The Effect of Water to Cement Ratio on Physical and Radiation Shielding Properties of Portland Concrete

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Abstract

Objective: To study the effect of the water to cement ratio (w/c) on 222Rn diffusion, gamma attenuation, neutron attenuation and Ultra-sonic Pulse Velocity (UPV) for local Portland concrete. Methods: The 222Rn diffusion coefficient was measured by putting a cubic concrete sample between two sealed compartments. The linear attenuation coefficients for g-rays from 137Cs and 60Co sources were measured using broad beam experimental setup. Neutron attenuation ratio was calculated by using Am-Be source with neutron flux $10^7$ (n.cm$^{-2}$.s$^{-1}$). The Ultrasonic-Pulse Velocities (UPV) (m.s$^{-1}$) were measured for all the faces of cubic concrete samples. Findings: Results showed that the effect of increasing w/c on linear attenuation coefficients of g-rays and neutron attenuation ratio were not significant and all results were within the uncertainties range. However, in the case of (UPV), the results showed linear relationship with negative slope. In addition, the effective diffusion coefficients $D_e$ of 222Rn in concrete samples increased with increasing (w/c) (on the opposite of the behavior of (UPV) and the value of $D_e$ changed from $(3.6\pm0.39) \times 10^{-10}$ m$^2$s$^{-1}$ for w/c = 0.35 to $(11.0\pm0.7) \times 10^{-10}$ m$^2$s$^{-1}$ for w/c = 0.6. Application/Improvements: This study establishes a reliable test and evaluating procedure for determination of $D$ and $D_e$ by measuring the (UPV) for cubic Portland concrete sample.

Keywords: 222Rn Diffusion, Gamma Attenuation, Neutron Attenuation, (UPV), (w/c)

Introduction

Concrete, in general, consists of coarse and fine aggregates and cement paste which fill the spaces between them and bonds together. Water to cement ratio (w/c) is the main factor determines the porosity and permeability in dry concrete. Decreasing w/c in the mixture of concrete will decrease both porosity and permeability and usually increasing the strength. In addition, during concrete preparation water usually comes from two sources, namely the added water and the moisture in the aggregates. In contrary, the contents of used water on hydrated Portland cement have many effects on the properties of the produced concrete.

Concrete is used in many applications for shielding nuclear constructions like hospital of therapy, Tel-therapy, diagnosis, biological shielding of nuclear reactors…etc. Many local and international studies on biological shielding were performed in the last few decades. Therefore, it is important to study the effect of some physical parameters such as w/c on the linear attenuation coefficient for gamma rays and the attenuation ratio of neutrons.

On the other hand, non-destructive test using Ultrasonic Pulse Velocity (UPV) measurement of concrete specimens is important method to determine the physical properties of dry concrete. The typical (UPV) for ordinary concrete is between 3700-4200 (m/s). Concrete is a porous materials and the porosity in concrete affects its properties such as density, permeability and the strength. So the permeability increases with increasing the porosity, which relates with the hydration degree, w/c, air content and kind of aggregates.

This work aims to study the effect of w/c on UPV, radon gas diffusion, linear attenuation coefficient and...
neutron attenuation ratios on local Portland concrete were studied.

2. Background Theory

The diffusion coefficient of radon gas in building materials is often used as an indication for radon transportability through a porous medium and, furthermore, as an essential tool for quantitative predictions of radon concentrations in dwellings. The values of radon diffusion coefficients for concrete are found (under laboratory or in-situ conditions) in the range from $7 \times 10^{-10}$ to $1.5 \times 10^{-7}$ $(\text{m}^2 \cdot \text{s}^{-1})^{14-16}$. These values were determined by putting a slab or a cubic concrete sample between two sealed compartments, one kept at a high radon concentration (compartment one), while the other empty of radon or background (compartment two) as illustrated in Figure 1. Then the building-up of $^{222}\text{Rn}$ in compartment 2 is measured by the time.

![Figure 1. The diffusion of $^{222}\text{Rn}$ in one dimension through concrete sample from the compartment one to the compartment two.](image)

This method has the following drawbacks:
1. Difficulty in radon-tight sealing between the concrete sample and the two compartments and between the inside and outside environments.
2. The pressure difference could lead to additional transport due to advection and/or convection.

Experimentally, the diffusion equation of a radioactive gas in porous medium could be used to determine the diffusion coefficient. Equation (1) shows the one dimensional diffusion of a radioactive gas in the steady state $^{14}$.

\[
(D_e / p)(d^2 C_x / dx^2) - lC_x = 0
\]

where:
- \(p\): the porosity of the medium (intestinal volume/total volume, a pure number),
- \(C_{(x)}\): radon concentration in the pores of the materials (Bq.m$^{-3}$),
- \(l\): Radon decay constant (s$^{-1}$), equal to $2.06 \times 10^{-6}$ (s$^{-1}$).
- \(D_e\): effective diffusion coefficient (m$^2$.s$^{-1}$). The relationship between the diffusion coefficient \((D)\) and \(D_e\) is given by the following equation$^{14,17}$:
\[
D_e = D.p
\]

Taking account of the boundary condition for this experiment specified in Figure 1, Equation (1) is solved as follows:

\[
C_{(d)} = C_0 \cdot \exp \left( - \frac{D_e}{p} \cdot d \right) \quad \text{or} \quad C_{(d)} = C_0 \cdot \exp \left( - \sqrt{\frac{2 \cdot l}{D_e}} \cdot d \right)
\]

where, \(C_{(d)}\) is the average concentration of $^{222}\text{Rn}$ recorded in the compartment two during flux measurements and \(d\) the thickness of the sample \((d = 0.1 \text{ m})\).

In this study the count rates (cpm: count per minute) were considered instead of $^{222}\text{Rn}$ concentration (Bq.m$^{-3}$) because of the direct proportion between of them.

3. Materials and Methods

Five concrete mixtures were prepared from local aggregates and Portland cement with equal ratios of the components excluding the water to cement ratio. The ratios of w/c were 0.35, 0.40, 0.45, 0.50, and 0.60. Tables 1 and 2 show the ratios of the components and sieve grading for the used aggregates respectively.

Table 1. The ratios of the components in the five examined concrete mixtures

| Cement   | Fine aggregates | Coarse aggregates | plasticizer | w/c |
|----------|-----------------|-------------------|-------------|-----|
| 1.00     | 1.33            | 2.66              | 0.03        | 0.35 – 0.60 |

![Figure 2. Schematic of radon diffusion measurement system.](image)
The results of X-rays diffraction (XRD) shows that the local dolomite contents are 97% to 95% CaMg(CO$_3$)$_2$ + 3% to 5% CaCO$_3$.

Five replicated cubic samples (10´10´10) cm were prepared from each mixture (total 25 samples). The cubes were conserved in water bath (22 °C) for 28 days and then left to dry for few days.

3.1 Description of Gamma and Neutron Attenuation Tests

The linear attenuation coefficients for g-rays from $^{137}$Cs and $^{60}$Co sources were measured using broad beam experimental setup illustrated in a study carried out previously. The linear attenuation coefficient was calculated by the following equation:

$$h = \frac{1}{10 \times (\ln (I_0 / I))}$$  \hspace{1cm} (4)

Neutron attenuation ratio was calculated also for neutron from Am-Be source with neutron flux $10^7$ (n.cm$^{-2}$.s$^{-1}$) as the ratio of measured dose rate with and without the sample.

3.2 Description of Ultra-Pulse Velocity (UPV) Test

The ultrasonic-pulse velocities (UPV) (m.s$^{-1}$) were measured for all the faces of cubic concrete samples. UPV was calculated using the following equation:

$$V = (\Delta x / \Delta t)$$  \hspace{1cm} (5)

Where $\Delta x$ the thickness of the concrete sample and $\Delta t$ the passage time of ultra-sonic pulse through this thickness.

3.3 Description of Radon Diffusion Tests

Figure 2 illustrates the experimental set-up to determine the diffusion coefficient of radon through a given concrete sample.

The volume of permeable voids (porosity %) were measured in concrete sample by using the procedure ASTM: C 642-06, this test method covers the determinations of density, percent absorption, and percent voids in hardened concrete, while, it does not involve a determination of absolute density.

4. Results and Discussion

Figure 3 shows changes of the averages linear attenuations coefficients for $^{137}$Cs, $^{60}$Co and Figure 4 shows the attenuation ratio of Am-Be neutron source. Statistical evaluation for the results has shown that there is no significant effect for the changes in w/c ratio on shielding properties of Portland concretes. The differences were within the statistical errors range. This agrees with the results of Kharita’s study.

Table 2. Sieve grading for the used aggregates

| Sieve no. (inch) | Percent passing, by weight (%) |
|-----------------|-------------------------------|
|                 | Coarse Aggregate | Fine aggregates |
| 1               | 100              | 100             |
| 3/4             | 90               | 100             |
| 1/2             | 59               | 100             |
| 3/8             | 2                | 100             |
| N4              | 0                | 96              |
| N8              | 0                | 52              |
| N16             | 0                | 26              |
| N30             | 0                | 16              |
| N50             | 0                | 11              |
| N100            | 0                | 7               |
| N200            | 0                | 4               |

Figure 3. Linear attenuation coefficient values for gamma ray of $^{137}$Cs and $^{60}$Co vs w/c.

The UPV significantly influenced by porosity of the concrete samples. Some authors studied the influence of porosity on the (UPV). They found that (UPV) decreases with increasing porosity of the concrete samples. Figure 5a shows the results that confirm this relationship, where the UPV attenuated by the voids. Also the porosity ratio influenced by the w/c ratio, Figure 5b shows this relation, where the porosity ratio increases by increasing the w/c ratio. Consequently, the (UPV) decreased with increasing the w/c ratio.
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Figure 4. Relation between neutron attenuation ratio for Am-Be source vs w/c.

Figure 5. The relation between the (UPV) vs. the (p %) and the (p %) vs (w/c).

In addition, Figure 6 shows the change of (UPV) vs (w/c) in the concrete mixtures for three different frequencies (24, 54, 150 kHz), where the frequency 24 kHz gives the highest velocities for the studied concrete samples.

Figure 6. The change of (UPV) vs (w/c) in the mixture concrete for three frequencies.

Moreover, $^{222}\text{Rn}$ diffusion through concrete samples was measured for all mixtures by using two-compartment method. The “Box Lucas1” Model built in OriginPro 8.5 program was used for fitting the results of the built up of the count rates in the compartment two through the time. The extrapolation that depends on building up $^{222}\text{Rn}$ at the steady state can help to solve the equations (3).

Figure 7 shows the building up of $^{222}\text{Rn}$ exhaled to compartment two through the concrete sample, which has the (w/c) ratio (0.35). It is clear that all values are converged to value (133 ± 14 cpm). From equation (3), the effective diffusion coefficient ($D_e$) was calculated to be $(3.6 ± 0.4) \times 10^{-10}$ (m$^2$.sec$^{-1}$).

Figure 7. The build-up $^{222}\text{Rn}$ for concrete mixture of w/c = 0.35.

In the same way, Figure 8 shows the building up of $^{222}\text{Rn}$ exhaled to compartment two through the concrete sample, which has the (w/c) ratio (0.40). It also found that

Figure 8.
the values are converged to value (143.3±14.3 cpm) and $D_e$ equal to (3.9±0.4) $\cdot 10^{-10}$ m$^2$.s$^{-1}$.

Figure 8. The build-up $^{222}$Rn for mixture concrete of w/c = 0.40.

Similar fitting was applied for all the concrete mixtures of (w/c) ratios (0.45, 0.5 and 0.6). Table 3 contains the count rate, the values of $D$ for $^{222}$Rn according (w/c), the porosity values $p\%$ (which were measured by using the method ASTM C 642-06) and the values of $D_e$ that calculated by equation (2). The obtained results were comparable to those published by other authors for (w/c) > (0.4) which were in the range from 7$\cdot 10^{-10}$ to 1.5$\cdot 10^{-7}$ (m$^2$.s$^{-1}$) $^{14}$, $^{15}$, $^{16}$. In addition, the results were close to the published value which was (4.5±0.4)$\cdot 10^{-10}$ m$^2$.s$^{-1}$ for the concrete mixture with (w/c = 0.35)$^{25}$.

Table 3. The values of the count rate, $D$, the measured porosity $p\%$ and $D_e$ for different w/c ratios

| w/c | Count Rate (cpm) | $D$ ($10^{-9}$) | The measured porosity ($p\%$) | $D_e$ ($10^{-10}$) |
|-----|-----------------|----------------|-------------------------------|--------------------|
| 0.35 | 133 ± 14 | 3.1 ± 0.3 | 11.70 ± 0.17 | 3.6 ± 0.4 |
| 0.40 | 145 ± 14 | 3.3 ± 0.3 | 11.88 ± 0.17 | 3.9 ± 0.4 |
| 0.45 | 200 ± 17 | 4.4 ± 0.4 | 12.02 ± 0.20 | 5.2 ± 0.5 |
| 0.50 | 240 ± 13 | 5.2 ± 0.3 | 12.20 ± 0.15 | 6.3 ± 0.4 |
| 0.60 | 396 ± 17 | 9.2 ± 0.4 | 12.36 ± 0.12 | 11.0 ± 0.7 |

Figure 9 reveals the relationship between the calculated $D$ and $D_e$ of $^{222}$Rn through the concrete samples for different values of w/c. As seen, the $D$ and $D_e$ increased by increasing w/c.

The Figure 10 shows the relationship between the diffusion coefficient and the effective diffusion coefficient against the UPV. It could be noticed from Figure (10) that the two curves were getting closer toward each other when the UPV increases, that could be attributed to a decrease in the porosity of samples. This agrees with the studies done by many authors$^{22-24}$.

Figure 9. The relationship between diffusion coefficient and effective diffusion coefficients of $^{222}$Rn through concrete samples and w/c.

Figure 10. The changing of diffusion and effective diffusion coefficients of $^{222}$Rn with (UPV) in five mixture of concrete samples.

The found relationship is useful for estimating the $D$ and $D_e$ of $^{222}$Rn through Portland concrete samples by measuring the (UPV). The (UPV) can be measured easily in short time, while measuring $D$ or $D_e$ by two-compartment method needs long time and sealing the sample without any leakage in the radon diffusion measurement system.

5. Conclusion

The change of w/c does not significantly affect the ionizing radiation attenuation as much as it affects the mechanical and physical properties of concrete.
The values of ultra-sonic pulse velocity (UPV) in concrete samples were strongly correlated with \((w/c)\) ratio. There was a linear relationship between (UPV) and \((w/c)\), where the (UPV) decreases with increasing \((w/c)\). The frequency 24 kHz gave greater (UPV) values than 54 and 150 kHz.

The ratio \((w/c)\) had a clear effect on \(^{222}\text{Rn}\) diffusion through concrete samples. The diffusion coefficients \(D\) were calculated by using the solution of the steady state equation for one-dimensional diffusion of a decaying substance. The values of \(D\) were changed from \((3.1 \pm 0.3) \times 10^{-9} \text{ (m}^2\text{s}^{-1})\) for \((w/c = 0.35)\) to \((9.2 \pm 0.4) \times 10^{-9} \text{ (m}^2\text{s}^{-1})\) for \((w/c = 0.6)\). Also, the values of effective diffusion coefficients \(D_e\) were changed from \((3.6 \pm 0.4) \times 10^{-10} \text{ (m}^2\text{s}^{-1})\) for \((w/c = 0.35)\) to \((11.0 \pm 0.7) \times 10^{-10} \text{ (m}^2\text{s}^{-1})\) for \((w/c = 0.6)\). Consequently, decreasing \(^{222}\text{Rn}\) diffusion can be made by decreasing the ratio \(w/c\).

Furthermore, the relation between \(^{222}\text{Rn}\) diffusion coefficient and (UPV) establishes a reliable test and evaluating procedure for determination of \(D\) and \(D_e\) by measuring the (UPV) for cubic Portland concrete sample.

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7. References

1. Roy DM. Relationship between permeability, porosity, diffusion and microstructure of cement pastes, mortar and concrete at different temperatures, materials. MRS Proceedings. 1988; 137:1–5. https://doi.org/10.1557/PROC-137-179
2. Reddy VV, Ramana NV, Gnaneswar K, Sashidhar C. Effect of Magnesium Chloride (MgCl2) on ordinary portland cement concrete. Indian Journal of Science and Technology. 2011 Jun; 4(6):1–3.
3. Sharma DK, Sharma R. Reduction of water soluble hexavalent chromium in hydrated portland cement. Indian Journal of Science and Technology. 2015 Jun; 8(11):1–6. https://doi.org/10.17485/ijst/2015/v8i11/71773
4. Nirmala R, Rajkumar R. Finite element analysis of Buried UPVC pipe. Indian Journal of Science and Technology. 2016 Feb; 9(5):1–5. https://doi.org/10.17485/ijst/2016/v9i5/87225
5. Jaeger RG. Engineering compendium on radiation shielding. Shielding Materials, Springer-Verlag: Heidelberg, 1975; 2:1–7. https://doi.org/10.1007/978-3-642-65001-7
6. Kaplan MF. Concrete radiation shielding. John-Wiley and Sons Inc: USA; 1989.
7. Knoll GF. Radiation detection and measurement. Third Edition, John Wiley & Sons: USA; 2010.
8. Khait MH, Takeyeddin M, Nassar M, Yousef S. Development of special radiation shielding concretes using natural local materials and evaluation of their shielding characteristics. Progress in Nuclear Energy. 2008; 50(1):30–6. https://doi.org/10.1016/j.pnucene.2007.10.004
9. Yousef S, AlNassar M, Naoom B, Alhajali S, Khait MH. Heat effect on the shielding and strength properties of some local concretes. Progress in Nuclear Energy. 2008; 50(1):22–6. https://doi.org/10.1016/j.pnucene.2007.10.003
10. Alhajali S, Khait MH, Naoom B, Yousef S, AlNassar M. Estimation of the activation of local reactor shielding concretes. Progress in Nuclear Energy. 2009; 51(2):374–7. https://doi.org/10.1016/j.pnucene.2008.04.007
11. Hobbs B, Tchoketch K. Non-destructive testing techniques for the forensic engineering investigation of reinforced concrete buildings. Forensic Science. 2007; 167(2–3):167–72. https://doi.org/10.1016/j.forsciint.2006.06.065 PMid:16904854
12. Albert S, Birks B, Robert E, Green JR, MacIntire P. Nondestructive testing handbook. Ultrasonic Testing, the American Society for Nondestructive Testing. 1991; 7(3):273–82.
13. Roy DM, Brown PW, Shi D, Scheetz BE, May W. Concrete microstructure porosity and permeability. National Research Council: Washington DC; 1993.
14. Folkerts KH, Keller G, Muth K. Experimental investigations on diffusion and exhalation of \(^{222}\text{Rn}\) and \(^{220}\text{Rn}\) from building materials. Radiation Protection Dosimetry. 1984; 7(1):41–4. https://doi.org/10.1093/rpd/7.1-4.41
15. Renken K, Rosenberg T. Laboratory measurements of the transport of radon gas through concrete samples. Health Physics. 1995; 68(6):800–8. https://doi.org/10.1097/00004032-199506000-00006 PMid:7759258
16. Gadd MS, Borak TB. In-situ determination of the diffusion coefficient of \(^{222}\text{Rn}\) in concrete. Health Physics. 1995; 65(6):817–22. https://doi.org/10.1097/00004032-199506000-00008
17. Raabe OG. Measurement of the diffusion coefficients of RaA. Nature. 1968; 217(3):11–43.
18. Yousef S, AlNassar M, Naoom B, Alhajali S, Khait MH. Heat effect on the shielding and strength properties of some local concretes. Progress in Nuclear Energy. 2008; 50(1):22–6. https://doi.org/10.1016/j.pnucene.2007.10.003
19. Albert S, Birks B, Robert E, Green JR, MacIntire P. Nondestructive testing handbook. Ultrasonic Testing, the American Society for Nondestructive Testing. 1991; 7(3):273–82.

20. ASTM C642-06. Standard test method for density, absorption, and voids in hardened concrete, ASTM International, West Conshohocken, PA [Internet]. [cited 2016 Mar 05]. Available from: www.astm.org.

21. Kharita MH, Youssef S, AlNassar M. The effect of the initial water to cement ratio on shielding properties of ordinary concrete. Progress in Nuclear Energy. 2010; 52(5):491–3. https://doi.org/10.1016/j.pnucene.2009.11.005

22. Winkler KW, Nur A. Seismic attenuation: effects of pore fluids and frictional sliding. Geophysics. 1982; 67(1):1–15. https://doi.org/10.1190/1.1441276

23. Ohdaira E, Masuzawa N. Water content and its effect on ultrasound propagation in concrete – the possibility of NDE. Ultrasonics. 2000; 38(1–8):546–52.

24. Vergara I, Miralles R, Gosalbez J, Juanes FJ, Ullate LG, Anaya JJ, Hernandez MG, Izquierdo MAG. NDE ultrasonic methods to characterise the porosity of mortar. NDT & E International. 2001; 34(8):557–62. https://doi.org/10.1016/S0963-8695(01)00020-2

25. Cozmuta I, van der Graaf R. Methods for measuring diffusion coefficients of radon in building materials. Science of the Total Environment. 2001; 272(1–3):323–35. https://doi.org/10.1016/S0048-9697(01)00711-2