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Durability of a reinforced concrete designed for the construction of an intermediate-level radioactive waste disposal facility

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ABSTRACT

The National Atomic Energy Commission of the Argentine Republic is developing a nuclear waste disposal management programme that contemplates the design and construction of a facility for the final disposal of intermediate-level radioactive wastes. The repository is based on the use of multiple, independent and redundant barriers. The major components are made in reinforced concrete so, the durability of these structures is an important aspect for the facility integrity. This work presents an investigation performed on a reinforced concrete specifically designed for this purpose, to predict the service life of the intermediate level radioactive waste disposal facility from data obtained with several techniques. Results obtained with corrosion sensors embedded in a concrete prototype are also included. The information obtained will be used for the final design of the facility in order to guarantee a service life more or equal than the foreseen durability for this type of facilities.

1. Introduction

Since 1950, the National Atomic Energy Commission (CNEA) of the Argentine Republic works on the development of nuclear energy applications for pacific purposes. The tasks include, among others, the research and development of basic and nuclear technology areas, the operation of important facilities in charge of the production of radioisotopes for medical and industrial applications and the performance of tasks in connection with the nuclear fuel cycle, mining and uranium processing activities, manufacturing of fuel elements, and the operation of two nuclear power plants. As a result of such activities performed in the nuclear field by CNEA and other private and public entities, various types of radioactive waste are being produced. The CNEA is the implementing authority to perform all activities related to the radioactive waste management and sets up the Radioactive Waste Management National Program (PNGRR) [1]. In order to achieve its objectives the PNGRR is in charge of the adoption of the most appropriate technological solution for the management of such wastes and the scientific-technological support. At the end of the 1990s, the CNEA started an extended research programme which final purpose was to design a facility for the disposal of Intermediate Level Radioactive Waste (ILRW) and to study the long-term behaviour of the materials used for its construction.

ILRW is originated from operation and maintenance activities of the two Argentine Nuclear Power Plants (Atucha I and Embalse), and consist mainly of mechanical filters from the primary circuit of the reactor and by spent ionic exchange resin beds. Such ILRW is under interim storage at the facilities of each power plant awaiting treatment and conditioning, but most of this waste will arise from the decommissioning of the nuclear power plants. Then, the start up of the ILRW repository is linked to the decommissioning of the nuclear power plants and it should be operative approximately by the year 2023.

In this context, a near-surface monolithic repository based on those in operation in El Cabril, Spain, is foreseen. The conceptual design of this repository is the use of multiple, redundant and independent barriers, and the model considers a 300 years post-closure institutional control. An sketch of the proposed model is shown in Fig. 1 [2].

The barriers foreseen for the ILRW repository can be divided into two categories: physical and chemical barriers. The physical barriers are meant to avoid the intrusion of water, people and animals, and the release of radionuclides; while the chemical barriers are intended to restrict radionuclide migration by adsorption and ion-exchange once the soluble radionuclides have been released from the source. The barriers which will be used in the near-surface repository include: waste forms and metallic disposal containers (waste will be immobilized in cement matrices and packed in 200 l drums or in special concrete containers), backfill and buffer

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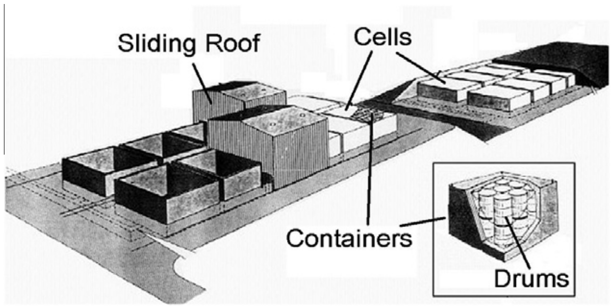

Fig. 1. Sketch of the repository design.

materials, vaults and cover and geological media. The vaults and covers are major components of the engineered barriers, and due to the fact that these barriers are made of reinforced concrete, its durability is an important aspect regarding the integrity of the facility. So, the research and development are focussed to design a durable concrete and to establish the methodology to determine its durability, through the knowledge of its performance in the disposal conditions. Of course, durability will be influenced by the environment to which the concrete is exposed to, by the quality of the constituents and their proportion in the mix design and by the structural designs and construction techniques, among other factors. Those uncertainties make the prediction of performance for 300 years impossible to be fully validated. Therefore, as a general criteria, a conservative approach for degradation has to be developed.

This work presents laboratory investigations performed on concrete specimens made with a formulation candidate to be used in the construction of the ILRW disposal facility in order to predict the service life of concrete vaults and cells from data obtained with available techniques. In addition, a concrete prototype was built and instrumented with sensors specially designed, in order to monitor the evolution of the rebar state in real conditions. Finally, a comparison between laboratory and field data were done.

2. Experimental technique

The study was performed using concrete specimens containing rebar segments (Fig. 2). The specimens are 25 cm high and have a square section of 15 cm per side. Each specimen contained 3 rebar segments (in the *as-received* condition, without traces of rusting) of the DNA-420 type, two straight ones and one containing two welds. The rebars are 10 mm diameter and present an exposed area of 30 cm^2. The chemical composition of the reinforcing bars used in the study is as follows: C, 0.41%; Mn, 0.73%; Cu, 0.27%; Ni, 0.13%; Si, 0.28%; P, <0.01%; S, 0.02%; N, 0.008%, Fe, balance. At the concrete-air interface the rebars were isolated by epoxy resin in order to avoid crevice corrosion. The rebar segments were positioned in such a way that a 3 cm concrete cover was achieved. Each specimen contains a metal–metal oxide (MMO) internal reference electrode (IRE) [3,4] with a diameter of 3 mm and 50 mm long, located in its centre.

Table 1
Composition and properties of the concrete used in the present work.

Water (kg/m^3)	154.1
Cement CAH40 (SR) IRAM 50,000 (kg/m^3)	416.4
Sand (kg/m^3)	813.4
Coarse aggregate (kg/m^3)	995.2
Additives (kg/m^3)	2.94
w/c	0.37
Compressive strength (MPa) 28 days	53.6 ± 1.3
Capillary suction (g/cm^2)	886
Air permeability (m^2)	0.018×10^{-16}

The concrete used in this study was selected based, among other characteristics, on the mechanical properties and workability. The composition of the concrete used, as well as some of its properties are summarised in Table 1.

Two specimens were cast and, after a curing period of 28 days at 100% relative humidity, the specimens were exposed to the laboratory atmosphere, with temperatures between 20 °C and 25 °C and 60–70% relative humidity.

The electrochemical parameters normally used to characterise the corrosion behaviour of reinforcing steel in concrete were monitored periodically during approximately 1600 days (about 4.4 years). These parameters include the corrosion potential (E_{corr}), the corrosion rate (*C.R.*) obtained from polarisation resistance (R_p) measurements, and the electrical resistivity of concrete (ρ) determined from resistance measurements between the straight rebar segments and the IRE.

The concrete electrical resistance (R_s) was measured using an earth ohm-meter Metrel Smarctec MI 2124. These values were used to estimate the electrical resistivity of the concrete as $\rho = k \cdot R_s$, where k is the geometric cell constant measured experimentally with solutions of known electrical resistivity.

The E_{corr} were measured using a high input impedance multimeter (Brymen BM815) connected to the IRE. These measurements were then corrected against a copper/saturated copper sulphate reference electrode (SCE) [3,4]. The results where evaluated according to ASTM standard C-876 [5]. R_p was evaluated as $\Delta V/\Delta I$ [6] from potential sweeps within E_{corr} ± 0.01 V at a scan rate of 10^{-4} V/s, using a Gamry CMS-100 potenciostat–galvanostat. The results were corrected to compensate the $I \cdot R_s$ drop error. R_p values were used to calculate the rebar corrosion current density (I_{corr})

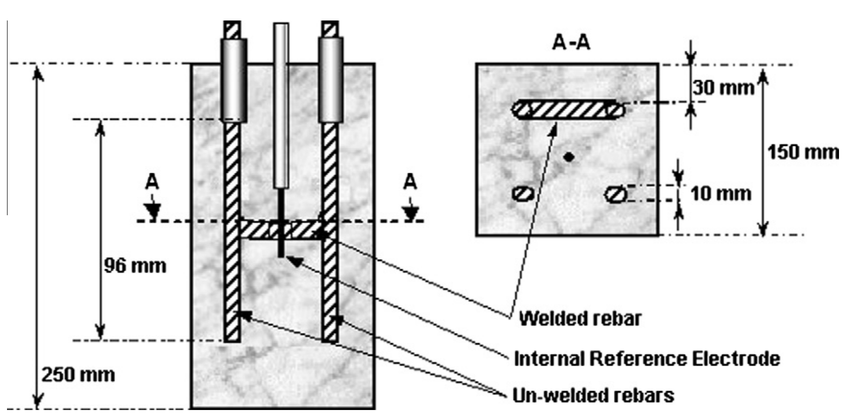

Fig. 2. Sketch of the specimens employed for laboratory measurements.

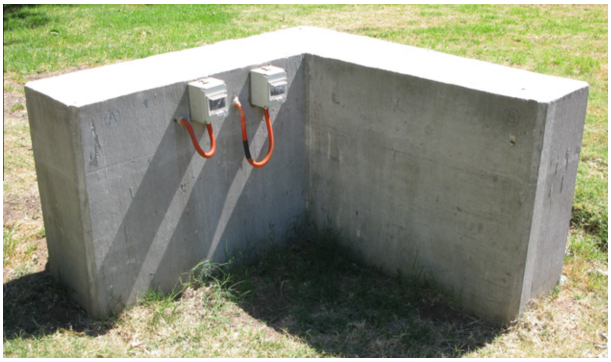

Fig. 3. Instrumented concrete wall simulating the corner of a cell showing the electrical contacts of the embedded sensor.

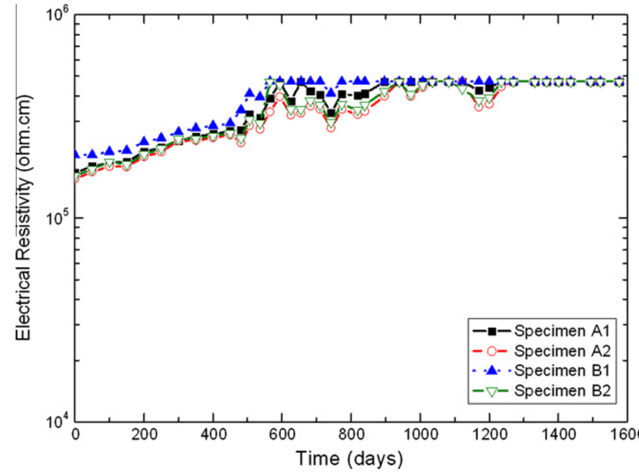

Fig. 4. Evolution of the electrical resistivity of the concrete a function of time, exposed to a laboratory environment.

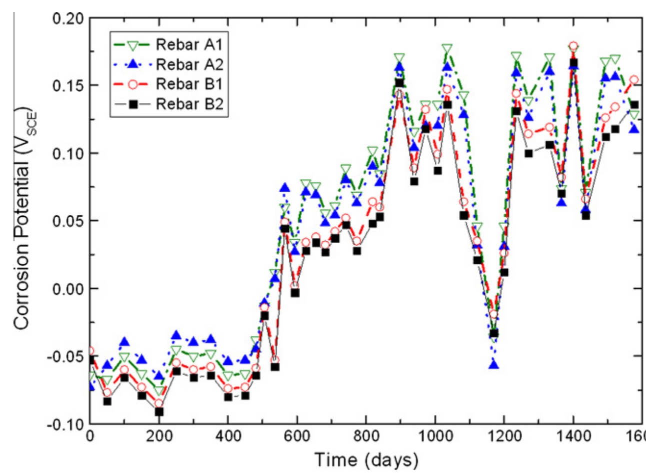

Fig. 5. Evolution of the corrosion potential as a function of time for steel bars embedded in concrete and exposed to the laboratory environment.

according to the Stern–Geary relationship as follows: $I_{corr} = B/R_p$, where B values should take into account whether the steel segments are in the active or passive state. Andrade et al. [7] reported typical values for steel embedded in mortar: the value of B for steel in the passive state ($E_{corr} > -0.2\ V_{SCE}$) is 0.052 V, while for steel in the active state ($E_{corr} < -0.3\ V_{SCE}$) is 0.026 V. Finally, the corrosion rate (*C.R.*) was calculated as $C.R. = I_{corr}$ (in μA/cm^2) × 11.6 [7].

The carbonation rate was determined after 3.4 and 5.7 years exposure using cylindrical concrete specimens (10 cm diameter and 10 cm height) by the phenolphthalein test. These specimens were exposed to laboratory environment, where the temperature was 24.4 ± 2.9 °C and the relative humidity was 50 ± 10%.

An "L shape" reinforced concrete wall (named "prototype") was built with the concrete previously described, using a pre-welded cage made with 10 mm diameter rebars (Fig. 3). The cover concrete of this prototype is 50 mm. This wall simulates the corner of the radioactive waste disposal cells, and was built on the Centro Atómico Constituyentes grounds, of the National Atomic Energy Commission, where the external temperature was measured on a periodic basis. The compressive strength of the concrete, measured at 28 days, was 52.1 ± 1.0 MPa.

Three corrosion sensors, specially developed in the laboratory [8], were embedded in the prototype in order to monitor the evolution of several parameters associated with the corrosion process along time: the temperature inside the structure, the corrosion potential and the corrosion rate of the reinforcing bars, the electrical resistivity of concrete and the availability of oxygen. The measurements started 28 days after the concrete was discharged into formwork. This monitoring has been followed for approximately 1200 days (about 3.3 years), and it will continue for the next years.

3. Results and discussion

Fig. 4 shows the evolution of the electrical resistivity of the concrete exposed to the laboratory environment. Two measurements were performed on each specimen (A and B), yielding a total of 4 data. It can be seen that the electrical resistivity continuously increases with the exposure time from 2×10^5 ohm cm, being this fact an indication of the continuous concrete hydration process. The apparent saturation of the resistivity value at about 5×10^5 ohm cm is due to the maximum value that can be measured with the available device.

Fig. 5 show the evolution of the corrosion potentials of the straight reinforcing bars as a function of time (up to 1600 days). It can be seen that the corrosion potential has a trend to increase along time and, in all cases, the value is more positive than −0.2 V_{SCE} that, according to ASTM C-876 standard [5], reveals a passive state of the steel.

The corrosion rate of the straight rebars embedded in concrete and exposed to the laboratory environment remained almost constant and close to 0.8 μm/year after 1600 days exposure (Fig. 6). Again, these results are a clear indication that steel presents a passive state.

As for the effect of welds on the reinforcing bars, it was found that no differences between the corrosion rate measured on welded and straight (un-welded) rebars were detected; so the presence of welds do not modify corrosion susceptibility of rebars.

The carbonation depth after 3.4 years exposure was 5.7 mm and after 5.7 years was 6.8 mm. These values give a carbonation rate (k in the equation $x = k \cdot t^{0.5}$, x being the carbonation depth and t the exposure time), of 3.09 mm years$^{-0.5}$ in the first case, and 2.75 mm years$^{-0.5}$ in the second one. Fig. 7 shows the values of the carbonation rate obtained in the present work compared with those obtained by Andrade et al. [9] for the same type of application. It can be seen that, up to the moment, the values obtained in the present case are higher than those measured in the "El Cabril" facility.

Fig. 8 shows the evolution of the temperature measured inside the prototype as a function of time. The figure also includes the evolution of the average external temperature measured in the same place where the prototype is installed. It can be seen that the internal temperature is always higher that the external one,

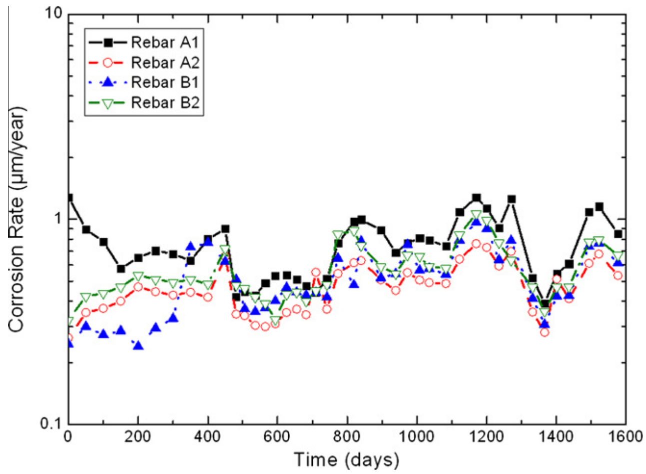

Fig. 6. Evolution of the corrosion rate as a function of time for steel bars embedded in concrete and exposed to a laboratory environment.

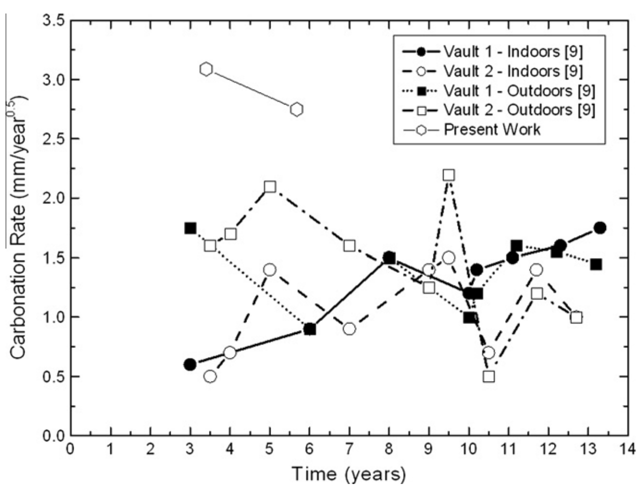

Fig. 7. Evolution with time of the carbonation rate calculated in the present work, compared with values obtained with control specimens in El Cabril [9].

being the difference higher during the summer periods, when the difference reaches almost 15 °C.

Fig. 9 shows the evolution of the concrete electrical resistivity as a function of time. The evolution of the internal temperature is also included (in this case, for the sake of simplicity, only the values provided by the sensor 1 are included). It should be noted that the progression of the hydration is well reflected by the increase in electrical resistivity. The impact of the cycle of temperature along the year on the electrical resistivity is remarkable: the higher the temperature, the lower the electrical resistivity. The same might happen with the other parameters measured so, care has to be taken when interpreting on-site results.

Fig. 10 shows the evolution of the corrosion potential of the reinforcing bars as a function of time (the temperature values provided by sensor 1 are also included). It can be seen that the corrosion potential is almost constant along the 3 year measurements period, with a small tendency to increase. In all cases, the value are close to −0.2 V_{SCE} that, according to ASTM C-876 [5] standard, is the borderline between intermediate and low corrosion risk.

Fig. 11 shows the evolution of the oxygen flow as a function of time. I can be seen a progressive decrease in the oxygen flow that reaches the rebars, without this being noticed by changes in the corrosion potential values. The trend of the oxygen flow follows

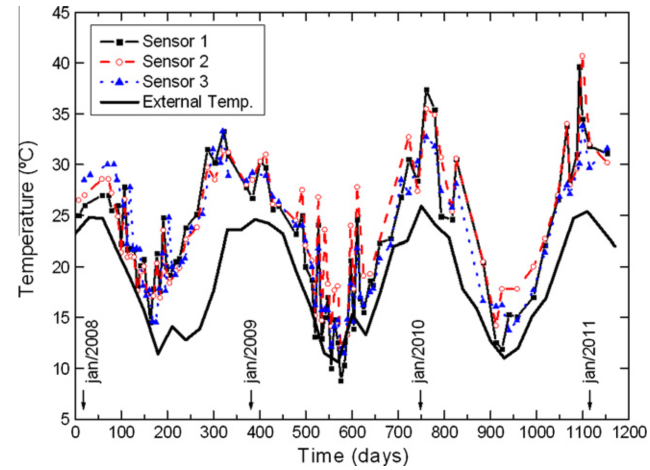

Fig. 8. Evolution of the temperature inside the prototype exposed to natural conditions, and the average external temperature as a function of time.

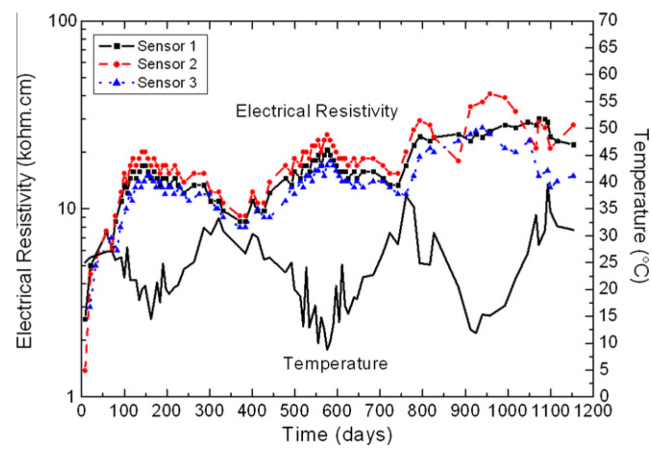

Fig. 9. Evolution of the electrical resistivity of the prototype exposed to natural conditions, and the internal temperature (sensor 1) as a function of time.

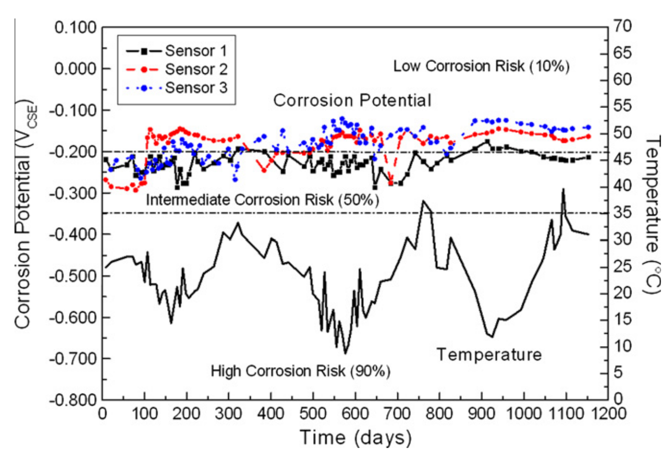

Fig. 10. Evolution of the rebar corrosion potential and the internal temperature (sensor 1) as a function of time, of the prototype exposed to natural conditions.

the trend of the internal temperature (the figure shows the values obtained with sensor 1): the higher the internal temperature, the higher the oxygen flow. Again, the impact of the cycle of temperature along the year on the measured parameter is noticeable.

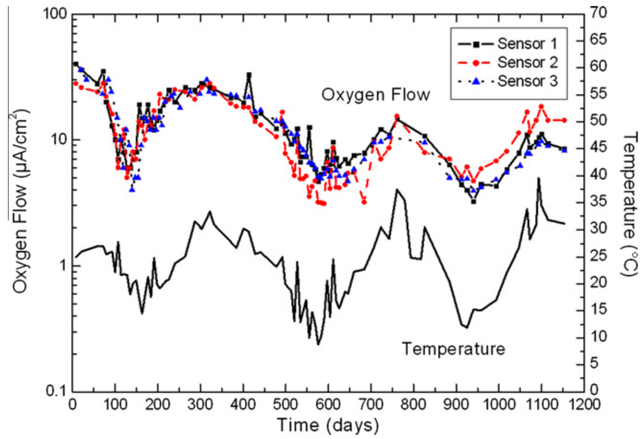

Fig. 11. Evolution of the oxygen flow and the internal temperature (sensor 1) as a function of time, of the prototype exposed to natural conditions.

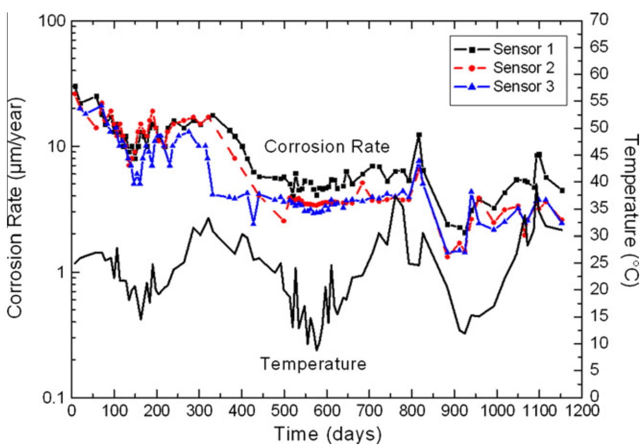

Fig. 12. Evolution of the corrosion rate and the internal temperature (sensor 1) as a function of time, of the prototype exposed to natural conditions.

Fig. 12 shows the evolution of the corrosion rate of the rebars (data obtained with the embedded sensors) as a function of time. The corrosion rate starts from 30 μm/year and, after 3 years, it reaches a value close to 2 μm/year. From this figure it is possible to deduce that the temperature evolution due to the seasonal changes is, again, the most influencing factor for the trends recorded. The reinforcement corrosion rate values are in the borderline between low and negligible corrosion rate [10].

According to the threshold corrosion potential and corrosion rate values for the passive to active transition for steel corrosion in concrete ($E_{corr} = -0.35\ V_{SCE}$ and $C.R. = 1$ μm/year), the results obtained in laboratory show that the steel reinforcement remains in the passive state or close to the boundary between low corrosion risk and intermediate corrosion risk. However, the comparison of the corrosion potentials and corrosion rates data obtained in laboratory and those obtained in the prototype seems to be not straightforward: the prototype shows lower corrosion potentials and higher corrosion rates than those obtained in laboratory specimens. The explanation for this phenomenon is that rebars embedded in the laboratory specimens were free of rusting, while the pre-welded cage of the prototype, after being exposed to the atmosphere for several days without any protection, underwent atmospheric corrosion and the steel were covered by a red type of corrosion product. These products were not properly eliminated before the cast of the concrete. It is a known fact that clean steel electrodes behave differently than pre-corroded ones. Mortar alkalinity by itself, or that provided by $Ca(OH)_2$ saturated solutions, which rapidly passivates a clean steel surface, does not ensure passivation of pre-rusted steel. In fact, rebars with pre-rusted surfaces exhibit unacceptable corrosion rates, even in non-carbonated concrete with a very low concentration of chloride [11–14]. The same results have been reported even in stainless steels, in which cheaper low-alloyed steels with bare surfaces are more resistant than expensive high-alloyed scaled steels [15]. One of the causes of this non-expected corrosion rate on pre-rusted surfaces might be the contamination of the rust layer during atmospheric exposure prior to the steel's embedding in concrete. However, the major cause might be the rust layer itself that acts as a barrier for alkaline solution from the concrete to reach the rebars. This fact is very interesting as in the actual construction practice pre-rusted steel bars are generally used.

Based on the Tuutí's service life model [16], no propagation period must be considered for ILRW disposal facilities because, according to the service criterion proposed by Andrade and Alonso [17], it is assumed that the durability of these structures ends once rebar corrosion has been initiated. In order to fulfil this criterion, the concrete must be designed to completely avoid corrosion during the period of almost 300 years. However, it should be mentioned that the effect of a passive state over 300 years deserves further study because, it is not known if a passive corrosion rate occurring during a very long time could lead to the formation of an oxide layer able to generate mechanical stresses in the concrete cover [18]. If corrosion (propagation period) is likely to occur during the ILRW disposal service life, the theoretical safety is very much reduced and a high risk of loosing the barrier properties exists before accomplishing the expected life. Two consequences of this statement are: ILRW facilities must not be located in places where chlorides, sulphates or the carbonation front could reach the rebar during the service life of the structure. In addition, all other degradation processes, which might affect cover integrity and impermeability (leaching, sulphate attack, freezing and thawing, etc), have to be avoided as part of the design phase.

The location selected for the construction of the ILRW disposal facility should be compatible with long-term concrete performance. That is to say, the site should be at a considerable distance from chloride containing environments (e.g. marine environments) and located in an area with no abnormal content of carbon dioxide and other acid contaminants. Then, the most important cause of reinforced corrosion is the carbonation during the period in which the vaults are kept in contact with air [16]. Knowing the diffusion rate of CO_2 in the concrete studied, after 300 years, the carbonation depth will be about 48 mm (assuming a constant value of the carbonation rate equal to that obtained after 5.7 years exposure). The design of the containers and cells foresee a concrete cover of 50 mm, that is to say, the concrete under study will fulfil the requirements for the expected life-time.

In conclusion, if the concrete is properly designed and carefully produced with good quality control forms, it is as an inherently durable material. However, several factors may affect the durability of concrete: constituent materials, construction practices, physical properties, environmental exposure conditions and types of loads. One of the most important factors is connected to the construction practices, i.e. to avoid the use of pre-rusted rebars. It is well recognised that concrete construction methods and practices influence the final quality of the concrete. Besides, the placement of concrete, that includes transporting the concrete to the jobsite delivery point, discharging into formwork, consolidating and providing proper curing conditions, will ensure adequate durability, serviceability and structural integrity in accordance to design specifications. It is worth to mention that, up to now, the site for the Argentine near-surface disposal facility has not been determined. So, as soon as this location is established, it will be necessary to fully characterise the environment for the correct identification of key parameters involved in the long term durability.

4. Conclusions

– The concrete studied in the present work in laboratory tests, shows an electrical resistivity that increases with time, which reflects the continuous hydration process. It provides a passive status of corrosion to steel rebars characterised by corrosion rates lower than 1 μm/year.
– The value of the carbonation rate is adequate to comply with the foreseen specifications.
– The corrosion rate as well as other parameters related to rebars durability measured in a prototype, shows values that are not in accordance with those obtained in laboratory specimens. This is due to the use of pre-rusted reinforced bars that increases the corrosion susceptibility of rebars. Besides, the impact of the cycle of temperature along the year on the parameter measured is remarkable so, this effect should be carefully taken into account when applying durability models.
– In order to fully evaluate the behaviour of reinforced concretes, once the selection of the site for the near surface disposal facility is made, the medium and long term evolution of the parameters under study should be followed up.

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