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Role of consolidants for limiting the radon exhalation rate in building materials of historical and artistic interest

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Abstract – **This study evaluates the effectiveness of consolidants in reducing radon (Rn-222) exhalation from building materials commonly found in historical structures. Radon, a radioactive gas derived from Ra-226 decay, represents a significant indoor health risk due to its tendency to accumulate. Experimental investigations were conducted within the ATHENA project, assessing radon emission from various lithotypes subjected to accelerated aging and treated with different consolidants, including PDMS, NanoEstel (silica-based materials), and NanoRestore calcium-based material). Radon exhalation rates were measured using a closed chamber method connected to a RAD7 detector, focusing particularly on Viterbo tuff. Results demonstrated an exponential increase in radon levels before reaching equilibrium, highlighting significant variations based on material composition, porosity, and applied consolidants. Notably, NanoEstel showed a remarkable reduction in radon emissions compared to untreated and other consolidant-treated samples. These findings underscore the importance of consolidants in heritage conservation, emphasizing their potential role in improving indoor air quality and safeguarding public health. This research was carried out as part of the PRIN 2022 PNRR ATHENA project, funded by the European Union through the Next Generation EU initiative.**

I. INTRODUCTION

Consolidants in building stones play a vital role in preserving the structural integrity and aesthetics of historic and modern stone structures by reducing the rate of decay and enhancing the stone's resistance to weathering [1-4]. They work by either binding the stone grains together or creating a barrier that protects the stone from moisture and other harmful environmental factors [3]. The main role of a consolidant is to reduce the rate of decay of the stone surface and the most successful treatments are those which least alter the characteristics of treated stone leaving it similar to the underlying sound stone [4].

The role of consolidants takes on extreme importance especially in those cases in which the emission of radioactive agents is involved, like radon (Rn-222).

This study, conducted within the ATHENA project, focuses on radon (Rn-222) exhalation from building materials commonly found in historical structures. Radon, a radioactive gas produced by the decay of Ra-226, poses a significant health risk due to its ability to accumulate indoors, making it essential to understand and mitigate its sources [6-8]. The research aims to systematically compare radon exhalation rates across different lithotypes and evaluate the impact of environmental factors such as humidity, temperature, and laboratory treatments, including accelerated aging and consolidant applications.

The role of consolidants is associated to the properties of aged rock samples which change their surfaces and porosity, sometimes favoring the emission of radon.

II. MATERIALS AND METHODS

The activity specifically investigates the radon exhalation rates of various stony materials, including Ignimbrite from Campania, Noto stone, Comiso stone, Lecce stone, Mendicino stone and Tuff from Viterbo all aged (salt weathering test) and consolidated with different consolidants (PDMS, NanoEstel, NanoRestore). Figure 1 displays an example of aged stones after several cycles of salt weathering test [5]. For each sample the radon exhalation rate was measured before and after the aging treatment.

Fig. 1. Stones (Viterbo tuff) before and after four cycles of salt weathering aging.

A. Consolidants

Characterized by its silicone-based polymeric structure, PDMS (Polydimethylsiloxane) is particularly effective in consolidating porous materials by penetrating and reinforcing their internal structure. This consolidant forms a protective barrier that enhances resistance against environmental deterioration factors such as moisture, pollutants, and weathering, without significantly compromising the original appearance of treated surfaces [1].

NanoEstel is an advanced consolidant formulated with silica nanoparticles, specifically designed for preserving and consolidating building materials of historical and artistic significance. Its nanoparticle composition facilitates deep penetration into porous substrates, effectively reinforcing structural integrity without significantly altering the aesthetic qualities or breathability of the material [2-3].

NanoRestore is an advanced consolidant specifically designed using nanotechnological solutions for the restoration and conservation of culturally significant carbonate-based materials such as frescoes and limestone. Its primary composition includes calcium hydroxide ($Ca(OH)_2$) nanoparticles dispersed in short-chain alcohols such as ethanol or 2-propanol. These nanoparticles penetrate deeply into the pores and fissures of degraded substrates, where they react with atmospheric carbon dioxide to form calcium carbonate ($CaCO_3$), effectively consolidating and stabilizing treated surfaces [2].

B. Radon exhalation rate measurements

The stone lithotypes were prepared by heating them to 110°C for one hour to assess the impact of humidity on radon release rates. This preparation step is essential to reduce moisture content, which can influence radon emission [6]. Subsequently, the samples were cut into cubes of approximately 5 cm per side, as a regular shape facilitates accurate calculation of the radon exhalation surface area. Prepared samples were then placed in an accumulation chamber connected to a Durridge-RAD7 radon detector. The chamber's air was dried using a desiccant and a DRYSTIK system (volume 1l), ensuring minimal humidity and forming a closed loop with the RAD7 to measure radon levels effectively (figure 2) [7].

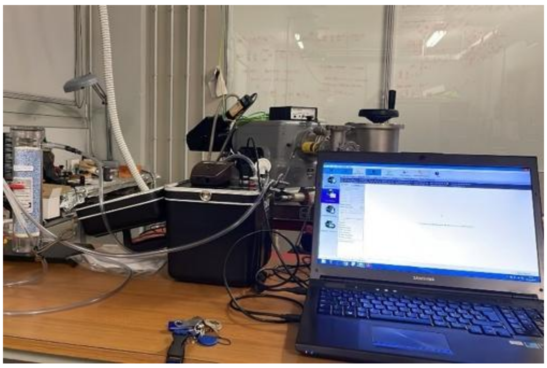

Fig. 2. Experimental setup with Durridge-RAD7 radon detector.

The RAD7 detector utilized the ^{218}Po peak to calculate radon concentrations, leveraging the rapid equilibrium between ^{218}Po and ^{222}Rn, achieved in approximately 15 minutes. The radon growth curve to equilibrium was monitored over a period of 10 days and the ^{222}Rn specific exhalation rate, E (Bq h^{-1} kg^{-1}), was calculated according to the following equation:

$$E = \frac{(C - C_0 e^{-\lambda T})/m}{1 - e^{-\lambda T}} \lambda V$$

where C is the equilibrium concentration (Bq m^{-3}), C_0 is the initial radon concentration (Bq m^{-3}), λ (h^{-1}) is the sum of the radon decay constant, the bound exhalation constant and the leakage constant, V is the total volume of the analytical system (m^3), T is time of exposure (h) and m is the mass of the sample (kg) [8-14]. The calibration accuracy is independently verified by direct determination of the radon chamber level from the calibrated activity and emission of standard radon source. In addition, periodically intercomparison with other radon chambers is performed periodically. Generally, a reproducibility of better than 2% with the standard RAD7 calibration is achieved. Overall calibration accuracy is in the range of 5%, ensuring a Measurement Accuracy of +/-5% (absolute), considering the range of 0% - 100% of RH [7].

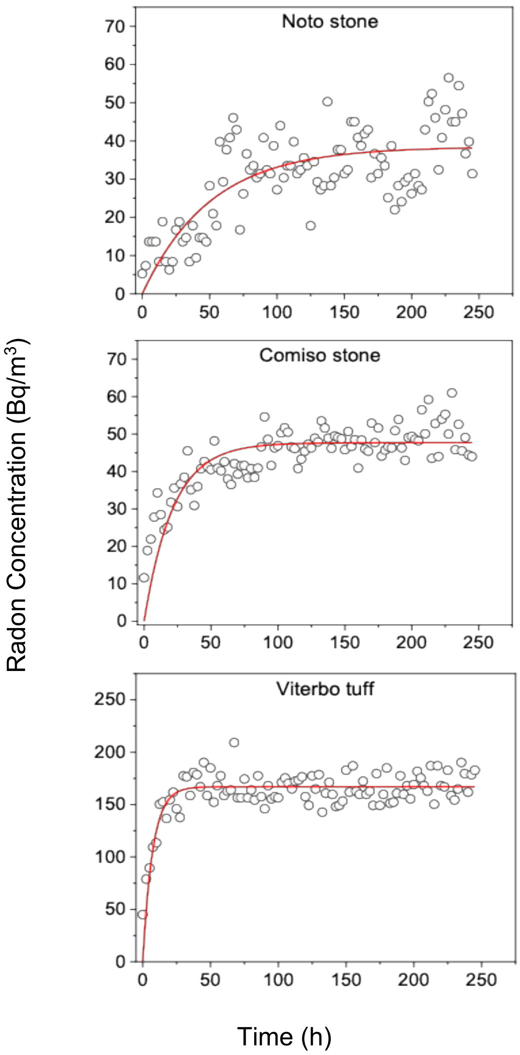

Fig. 3. Radon activity concentrations during the accumulation (dots) and fit curves for evaluating the radon exhalation rate (red line).

III. RESULTS

Preliminary experimental data and fitted curves are shown in the following graphs. Figure 3 proposes the Radon concentration for three different exemplificative samples: Noto stone, Comiso stone and Viterbo tuff.

Experimental results clearly indicate that Viterbo tuff exhibits the highest radon concentration at saturation compared to all other studied stone materials. Following these significant findings, we conducted a preliminary investigation focusing on the radon exhalation rates from artificially aged samples of Viterbo tuff. These samples were systematically compared with untreated samples and those treated using three distinct consolidants. Results revealed that the artificially aged sample displayed a slightly reduced radon exhalation rate compared to the untreated sample. Figure 4 reports the trend of radon accumulation in the RAD7 chamber as a function of the time.

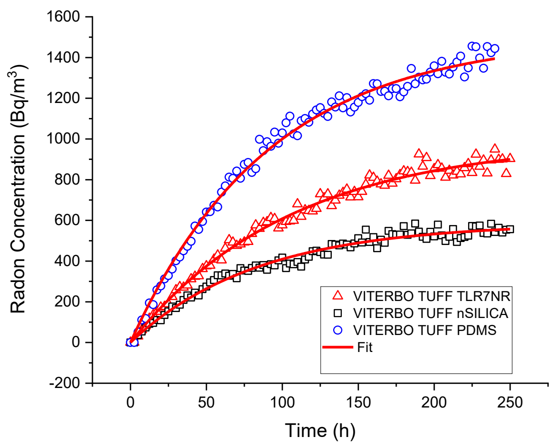

Fig. 4. Radon activity concentrations during the accumulation (dots) for different consolidant applied to Viterbo tuff. Fit curves for evaluating the radon exhalation rate are reported in red line.

The curves express clearly the role of the different consolidants used in the study: nSilica (NanoEstel) reduces drastically the radon exhalation rate in relation to what obtained with the other applied consolidants. NanoEstel decreases about three time the radon emission if compared, for example, to PDMS which leaves the rate quite unaltered. Calculated values of radon activity concentrations and radon exhalation rates are summarized in Table 1.

Table 1. experimental results for Viterbo tuff with different consolidants.

ID	Weight after heating (kg)	Radon activity concentration (Bq m^{-3})	λ (h^{-1})	Radon exhalation rate (Bq kg^{-1}h^{-1})
Viterbo Tuff	0.160	170 ± 20	0.024 ± 0.002	0.755 ± 0.263
Viterbo Tuff (Aged)	0.138	32 ± 0.7	0.07 ± 0.02	0.600 ± 0.050
Viterbo Tuff (PDMS)	0.163	1503 ± 15	0.0109 ± 0.0002	0.574 ± 0.023
Viterbo Tuff (NanoEstel)	0.151	586 ± 7	0.0120 ± 0.0004	0.247 ± 0.019
Viterbo Tuff (NanoRestore)	0.152	993± 12	0.0095 ± 0.0003	0.370 ± 0.025

IV. CONCLUSIONS

The experimental findings of this research clearly demonstrate the effectiveness of consolidants in significantly modifying the radon exhalation behavior of historical building materials, notably in the case of Viterbo tuff. Among the consolidants tested, NanoEstel exhibited the most substantial reduction in radon emission rates, decreasing radon release by approximately threefold compared to untreated and other treated samples. Conversely, PDMS showed minimal influence, underscoring that consolidant composition plays a critical role in radon mitigation.

These outcomes highlight the potential of nano-silica-based consolidants as a strategic intervention in the conservation of culturally significant structures, effectively enhancing both their structural integrity and indoor air quality. The results emphasize the necessity of carefully selecting consolidants based on their specific impact on radon dynamics, advocating for integrated conservation practices that address both structural preservation and occupant health protection. Future work should focus on long-term monitoring and comprehensive field evaluations to validate laboratory results under real-world conditions.

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