STOCHASTIC METHODS APPLIED TO THE MORTAR LEACHING MODEL BASED ON INDUSTRIAL MARBLE BY-PRODUCT WASTE

AZHAR BADAOUI[1]* , M'HAMMED BADAOUI [2]
[1]*Ecole Nationale Supérieure des Travaux Publics-Francis Jeanson- ENSTP Sidi Garidi Kouba Laboratoire de Recherche LTPiTE, Algiers, Algeria*
[2]*FEHR Technologies Teppes Châteauneuf-Sur-Isere, France*

The industry poses many problems to the disposal of waste materials; the impact on the environment and space occupied by storage sites. It is necessary to find a way to valorize and re-use this waste to protect the environment.

The research aims to apply a probabilistic approach to the simple leaching of mortars based on mineral fines of industrial by-products such as marble powder by pure or low mineralized water to maximize its recycling in the formulation of cementitious matrix building materials and to solve ecological and economic problems. The leaching phenomenon is studied using a probabilistic approach in this article. The statistics of the dissolving front in the material are investigated using a parametric approach that takes into account the variation of the mortar's calcium diffusion coefficient porosity. A lognormal probability distribution is used in Monte Carlo simulations. In comparison to solely deterministic studies, the study found that probabilistic approaches are effective tools coupled with adequate deterministic models, providing extra information. As a result, concrete buildings may be better tailored for specific service life and environmental conditions.

Keywords: *leaching; coefficient of variation, simulation, mortar, marble waste*

1. Introduction

Waste is one of the best indicators of a society's economic vitality and lifestyle. Any production or consumption activity generates waste, which is often associated with the deterioration of our environment. Due to their specificity and complexity, part of the waste is sent to specialized sectors. Among the various management methods applied to them, recovery is privileged for environmental and economic reasons.

The main waste streams recovered are residues from thermal operations (slag or fly ash) recovered in cement works or civil engineering, vegetable waste recovered as animal feed, and ferrous metal waste recycled in metallurgy and metal production. Marble powder is one of the industrial by-products wastes that occupy large storage sites and pollute the environment. Using marble waste in the manufacture of mortars and concrete helps to reduce the cement cost production and to manage the problem of storing industrial by-products.

Probabilistic approaches to leaching and modeling via the Monte Carlo technique are used to study the longevity of mortars based on mineral fines from industrial by-products such as marble powder. The degradation of the cementitious matrices during leaching is mainly related to the diffusion of interstitial water ions into the aggressive medium, coupled with the progressive dissolution of hydrates. This ionic diffusion is, therefore, the motor of cement degradation during leaching.

2.Leaching theory

Leaching is the re-dissolution of solid calcium phases in the hydrates of cement pastes when the porous material is exposed to site water that is weakly ionized or low in calcium. In this case, the calcium present in the interstitial solution in ionic form diffuses towards the outside environment, leading to a decrease in calcium concentration in the pores [1, 2]. To restore the local chemical balance between the interstitial solution and the solid phases, the hydrates are dissolved. The dissolving of the material increases its porosity, increasing the material's permeability and decreasing its strength.

2.1.Simple leaching

The most straightforward concrete degradation process is simple leaching with pure or low mineralized water. Since the pore solution in concrete pores is fundamental (pH around 13), weakly mineralized water constitutes an aggressive environment. The concentration gradients between the pore solution of the cementitious material and "pure" water, therefore, lead to the diffusion of most ions (alkaline, calcium, hydroxyl, etc.) towards the outside environment. The main chemical reactions of hydrolysis/decalcification of the two main phases of cementitious materials (portlandite and C-S-H) appear in equation (1), with n varies between 0.1 and 1.8 and characterizing the CaO= SiO_2 ratio of C-S-H (in the presence of portlandite, n is equal to the maximum value, i.e. between 1.7 and 1.8) [3, 4]

$$Ca^{2+}+2H_2O \longleftrightarrow Ca(OH)_2+2H^+ \quad (1)$$

$$nCa^{2+}+(SiO2)aq+2nH2O \longleftrightarrow (C\text{-}S\text{-}H)+2nH^+ \quad (2)$$

$$H_2O \longleftrightarrow H^++OH^- \quad (3)$$

The original solid/solution chemical equilibrium arising from the hydration activities is changed due to the ion transport mechanisms.

* Autor corespondent/*Corresponding author*,
E-mail: azharbadaoui@yahoo.fr

2.2.Simple leaching model

Based on the diffusion of pore water ions and the local chemical equilibrium of portlandite, ettringite, and mono-sulfoaluminate hydrates, ADENOT established a deterministic model for leaching degradation of cement matrices. This model divided the leaching zone into several sub-zones of constant mineralogy. It provided information on the thickness degraded over time and the mineralogical fronts developed within the leaching zone; this approach is illustrated in Fig. 1 [5].

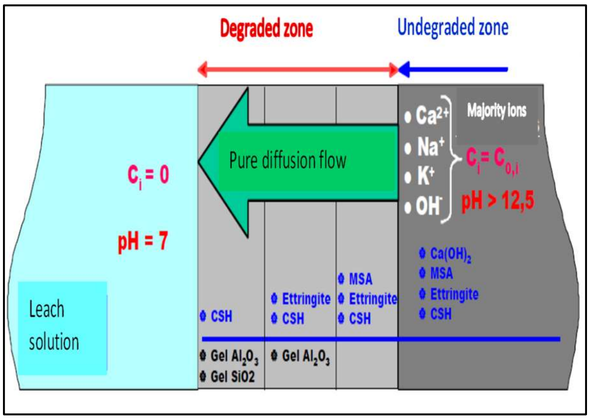

Fig. 1 - Degradation model of water-leached cementitious matrices from ADENOT .

The thickness of the degraded zone is defined by the portlandite solubility front, which is the most soluble hydrate of the cement; the amount of calcium released is dependent on the square root of time (solution of the diffusion equations). A characteristic leaching time (T_{leach}) for the different hydrate is defined and compared to a reference value. [6, 7]

$$T_{leach} = \frac{l^2_{leach}}{Dca}\left(1 + \frac{\partial SCa}{\partial CCa}\right) \quad (1)$$

l_{leach} : Characteristic length of the Leach phenomenon; l_{leach} = 50 µm.

$\frac{\partial SCa}{\partial CCa}$: Derived from the mass concentration of solid calcium relative to the concentration of calcium in solution. It ranges between 10 and 20; the higher this term, the faster the corresponding hydrate dissolves.

Dca : Calcuim coefficient of diffusion in the water. It is equal to $3.7.10^{-12}$ m^2.s for a healthy material and 3.10^{-10} m^2.s for a degraded material.

3.Theoretical formulation

3.1.Probabilistic Analysis of mortar leaching

In a marble powder mortar, the effect of spatial variability of the leaching degradation kinetics parameters is investigated. The difference between a deterministic and a probabilistic simulation life estimate is quantified. To assess the impact of material variability on service life, we used two numerical applications.

The statistical investigation was carried out using the Monte-Carlo method, which is the simplest probabilistic integration method since it is simply a matter of calculating many simulations with independent realizations of the input random variable [7].

The first application compares the lifetime of a marble powder-based mortar and a reference mortar subjected to degradation in pure water determined by a standard deterministic calculation with the results of Monte-Carlo simulations using random fields with spatial correlation as input variables.

The second application investigates the effect impact of uncertainty on the most crucial leaching parameters, which are porosity and the coefficient of calcium diffusion in water, on leaching kinetics. Statistics on leaching time were compiled using a parametric approach that factored in the influence of these parameters' coefficients of variation.

The calcium diffusion coefficient and mortar porosity are assumed to be random variables with a lognormal distribution when simulations are run. The calcium diffusion coefficient (DCa) and the parameters of the lognormal distribution of porosity (ε) are expressed as follows [8].

$$\mu_{\ln\varepsilon} = \ln(\mu_\varepsilon) - \frac{1}{2}\sigma^2_{\ln\varepsilon} \quad (2)$$

$$\sigma^2_{\ln\varepsilon} = \ln\left(1 + \frac{\sigma^2_\varepsilon}{\mu^2_\varepsilon}\right) \quad (3)$$

$$\mu_{\ln DCa} = \ln(DCa) - \frac{1}{2}\sigma^2_{\ln DCa} \quad (4)$$

$$\sigma^2_{\ln DCa} = \ln\left(1 + \frac{\sigma^2_{DCa}}{\mu^2_{DCa}}\right) \quad (5)$$

Where ($\mu_{\ln\varepsilon}$, $\sigma^2_{\ln\varepsilon}$) and ($\mu_{\ln D_{Ca}}$, $\sigma^2_{\ln D_{Ca}}$) are statistics (mean and variance) of porosity (ε) and calcium diffusion coefficient (DCa), these parameters are assumed uncertain with lognormal probability, which is suitable for strictly non-negative. Since for large values of coefficient of variation, simulations with a normal distribution can give negative values, this is ideal.

Monte Carlo simulations are performed, 10000 independent samples of the parameters (ε) and (DCa) with lognormal distribution are generated, and a deterministic numerical procedure is applied to each simulation, providing 10000 values of the leach time parameters [9].

Finally, the time factor statistics (mean, standard deviation, and confidence interval) are computed in the last step.

The cement used in the formulation of the mortars is a portland cement NA442 CEM II/B-L 42.5N, of Blaine surface area equal to 3895 cm^2/g , its chemical composition is shown in Table 1.

The sand used is a quarry sand of granular class 0/3, of fineness modulus Mf = 2.9, the apparent and absolute densities of this sand are;

Table 1

Cement chemical composition

Element	Value %
SiO_2	22,33
Al_2O_3	4,35
Fe_2O_3	3,30
CaO	58,00
MgO	1.93
SO_3	2 ,31
Na_2O	0,14
K_2O	0,82
P.F	6.82

ρapp= 1.59 g/cm^3, ρabs= 2.55 g/cm^3.

The marble powder is a white powder with a granulometry equal to 450 μ, a fineness of 3600 cm^2/g, absolute density ρabs = 2.70 g/cm^3 , and a mass absorption of 0.30%. The chemical analysis carried out on this powder is shown in Table 2.

Table 2

Marble powder chemical composition

Element	Value %
SiO_2	0.42
Al_2O_3	0,13
Fe_2O_3	0,06
CaO	56,01
MgO	0.12
P_2O_5	0 ,03
Na_2O	0,44
SO_3	0,01
P.F	42.78

The formulated mortars comply with the EN 196-1 standard, the sand/cement, and water/cement ratios are respectively: S/C = 3 and W/C = 0.4

The mortars used are M0 and M1which M0 is the reference mortar and M1 is the mortar based on marble powder with a 20% substitution rate (Table 3).

Table 3

Formulated mortars

Constituent	M0 (control)	M1
Cement (g)	400	320
Marble powder(g)	00	80
Sand(g)	1200	1200
Water(g)	160	160

The mean values (μ) and the coefficients of variation (CV) of the different parameters were estimated respectively from Model Code FIB proposals.

μ_ε = 40% and CV_ε varies from 0.1% to 0.5%.

μ_{DCa} = 3,7.10^{-12} m^2.s and CV_{DCa} varies from 0 to 0.05

$\mu_{(SCa/CCa)}$ = 4,7.10^{-12} m^2.s^{-1} and $CV_{(SCa/CCa)}$ =0

The mass concentration of solid calcium SCa = 14700 mol m^{-3} and the concentration of calcium in solution CCa = 22 mol m^{-3}

4.Results and discussions

The leaching time coefficient of variation behavior versus the realizations number is investigated. For several realizations Nsamp about 800, the ultimate settlement coefficient of variation convergence versus (ε) and (DCa) is seen; this value is selected equal to 10000 (Fig. 2) [9].

The Chi-Square fitness test is used to evaluate assumed probability distribution time parameters related to the appropriate histogram forms suggest a log-normal distribution, that is suitable for strictly non-negative random variables with a significant variation value coefficient, because simulations can produce negative values in this case as a result of the normal distribution (Fig. 3).

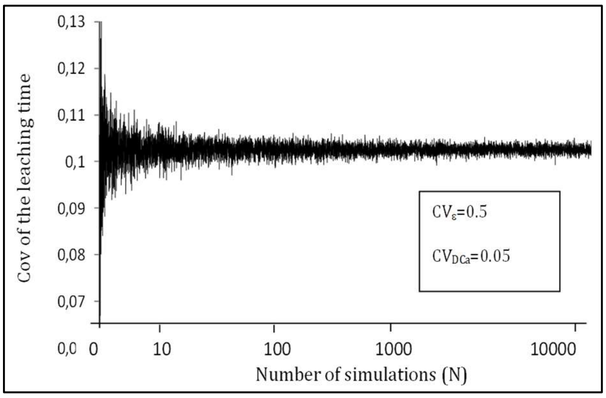

Fig. 2 - Leaching time versus ε and DCa coefficient of variation.

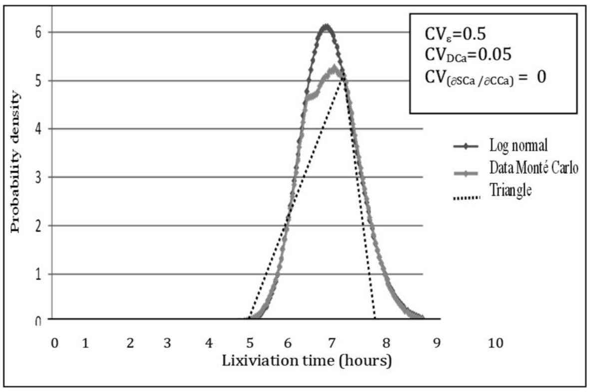

Fig.3 - Probability density versus leaching time.

Figures 4 and 5 show the results of Monte-Carlo simulations comparing probabilistic and deterministic lifespans related to M0 (control), and M1 (marble powder mortar).

Probabilistic lives are significantly shorter than deterministic lifetimes. Moreover they decrease as the coefficients of variation $CV\varepsilon$=0.5,

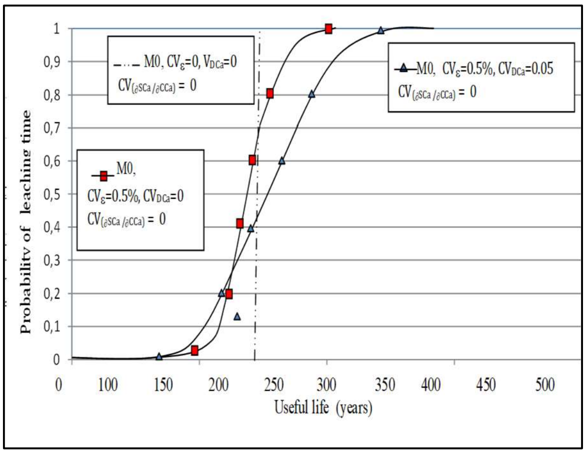

Fig. 4 - Service life probability of M0.

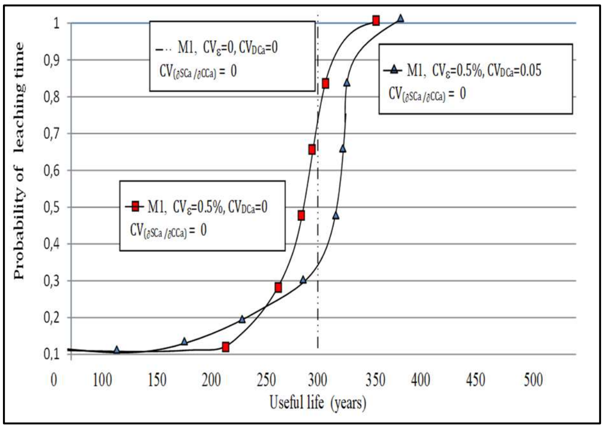

Fig. 5 - Service life probability of M1.

and CV_{DCa}=0.05 for mortars M0 and M1 become more prominent.

The deterministic and probabilistic lifetimes of the marble powder mortar (M1) are larger than those of CEMII cement alone (M0); this is due to the extra mineral (marble powder) in the M1 formulation. The addition of marble powder to cement alters its physicochemical properties, making the paste less rich in portlandite than concrete pastes.

On the one hand, marble powder decreases the interstitial solution pH by simple diluting action, directly related to portlandite consumption, the C/S ratio of C-S-H, and hydrate forms [10].

On the other hand, the clinker containing less CaO, and the pozzolanic reactions due to the additions consume the portlandite by secondary reactions.

Furthermore, as demonstrated by the work of Duchesne and Berube [11, 12], the C-S-H created by such pozzolanic processes often have lower C/S ratios than the C-S-H produced by clinker hydration. The diffusiveness of a material decreases with mineral additions. The transfer properties are determined by the pore network parameters, including the number of pores, the size, and the pores of the solid matrix connections. Marble powder may not necessarily lower total porosity. However, the tortuosity of porosity increases as the size and interconnectivity of the pores decreases, resulting in a drop in the diffusion coefficient and a slowing of the hydrolysis process.

We should also add the Codina experiment, which shown that low pH binder pastes deteriorate in leaching at roughly four times the rate of Portland cement pastes [13].

This first part aims to study the spatial variability effect of the material properties on the propagation speed of the portlandite dissolution front in the structure. The lifetime estimated by a probabilistic simulation increases by 60% with the homogenous fields.

In the second part, the uncertainties effect of the porosity and the calcium diffusion coefficient in water is examined. Performing a parametric analysis that incorporates the influence of these parameters' coefficients generated Leach time statistics.

Simulations are carried out assuming that the calcium diffusion coefficient and mortar porosity are random variables with a log-normal distribution. The mean, standard deviation, and confidence interval statistics are obtained from 10,000 simulated random samples by performing a parametric study.

4.1.Leaching time statistics as a function of the mortar porosity variability for M0

The leaching time statistics as a function of mortar porosity variations are shown in Figure 6 and 7. By varying the coefficient of variation of calcium diffusion coefficient CV_{DCa} from 0 to 0.05, Figure 6 depicts the mean leaching time variation with mortar porosity coefficient of variation ($CV\varepsilon$) and for a coefficient of variation of the solid calcium relative mass concentration to the calcium in solution concentration $CV_{(SCa/CCa)}$ =0. This figure illustrates that when the coefficient of variation of the calcium diffusion coefficient increases, the mean leaching time increases for all values of the porosity coefficient of variation $CV\varepsilon$.

Figures 6 and 7 show the leaching time statistics as a function of mortar porosity variability. Figure 6 expresses the mean leaching time variation with mortar porosity coefficient of variation ($CV\varepsilon$) and for a coefficient of variation of the solid calcium relative mass concentration to the calcium in solution concentration $CV_{(SCa/CCa)}$ =0, by varying the coefficient of variation of calcium diffusion coefficient CV_{DCa} from 0 to 0.05. This figure shows that, for all values of porosity coefficient of variation $CV\varepsilon$, an increase in the mean leaching time is observed when the coefficient of variation of calcium diffusion coefficient increases.

Figure 7 depicts the relationship between the standard deviation of leaching time and the porosity coefficient of variation. Again, the curves are strewn around, and the standard deviation is significant for high CV_{DCa} levels.

The confidence intervals of the mean and standard deviation of the leaching time to the

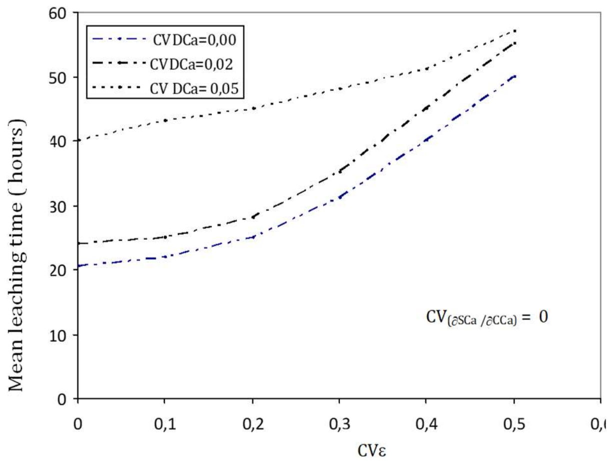

Fig. 6 - Mean leaching time versus CVε for M0.

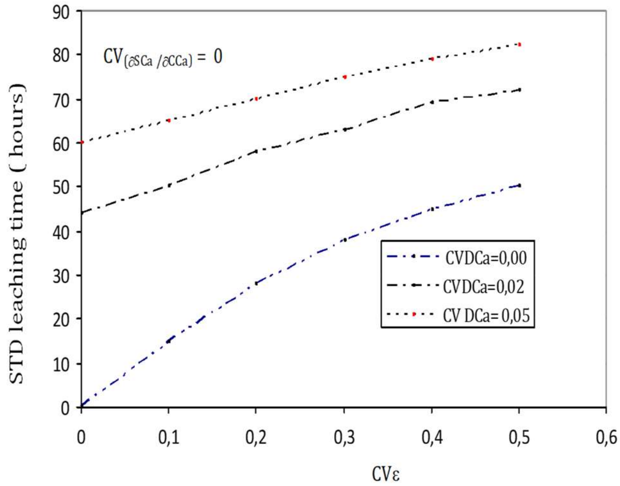

Fig. 7 - STD leaching time versus CVε for M0.

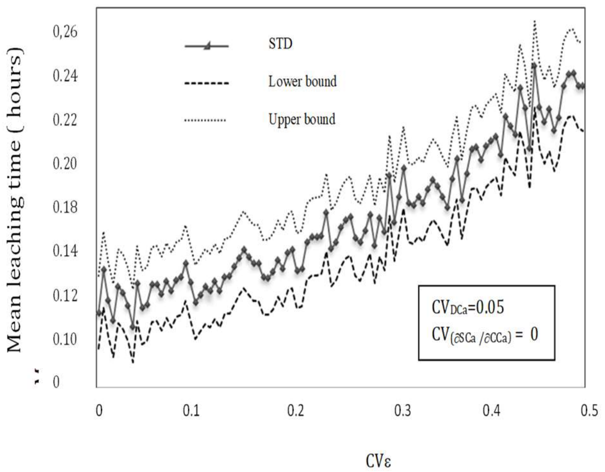

Fig. 8 - Mean statistics and confidence intervals versus CVε for M0.

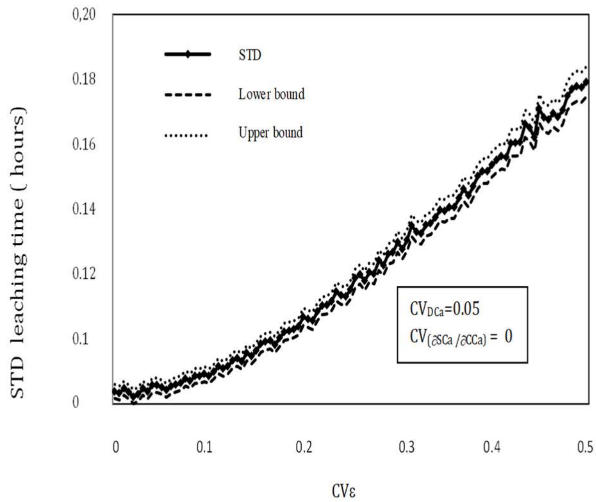

Fig. 9 - STD statistics and confidence intervals versus CVε for M0.

porosity coefficient of variation are shown in Figures 8 and 9. The confidence interval measures the sample estimates' imprecision.

For an assumed log-normal distribution and from the statistics (mean and standard deviation, as well as the 95% confidence interval) of the leaching time to mortar porosity coefficient of variation, it is shown that the mean value increases that as the coefficient of variation of CV_{DCa} and $CV\varepsilon$ increases.

The standard deviation also increases uniformly, suggesting that porosity variability has a substantial impact on the mean leaching time and strongly influences its dispersion. The diffusion processes involved in leaching are aided by an increase in porosity and, as a result, its diffusion coefficient. The dissolution of portlandite crystals, which range from a few micrometers to several hundred, creates pores sufficiently to contribute to the increase in capillary porosity, while C-S-H hydrolysis only affects the Nano porosity of hydrate gels. It appears that decalcification affects just mesoporosity between micro and macroporosity [14]._ As the portlandite and C-S-H hydrolysis are not simultaneous, an evolution in porosity is observed over the degraded thickness with an area more exposed to the aggressive solution. These findings imply that porosity (ε) uncertainty should be included in any stochastic concrete or mortar study.

4.2. Leaching time statistics as a function of the calcium diffusion coefficient variability for M0

Figures 10 and 11 show the leaching time statistics as a function of calcium diffusion coefficient variability.

Figure 10 presents the mean leaching time variation with the coefficient of variation of calcium diffusion coefficient (CV_{DCa}) for a coefficient of variation of the mass concentration of solid calcium related to the concentration of calcium in solution $CV_{(SCa/CCa)}=0$, varying the porosity coefficient of variation $CV\varepsilon$ from 0 to 0.5.

The mean values curves of the leaching time become significant only when CV_{DCa} increases,

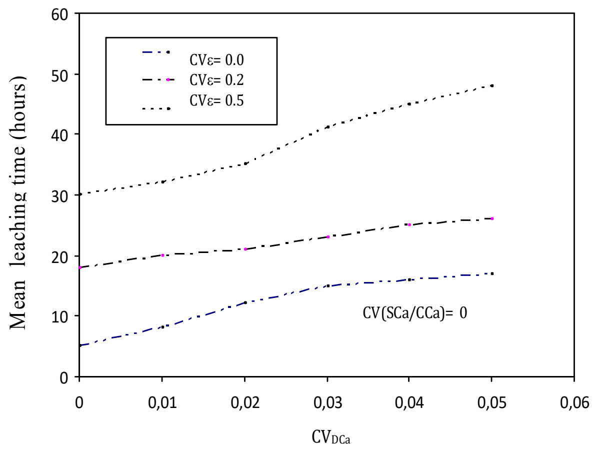

Fig. 10 - Mean leaching time versus CV_{DCa} for M0.

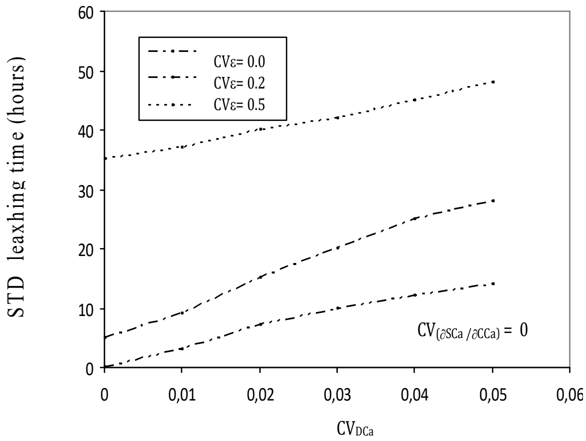

Fig. 11 - STD leaching time versus CV_{DCa} four M0.

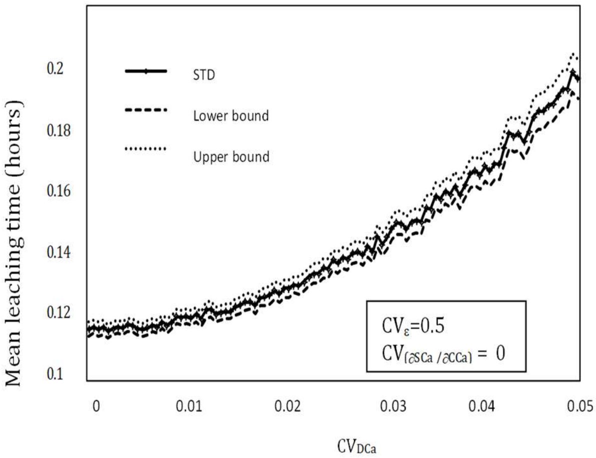

Fig. 12 - Mean statistics and confidence intervals versus CV_{DCa} for M0.

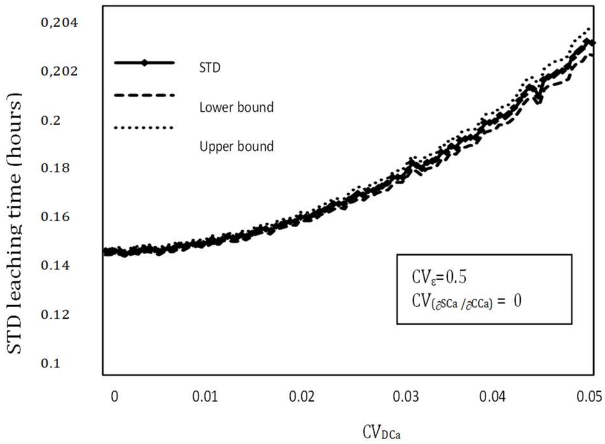

Fig. 13 - STD statistics and confidence intervals versus CV_{DCa} four M0.

suggesting that DCa variability has a more substantial influence on the leaching time.

In Figure 11, the standard deviation is more minor for low values of $CV\varepsilon$, it is equal to zero for $CV\varepsilon= CV_{(SCa/CCa)}= 0.0$, it increases significantly with CV_{DCa}. This suggests that the calcium diffusion coefficient and porosity uncertainty is an essential parameter that should be considered in any stochastic research.

Figures 12 and 13 illustrate the mean and standard deviation confidence interval of the leaching time to the coefficient of variation of calcium diffusion coefficient CV_{DCa}. The mean value of the leaching time increases, indicating that the uncertainty in calcium diffusion coefficient (DCa) leads to a delay in the leaching process. An increase in the standard deviation with a parabolic curve can also be seen when the calcium diffusion coefficient variation increases.

The porosity evolution and the increase in the connectivity of the pores for the mortar M0 and its impact on the diffusion coefficient have also been studied. For a cement paste with a W/C ratio of 0.4, the diffusion coefficient in the in the healthy material is 3.710^{-12} m^2 s^{-1} and rises to 3.10^{-10} m^2 s^{-1} for a degraded or leached material [15, 16].

As the capillary porosity created by leaching is lower, the porosity created in the C-S-H gels is too low to form a percolation network, which would significantly impact the diffusion coefficient.

4.3.Leaching time statistics as a function of the mortar porosity variability for M1

The leaching time data are shown in Figures 14 and 15 as a function of mortar porosity variations.

Figure 14 depicts the mean leaching time variation with the coefficient of variation of mortar porosity ($CV\varepsilon$) for $CV_{(SCa/CCa)}$ =0 and changing the calcium diffusion coefficient of variation from 0 to 0.05. The graph depicts this for $CV\varepsilon$ values. The mean values curves of the leaching time are scattered. The leaching time increases only when the CV_{DCa} and $CV\varepsilon$ increase.

Figure 15 shows the standard deviation leaching time variation with the porosity coefficient of variation. We note that for $CV\varepsilon = CV_{DCa}= CV_{(\partial SCa/\partial CCa)} = 0$, the standard deviation equals zero; it increases considerably with $CV\varepsilon$ and CV_{DCa}. It can be observed that the standard deviation

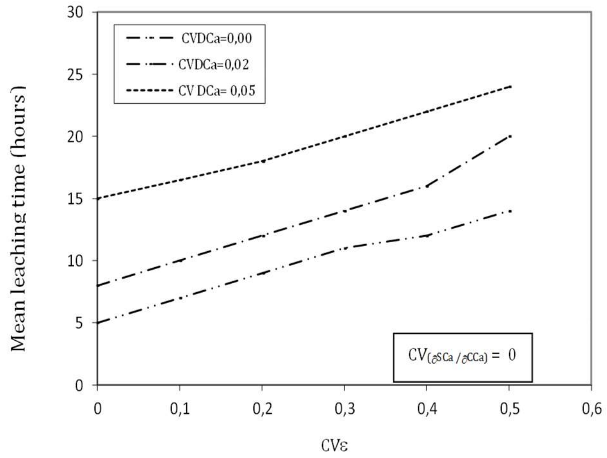

Fig. 14 - Mean leaching time versus CVε for M1.

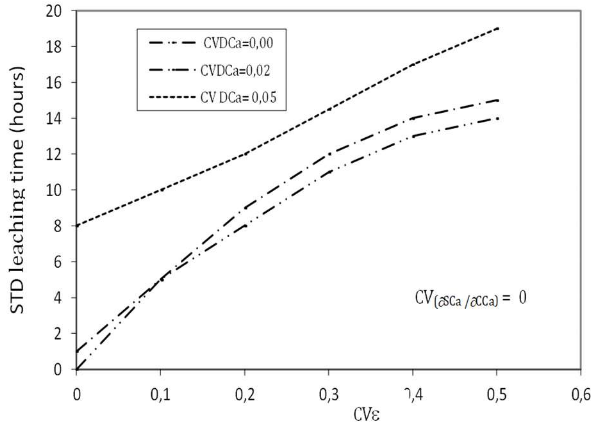

Fig. 15 - STD leaching time versus CVε for M1.

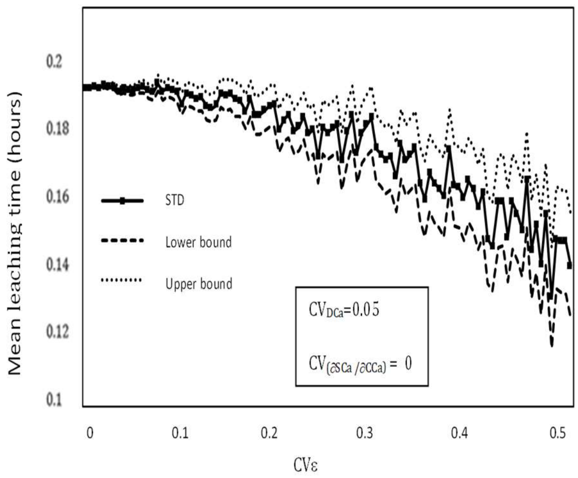

Fig. 16 - Mean statistics and confidence intervals versus CVε for M1.

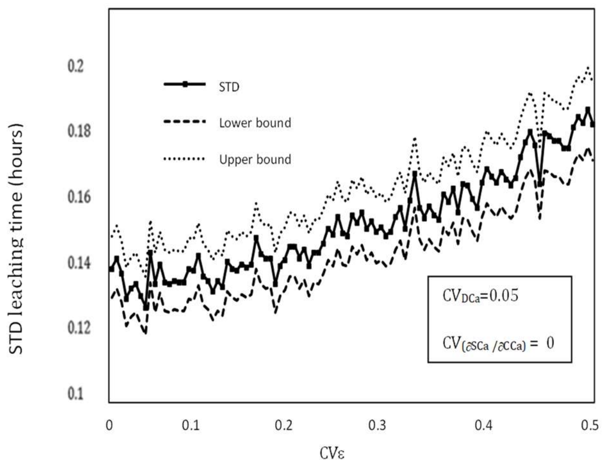

Fig. 17 - STD statistics and confidence intervals versus CVε for M1.

leaching time curves are confounded for the values of CV_{DCa} = 0.02 and 0.0, the dispersion becomes large for CV_{DCa} = 0.05, that as the CVε increases.

The influence of the porosity coefficient of variation on the leaching time is small for deterministic values of CV_{DCa}. The value of leaching time increases significantly with CV_{DCa} and CVε.

For CV_{DCa} deterministic values, the influence of the porosity coefficient of variation on the leaching time is negligible; it increases with CV_{DCa} and CVε.

The leaching time mean and standard deviation confidence interval to the porosity coefficient of variation are shown in Figures 16 and 17.

The variation of CVε from 0.0 to 0.5 for values of CVDCa = 0.05 and $CV_{(\partial SCa/\partial CCa)}$ = 0 reduces the carbonation depth by 26%. There is a slight increase in the standard deviation, but the confidence interval remains constant, indicating that variability in porosity does not affect the accuracy of the standard deviation.

4.4. Leaching time statistics as a function of the calcium diffusion coefficient variability for M1

Figures 18 and 19 present the leaching time statistics as a function of calcium diffusion coefficient variability. They illustrate the variation of the mean and standard deviation leaching time with the coefficient of variation of calcium diffusion coefficient (CV_{DCa}), for $CV_{(SCa/CCa)}$ =0 and varying the porosity coefficient of variation CVε from 0 to 0.5. It is observed that the mean leaching time increases when the porosity coefficient of variation increases, it is more important for CV_{DCa} = 0.05 than for CV_{DCa} = 0.0.

We note that the standard deviation curves are slightly scattered and the value of the standard deviation increases as CVε increases.

Figures 20 and 21 illustrate the confidence interval of the mean and standard deviation of the leaching time to the coefficient of variation of porosityε.

A slight decrease in the mean and the

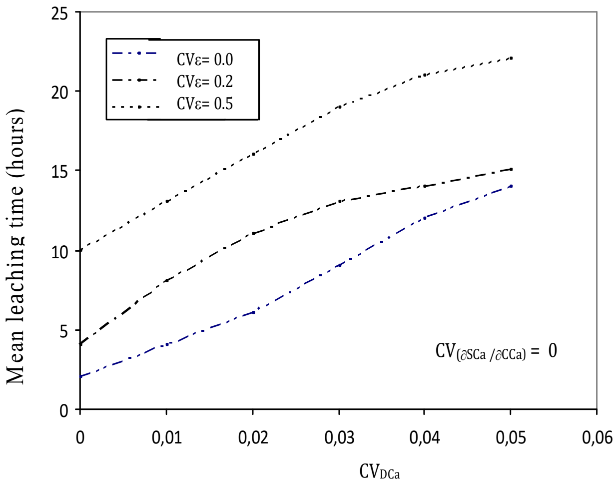

Fig. 18 - Mean leaching time versus CV_{DCa} for M1.

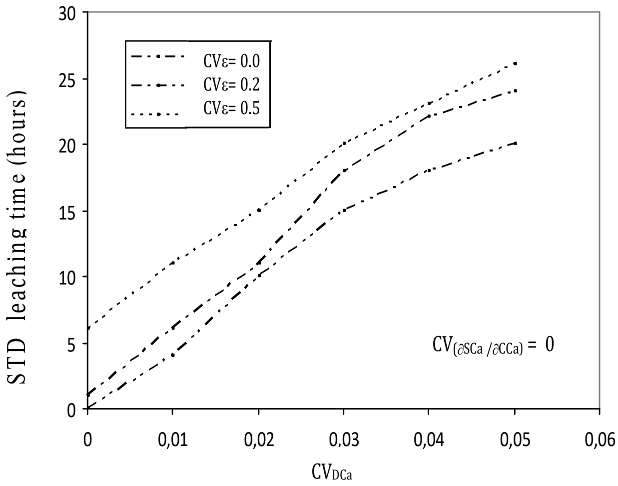

Fig. 19 - STD leaching time versus CV_{DCa} for M1.

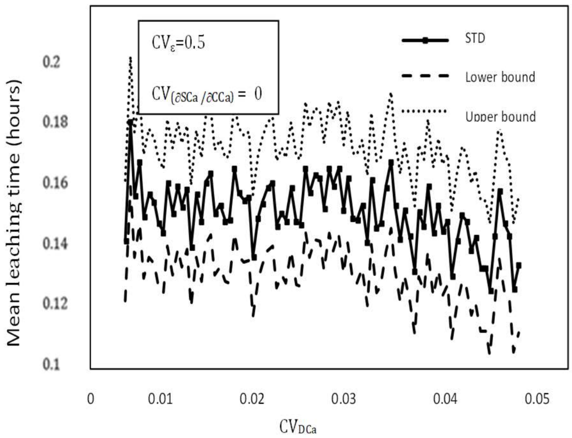

Fig 20. Mean statistics and confidence intervals versus CV_{DCa} for M1.

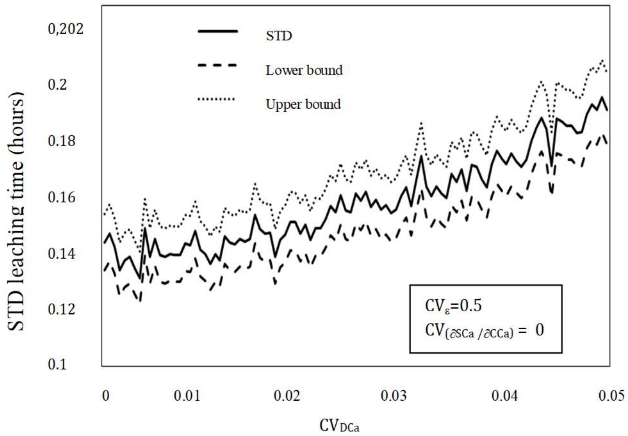

Fig 21 - STD statistics and confidence intervals versus CV_{DCa} for M1.

confidence interval remains large and constant for the means and standard deviation, which indicates that the CV_{DCa} variability influences the leaching time with a small effect on the mean value.

The marble powder reduces the portlandite content which can be leached (by pozzolanic reaction). The higher the rate of substitution of cement by marble powder, the lower the capillary porosity created by leaching, so that the porosity created in the C-S-H gels is too low to form a percolation network that would significantly influence the diffusion coefficient.

5. Conclusion

The primary purpose of this study is to apply stochastic methods to the simple leaching by pure or low mineralized water in mortars based on industrial marble by-product waste and compare it to a reference mortar without marble powder, to valorize its recycling in the formulation of cementitious matrix building materials and solving the ecological and economic problems. The statistical investigation was carried out using the Monte-Carlo method, which is the simplest probabilistic integration method since it is simply a matter of calculating many simulations with independent realizations of the random input variables.

In addition, the spatial variability of the leaching degradation kinetics parameters in a marble powder mortar is investigated, and the difference between a deterministic and a probabilistic simulation life estimate is quantified.

Two numerical applications were conducted to evaluate the effect of material variability on the service life. The first application involves comparing the lifetime estimated by a classical deterministic calculation of a marble powder-based mortar and reference mortar subjected to degradation in pure water with the results of Monte-Carlo simulations considering the input variables as random fields with spatial correlation.

The second application analyzes the influence of uncertainties on the most influential parameters on leaching: porosity and the diffusion coefficient of calcium in water on the leaching kinetics in mortars.

The leaching time statistics have been carried out by performing a parametric analysis that integrates the influence of these parameters' coefficients. The mean, standard deviation, and confidence interval statistics are generated using a parametric survey of 10,000 simulated random samples.

Results demonstrate that the added marble powder to cement used to prepare the mortar modifies the paste's physico-chemical characteristics, which contains less portlandite. In addition, the marble powder reduces the interstitial solution pH related to portlandite consumption and the C/S ratio of C-S-H and hydrate forms.

The C-S-H produced by pozzolanic reactions due to the addition of marble powder has generally lower C/S ratios than the C-S-H from direct hydration of clinker; the work of Duchesne and Berube has confirmed these results.

The transfer properties depend on the pore network characteristics, pores to the solid matrix, size, and connectivity of the pores. Marble powder increases the tortuosity of the porosity while reducing pore size and connectedness, which explains the decrease in the diffusion coefficient and, as a result, the slowing of the hydrolysis process. It is found that a deterministic simulation with uniform fields equal to the mean value of examined parameters provides a lifespan more significant than 60% of the probabilistic simulation, including variability.

Leaching time statistics show that the variability of porosity (ε) has a significant effect on the mean leaching time and strongly influences its dispersion. The dissolution of portlandite crystals, which vary in size from a few micrometers to several hundred, creates pores sufficiently to contribute to the increase in capillary porosity.

Portlandite and C-S-H hydrolysis is not simultaneous; it affects the Nano porosity of the hydrate gels. It would seem that decalcification only affects the mesoporosity between the micro and macro porosity, which varies in size from a few micrometers to several hundred, and creates enough pores to contribute to the increase in capillary porosity.

The evolution of porosity and its impact on the calcium diffusion coefficient, as far as decalcification also increases pore connectivity. The calcium diffusion coefficient (DCa) uncertainty causes a delay in the leaching process. The presence of the marble powder in mortar reduces the content of portlandite that can be leached. The more marble powder is present as a substitute for a certain amount of cement, the lower the capillary porosity created by leaching. The porosity created in the C-S-H gels is too small to form a percolation network, which would affect the diffusion coefficient.

These results indicate that uncertainty about porosity (ε) is among the parameters that should be incorporated into any stochastic concrete or mortar investigation.

Acknowledgments

This work is supported by the General Direction of Scientific Research and Technological Development (DGRSDT) of Algeria.

REFERENCES

[1] I. Arribas, I and I. Vegas, The deterioration and environmental impact of binary cement containing thermally activated coal mining waste due to calcium leaching, Journal of Cleaner Production, 2018, **183**, 887-897.

[2] Y.Tang and X. Zuo, Influence of calcium leaching on chloride diffusivity in cement-based materials", Construction and Building Materials,2018, **174**, 310-319.

[3] M. Jebli and F. Jamin, Leaching effect on mechanical properties of cement-aggregate interface. Cement and Concrete Composite,2018, **87**, 10-19.

[4] R. Ragoug and O. Omikrine, Durability of cement pastes exposed to external sulfate attack and leaching: Physical and chemical aspects, Cement and Concrete Research., 2019, **116**, 134-145

[5] F. Adenot, Durabilité du béton : Caractérisation et modélisation des processus physiques et chimiques de dégradation du ciment , Doctorate thesis, Université d'Orléans,1992.

[6] Y. Zhang and A. Khennane, Numerical modelling of degradation of cement-based materials under leaching and external sulfate attack, Computers and Structures, 2015,**158**, 1-14.

[7] D. Gawin and F. Pesavento, Modeling deterioration of cementitious materials exposed to calcium leaching in non-isothermal conditions, Computer Methods in Applied Mechanics and Engineering , 2009, **198**, 3051–3083.

[8] R. Caflisch, Monte Carlo and Quasi-Monte Carlo Methods". Acta. Numerica, 1998, **7** ,1- 49.

[9] A. Badaoui, Probabilistic carbonation simulations in concrete based on marble powder, Materials Sciences Forum, 2020, **1013**, 14-119.

[10] H.Yang and Y. Che, Calcium leaching behavior of cementitious materials in hydrochloric acid solution, Scientific Reports, 2018, **8**, 8806 L.

[11] A. Issaadi, Experimental assessment of the variability of concrete air permeability: repeatability, reproducibility and spatial variability, Ener. Proc, 2017, **139**, 537–543.

[12] T.Rougelot and C. Peng, Why is it necessary to use a damage model to simulate the mechanical behavior of concrete under drying and leaching ?, European Journal of Environmental and Civil Engineering , 2010, **14**, 923–935

[13] M. Codina, Les bétons bas pH - Formulation, caractérisation et étude à long terme, Doctorate thesis, INSA Toulous, 2007.

[14] J. Golaszewski, Influence of cement properties on new generation superplasticizers performance, Construction and Building Materials,2012, **35**, 586-596.

[15] A. BehnoodKim and V. Tittelboom, Methods for measuring pH in concrete: A review, Construction and Building Materials, 2016, **5**, 176-188.

[16] N. Nguyen , Leaching of Alkali from Concrete in Contact with Waterways, Water, Air, & Soil Pollution, 2009, **9**, 381.

****