-ash addition, CEM II/A-V 42,5 R, the values of radioactivity index are noted to be close to the maximum permitted value.

Aggregates

The values achieved for the 4 sorts used in concrete preparation for radionuclides Ra-226, Th-232 and K-40 are much lower than maximum permitted limits in Romania.

Using the values achieved for cements and aggregates and respectively the percentages of aggregates used in concrete preparation and the cement quantity used for each concrete type tested the concentration of radionuclides activity due to concrete may be assessed using the concrete design.

Concretes. Radioactivity indexes

In case of concretes, we noted that the water submerging period thereof influence the concentrations of radionuclides activity and respectively the radioactivity indexes. Thus, for concretes prepared with the same cement type, values of radioactivity indexes achieved for concretes submerged in water for 7 days after unmolding and then maintained in air until the test age range between 0,25 and 0,38, for concretes submerged in water for 2 days achieving a radioactivity index 30% higher. The increase of water submerging period determines an improvement in concrete’s microstructure.

An increase of the cement dosage used in the preparation of concretes causes an increase of the radioactivity index. For the same cement dosage and for various cement types we can note that concretes submerged in water for 7 days and then cured in air have a radioactivity index that ranges between 0,25 (cement dosage of 320 kg/m^3) and 0,38 (cement dosage of 400 kg/m^3).

Concretes. Compressive strength

The strength features discussed confirm the quality of concretes presented, especially of concretes prepared with superplasticizing admixture and respectively air entrainer.

For the same concrete class, we can note that concretes prepared with cement with slag addition have higher compressive strength than other concretes, disregarding the presence / absence of admixtures. Lower compressive strengths were achieved in case of concretes prepared with cement with limestone and fly-ash additions. Of course, the compressive strength of cements is very important.

Concretes. Porosity

Porosity decreases in time, more significantly in case of concretes prepared with cement with slag and respectively fly-ash additions comparative with concretes prepared with cement with slag and limestone.

Concretes. Air and water permeability

Of all concretes assessed, those prepared with cement with slag have a denser structure, are less permeable and therefore less porous than other concrete types, fact that results from the outcomes achieved for water infiltration depth, air flow depth and respectively for the air permeability coefficient.

Air permeability is influenced by time, especially in case of concretes having low and medium strengths.

Comparing the concretes prepared with the same cement dosage, those prepared with cement with slag addition present air flow depths lower than other types of concretes surveyed, disregarding the presence / absence of admixtures. Generally, high air flow depths were achieved for concretes prepared with cement with limestone and fly-ash additions.

The air flow depth varies based on the cement type, on the admixture type and on the concrete age.

Values achieved for air flow depth account, averagely, for approximately 10-65% of the water infiltration depth determined for samples submitted to a pressure of 4, 8 or 12 bar. In case of admixture-free concretes, this interval ranges between 30-65%, for concretes prepared with superplasticizing admixture 10-60%, and for those with entrained air 15-55%, values increasing in time.

Water infiltration depth and respectively the air flow depth decrease with the reduction of W/C ratio.

In case of admixture-free concretes, at higher cement dosages, 400-470 kg/m^3, we note a slower increase of air flow depth in time compared to the case of concretes prepared with concretes with lower cement dosages, especially in the case of concretes prepared with cements with slag. Same conclusions may be noted for concretes prepared with superplasticizing admixture.

Also, in case of concretes prepared with air entrainer, those with cement with slag addition have an air flow depth lower than other concretes surveyed.

Concretes. Permeability coefficient

For concretes prepared with superplasticizing admixtures and respectively air entrainer, values achieved for the permeability coefficient are several level orders lower than those achieved for admixture-free concretes.

The reduction of permeability coefficient is faster as the W/C ratio is lower. The use of admixtures in the preparation of concretes causes lower W/C ratios and implicitly improved strength and durability features.

The permeability coefficient follows the same trend with the air flow depth, with lower values for concretes with cements added with slag, for the same cement dosage.

The permeability coefficient progresses in time, between 28 days and 1 year the increase varying between 25%-60%, slower evolutions being seen for concretes prepared with cements added slag and respectively for concretes prepared with air entrainers and cements added with fly-ash.

For admixture-free concretes at 28 days, the air permeability coefficient has values 6-20 times higher than air permeability coefficient values achieved for concretes prepared with admixtures, at the same W/C ratio.

At the age of 1 year, values achieved for the permeability coefficient of concretes prepared with admixtures are 15-20 times lower than values achieved for admixture-free concretes, at the same W/C ratio.

The air permeability coefficient decreases with the increase of concrete density and respectively with the increase of compressive strength, higher coefficient values being achieved for admixture-free concretes, at the same W/C ratio. The increase of the water submerging period reduces permeability and porosity of concrete improving its microstructure.

At the age of 28 days, admixture-free concretes with W/C ratios between 0,5 and 0,4, have permeability coefficients determined experimentally similar to those determined theoretically. In case of admixture-free concretes, with W/C ratios exceeding 0,5, values determined experimentally for the permeability coefficient are higher than theoretic values.

Concretes prepared with admixtures have experimentally determined permeability coefficients lower than the coefficients calculated using Rogers formula, disregarding the W/C ratio or the cement type used in the preparation thereof

At 1 year after pouring, admixture-free concretes show measured values of permeability coefficient much higher than calculated values, disregarding the W/C ratio and the cement type use. In case of concretes with admixtures, values determined experimentally for the permeability coefficient are lower than theoretical values, disregarding the W/C ratio, the admixture type and respectively the cement type used in the preparation of concretes.

At the age of 28 days, values determined experimentally for permeability coefficient are similar to those determined theoretically.

For concretes prepared with admixtures, with ages of one year and W/C ratios lower than 0,45, permeability coefficients values determined experimentally ate similar to those determined using Rogers formula.

For concretes with densities higher than 2350 kg/m^3 (values achieved for concretes prepared with admixtures), values determined experimentally for permeability coefficient are similar to those determined theoretically. Rogers formula is verified for concretes with densities exceeding de 2350 kg/m^3.

Concretes. Exhalation rate

As for the radon exhalation rate from concretes, several conclusions may be drawn, synthesized below:

- The radon exhalation rate value per mass unit is noted to decrease with the increase of concrete density, disregarding concrete's age.
- The exhalation rate values per mass unit increase with concrete porosity, disregarding concrete's age.
- The exhalation rate values Em increase in time, with values between (7-12,5) mBq/kgh for concretes at 28 days, (11-16,5) mBq/kgh for concretes at 180 days and (14,5-20,5) mBq/kgh for concretes at one year, the upper section of such intervals being achieved for admixture-free concretes.
- The radon exhalation rate per mass unit vary linearly with the permeability coefficient, achieving higher Em values for high values of the permeability coefficient, i.e. for concretes with low density and high porosity.
- Exhalation rates achieved for concretes prepared with cement with limestone and fly-ash additions are approximately 20-60% higher than the values achieved for concretes prepared with cement with slag addition.
- The radon exhalation rate from concrete vary based on the type and percentage of addition in cement, on the cement dosage, on the presence type and dosage of admixture, on the water / cement ratio, on concrete's porosity, density and age.
- The radon exhalation rate value per surface unit increase in time, values achieved at 28 days after pouring ranging between (1,29-2,44) mBq/m^{2}s, and 1 year after concrete preparation the values range between (2,33-3,79) mBq/m^{2}s.
- The air flow depth and radon exhalation rate vary based on the cement type, on the admixture type and on the concrete age.
- The radon exhalation rate, water infiltration depth and respectively the gas flow depth decrease with the reduction of the W/C ratio.
- The radon exhalation rate from admixture-free concretes is higher than in case of concretes prepared with superplasticizing admixture or air entrainer, at the same W/C ratio.
- In case of concretes prepared with cements with slag, lower values were achieved for the radon exhalation rate than in case of other types of concretes. Highest values were achieved for concretes prepared with concretes with limestone and fly-ash additions.
- The radon exhalation rate from concretes varies linearly with the gas flow depth.

The factors that influence the radon exhalation rates are presented in figure 7.2.1. We may note that this rate is dependent on the concrete features, on environmental factors and on concrete ingredients.

Fig. 7.2.1 – Factors influencing the exhalation rate of radon from concrete

Concretes. Diffusion coefficient

For W/C ratios determined experimentally, ranging between 0,38 and 0,58, diffusion coefficient values were achieved ranging between 8,1 x10^{-9} and 1,1 x10^{-7} m^2/s.

The radon diffusion coefficient decreases with the increase of concrete density and respectively with the reduction of water / cement ratio.

The diffusion coefficient increases linearly with the permeability coefficient, disregarding the concrete's age.

Concretes. Indoor radon concentrations

In case of a room made of concretes prepared with cement with limestone and fly-ash additions, radon concentrations are higher than when using other types of concretes prepared with cement with slag and limestone additions and respectively with cement with slag addition, disregarding the age of concretes surveyed.

Lowest radon concentrations were achieved in rooms made of concretes with cement with slag addition, especially when concrete admixtures were used.

In case of a room made of concretes prepared with cement with slag additions and superplasticizing admixtures, after the age of 1 year after pouring, the lowest value of radon concentration was achieved, approximately 14 Bq/m^3, at a room ventilation rate 0,3 h^{-1} .

The maximum value achieved for the indoor radon concentration is of 20 Bq/m^3, and it was achieved for a room made of admixture-free concrete with cement with limestone and fly-ash, at 1 year after preparation, at a room ventilation rate of 0,3 h^{-1}.

If there are no access ways for radon from soil and if the outdoor air has a concentration of 8 Bq/m^3 (the normal value for outdoor environment), the building may determine an increase of the radon concentration of up to 28 Bq/m^3 .

The values achieved for the indoor radon concentration for the concrete types surveyed are not significant.

Concretes. Barriers against radon

As for the determinations made to test the efficiency of various barriers against radon, significant reductions were achieved using epoxy products applied on the surfaces of concrete samples prepared with cements with slag and respectively slag and fly-ash additions.

7.3. Author's contributions

The main contributions of the author, whose results are shown herein in sections 4, 5 and 6, relate to:

- Determination of the influence of cement additions, cements and aggregates to the concrete's radioactivity;
- Correlation of certain strength (compressive strength) and durability (porosity, water and air permeability) features of concretes prepared with cements with various additions and various admixture types, with the radon exhalation rates;
- Determination of the influence of cement dosages and W/C ratio on radon exhalation;
- Determination of the influence of W/C ratio, of concrete's density and permeability on radon diffusion;
- Determination of exhalation rate from concretes prepared with various cement types on the indoor radon concentrations;
- Determination of the radon exhalation rate reduction using epoxy products.

Data illustrated herein shows a dependence of radionuclides concentration and respectively of the radioactivity index on the type and percentages of additions used when manufacturing cements, especially in case of fly-ash and slag. In case additions with high concentration of nuclides are used, the usable percentages thereof must be determined to prevent exceeding the permitted limitations in Romania.

Also, following experimental research performed, certain conclusions may be drawn on the influence of concrete features on radon transport (figure 7.3.1).

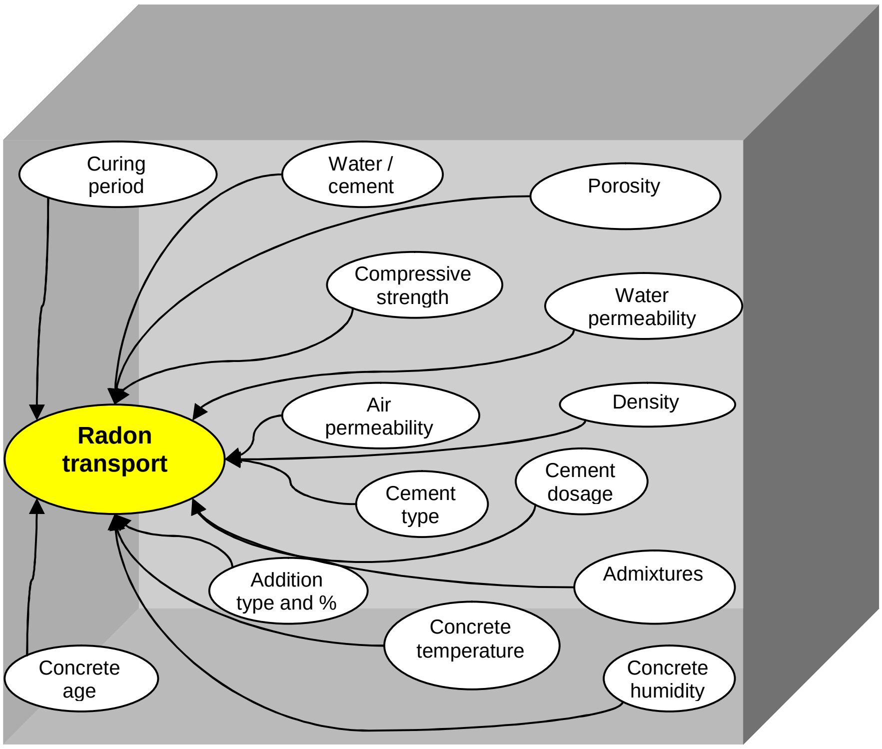


Fig.7.3.1 – Radon transport dependence on concrete's features

When preparing concretes, admixtures must be used as per the legislation in force. The admixtures determine a uniform distribution of cement particles within the concrete mix, reducing W/C ratio and therefore increasing the compressive strength, improving the internal structure and reducing porosity.

We can note that wet treatment for 7 of concretes prepared with blended cements leads to an improvement of concrete's microstructure and implicitly to the reduction of radon exhalation rate and of indoor radon concentration.

The results achieved confirm the quality of concretes prepared with cements with slag, even if results achieved for slag in radioactive contents did not anticipate this. Concretes prepared with cement with slag addition are less permeable to air and water, display a lower porosity compared to other concrete types surveyed, a lower exhalation rate and a lower indoor radon concentration compared to other concrete types (figure 7.3.2).

Fig. 7.3.2 – Indoor radon concentration based on addition types used in cements

A highly important parameter in the relation concrete - radon is the water / cement ratio, a ratio that influences the concrete features and implicitly the radon concentration within buildings.

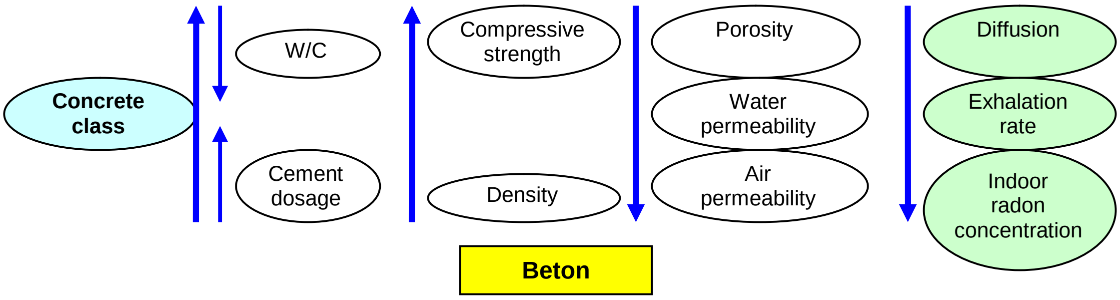


Fig.7.3.3 – Influence of concrete class on diffusion, exhalation rate and indoor concentration of radon

Concluding the results achieved in experimental research, in figure 7.3.3, we may easily note the significance of high concrete classes for the indoor radon concentrations.

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