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An optimized radiochemical method for the quantification of ^{36}Cl in irradiated concrete from nuclear decommissioning

Andrea Broglia[1] · Gabriele Magugliani[1] · Luca Fornara[2] · Cristiano Piras[3] · Gianmarco Bilancia[2] · Francesco Galluccio[1] · Elena Macerata[1] · Mario Mariani[1] · Eros Mossini[1]

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Abstract
The radiochemical methods for the characterization of hard-to-measure radionuclides are often complex, laborious and expensive, especially in the case of solid samples. The present work aimed at developing a new optimized method based on Cherenkov counting for the determination of ^{36}Cl in activated concrete. The primary goal consisted in improving simplicity and cost-effectiveness with respect to currently available methods without sacrificing performance. A chemical recovery of ≈ 90% and high decontamination from interferents were demonstrated. The method also proved to be scalable to large sample masses for clearance purposes, thereby achieving a detection limit < 5 mBq g^{-1}.

Keywords ^{36}Cl · Activated concrete · Cherenkov liquid scintillation counting · Nuclear decommissioning · Radiochemical procedure · Radioactive waste characterization

Introduction

From the beginning of the operations of a nuclear facility to its decommissioning and to the final disposal of wastes produced, the characterization of all materials involved is an activity of extreme importance. In particular, the classification of waste through radiological characterization is fundamental to design its correct and optimized route towards its final disposal: an inappropriate classification may result in either a hazard for the public or a waste of valuable resources [1]. In the radiological characterization of materials, one challenge is often posed by the possible presence of radionuclides which are called hard-to-measure (HTM) due to their radiation being poorly penetrating [2]. Therefore, the radiation emitted by said radionuclides is self-absorbed in the source item and their quantification may require ad hoc procedures. The radiochemical and radioanalytical methods necessary for the quantification of HTM radionuclides are often costly, time-intensive, complex to implement, and hence unsuited for routine characterization activities [3]. The scaling factors approach is therefore commonly adopted to deduce activity concentrations, but the identification and demonstration of suitable correlations between key radionuclides and HTM nonetheless requires dedicated matrix-specific sampling and analysis campaigns [2]. For this reason, the need for optimized, agile and cost-effective radiochemical methods is still of paramount importance.

^{36}Cl is a pure β-emitting HTM radionuclide with a half-life of 3.01×10^5 years. Such a long half-life, together with its volatility and mobility in solid matrices, makes it a challenge for long term waste disposal [4]. ^{36}Cl may be found in radioactive waste due to the neutron activation of stable ^{35}Cl. The quantification of ^{36}Cl may be difficult not only because it requires a complete purification from interferents, i.e. any other beta emitter, but even due to the complexity and variety of the matrices that contain it, ranging from aqueous effluents, concentrates, sludges, graphite and, most notably, irradiated concrete [5]. In nuclear facilities this material is extensively employed, also in places where it can experience intense neutron fluxes (e.g. biological shields). Since a non-negligible amount of stable chlorine may be found in concrete (between 250 and 1250 ppm [6]), at the decommissioning stage it can be expected to find a significant amount

✉ Gabriele Magugliani
gabriele.magugliani@polimi.it

[1] Department of Energy, Nuclear Engineering Division, Politecnico di Milano, Milan, Italy

[2] Joint Research Centre, European Commission, Ispra, Varese, Italy

[3] Nucleco S.p.A., Ispra Site, Rome, Italy

of ^{36}Cl-activated concrete waste, with activity levels ranging from tens of Bq g^{-1} down to clearance levels [7, 8]. The large inventory of this material entails high management costs if not properly classified or cleared from regulatory control. To conduct a representative and comprehensive characterization campaign, the costs associated with the thorough sampling operations can be extensive. Hence, the need for a method for the accurate quantification of ^{36}Cl in irradiated concrete arises. Moreover, since it is already expected that the characterization of this type of waste material will be expensive due to its vast volume and its complexity, such a method must be optimized to reduce the associated costs by lowering the time-intensiveness and laboriousness of the analysis [9].

To develop an improved method, the starting point was a gap analysis of the state of the art for the quantification of ^{36}Cl, aiming at identifying the strengths of preexisting methods as much as their flaws [4, 7, 10–13]. Particular attention was given to previous works by Hou et al. and Ashton et al., as they investigated similar waste samples. They achieved remarkable performances in terms of accuracy and precision, with almost quantitative chemical yields and detection limits of around 10 mBq g^{-1} [8, 14]. However, these procedures are quite laborious and involve energy- and material-intensive sample treatments, such as alkali fusion and analyte volatilization, and several sequential steps aimed at achieving the required decontamination from interferents. Simplification of state-of-the art procedures applicable to complex sample matrices such as irradiated concrete is therefore a crucial step in allowing the safe and effective implementation of radiological characterization campaigns.

The present work aims to develop a simpler radioanalytical method for the determination of ^{36}Cl in concrete, without sacrificing accuracy and sensitivity. A key distinguishing feature of the procedure is the reliance on Cherenkov counting, thus avoiding the use of liquid scintillation cocktails, negating metrological challenges related to chemical quenching and luminescence [15], and greatly simplifying the requirements in terms of radiochemical purity: since Cherenkov emission is a threshold effect, this counting method is naturally able to discriminate all beta/gamma emitters with energy < 250 keV. Also, counting efficiency steeply decreases as the involved energy falls below 500 keV [16], in contrast to cocktail-based LSC where counting efficiencies are essentially unitary for energies above 100 keV. This helps in attenuating—or removing altogether—the bias induced by some low-energy interfering emitters without requiring their quantitative removal, hence greatly alleviating the requirements imposed on radiochemical manipulations.

This work was developed in the framework of Work Package 5—ICARUS ("Innovative characterisation techniques for large volumes") of EURAD-2 (European Joint Programme on Radioactive Waste Management) [17, 18].

Materials and methods

Materials

All reagents employed throughout the procedure were of analytical grade, and were used without further purification. Two types of concrete samples were prepared, as detailed in the following: one set was spiked with a known amount of stable Cl, and the other one was not.

In particular, spiked concrete samples were prepared to assess Cl leaching by embedding the analyte directly into the sample matrix, thus allowing the optimization of the sample treatment step under realistic conditions (see Table 1). CEM I 42.5 R, sand (0–4 mm), and Cl-free osmotic water were employed to prepare a representative concrete formulation (water to cement ratio 0.36, aggregates 40 wt.%) [19]. A known amount of Cl was added in the form of NaCl to the water used to prepare the paste. The amount of Cl initially present in the raw materials, i.e. sand and CEM I, was quantified via X-Ray fluorescence analysis and was determined to be < 1 wt.% relative to the added quantity, and was therefore considered negligible. Hence, the concentration of Cl in the produced specimen amounted to 1.4 wt.%. The paste was then poured into a mold and cured for 28 days in a climatic chamber (at 20 ± 2 °C with relative humidity > 90%).

The second set of samples was prepared in an identical manner, but with no added Cl. These were employed as blank/surrogate samples once the optimal leaching conditions were identified.

As a pre-treatment step, all concrete samples were ground to a particle size of < 2 mm and homogenized to ensure representativeness.

Overview of the method

The proposed method is represented in Fig. 1. It consists of four main steps: analyte extraction from the concrete matrix by leaching with a 1 M sodium hydroxide solution; AgCl precipitation; re-dissolution of the precipitate in aqueous ammonium hydroxide; Cherenkov counting. The radiochemical yield of the procedure, or of one of its sub-steps, was determined by ion chromatography (IC).

Leaching

Approximately 1 g of concrete powder is added to a beaker and contacted with 20 mL of 1 M NaOH solution under stirring at 70 °C.

During the study and optimization of the leaching performance, no external Cl carrier was added to the sample, as the assessment of the yield relied on its known content in the Cl-spiked concrete. This approach was adopted to more

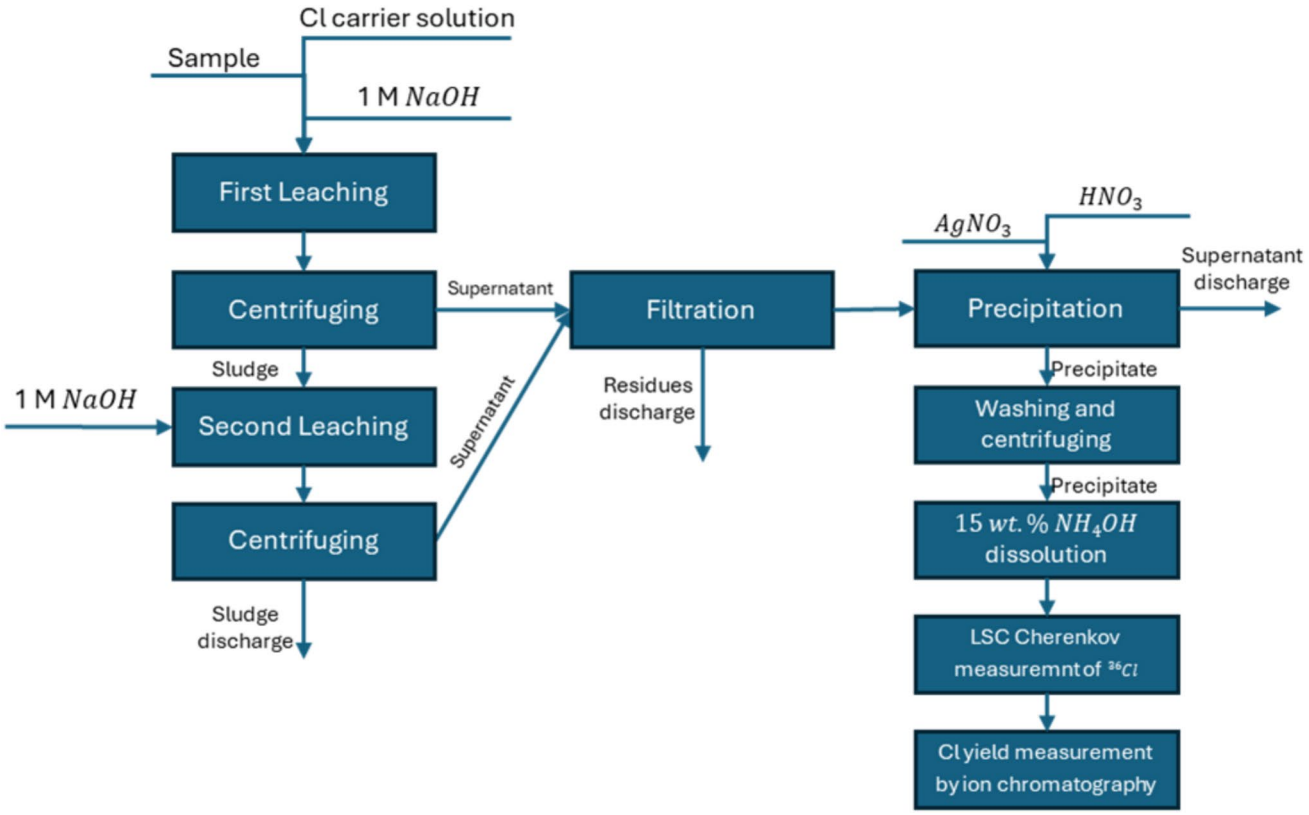

Fig. 1 Outline of the proposed method. During the study of leaching conditions performed with the Cl-spiked concrete sample, no Cl carrier solution was added

realistically evaluate the extraction of the analyte when embedded in the sample matrix, as this situation is more conservative than simply evaluating recovery of a liquid carrier spike added after grinding.

In all other cases, 25 mg of Cl carrier (as aqueous NaCl solution) were added to the powder. This quantity was selected to be much larger than that naturally present in the concrete sample.

Then, the supernatant is separated from the solid by centrifugation at 2000 rpm. Different leaching times (1–2 h) and numbers of sequential leaching cycles (single vs multiple) have been tested. The second and third leaching steps are performed with 10 mL of 1 M NaOH solution under the same experimental conditions. All the resulting supernatants are then combined in a single container. If necessary, the supernatant collected up to this point can be filtered through a 0.2 µm syringe filter.

The Cl leaching yield is determined on duly diluted aliquots of the leachates by IC as:

$$\eta_{Cl} = \frac{[Cl]_{leachate} m_{leachate}}{[Cl]_{sample} m_{sample}}, \quad (1)$$

where $[Cl]_{leachate}$ is the measured chloride concentration in the leachate ($g_{Cl}/g_{leachate}$), $[Cl]_{sample}$ is the chloride concentration in the concrete sample (g_{Cl}/g_{sample}), $m_{leachate}$ is the mass of the leachate and m_{sample} is the mass of the concrete sample.

Selective Cl precipitation and re-dissolution

The leachate is brought to a pH between 4 and 8 with HNO_3, then a suitable amount of silver nitrate solution is added, such that the silver amount stoichiometrically matches the amount of Cl expected assuming complete recovery, plus a 10% excess. The sample is gently mixed and allowed to sit for 10 min. This ensures quantitative analyte precipitation as AgCl.[1] The supernatant is removed from the precipitate by centrifugation at 2000 rpm for 10 min and discarded. The precipitate is rinsed once with 20 mL of ultrapure water, and the supernatant is again removed by centrifugation and discarded. The washed precipitate is dissolved with a suitable amount of 15 wt.% NH_4OH. The Cl precipitation and re-dissolution yield is obtained by IC measures of duly diluted aliquots of the corresponding solutions and supernatants.

[1] If the sample is left to sit for more than one hour, it is advisable to shield it from external light, as this can reduce Ag^+ to metallic silver, lowering the yield of the method.

The overall analyte recovery of the procedure is determined as the product of the leaching and precipitation/re-dissolution yields or, in the real case, as the ratio between Cl mass recovered in the counting solution and the added carrier amount.

Decontamination from interferents

To assess the effectiveness of the method in removing interferents, 1 g of blank concrete was traced with a solution containing known amounts of Co, Ba, ^{152}Eu, Sr, Cs and ^{226}Ra; a detailed discussion on the reason behind this choice of elements is provided in Sect. "Method robustness". The slurry was homogenized and dried at 105 °C overnight. The traced concrete was then subjected to the analytical procedure described earlier. Recovered amounts of Co, Ba, Sr and Cs were determined via ICP-MS analyses on the 1 M NaOH leachate and on the 15 wt.% NH_4OH solution. ^{152}Eu and ^{226}Ra were instead monitored at the same procedural steps via gamma and alpha spectrometry, respectively. For all elements except Cs, an additional known quantity of each interferent was added to the leachate before inducing AgCl precipitation, since the amount remaining in solution was very low due to high decontamination achieved by the leaching step.

^{36}Cl Cherenkov counting

Finally, the analyte solution is transferred to a polyethylene scintillation vial and measured by LSC in Cherenkov mode. Since the concluding step of the procedure, i.e. re-dissolution of the AgCl precipitate in 15 wt.% NH_4OH, can be performed in a variable amount of solvent, three sample geometries, i.e. 5, 10 and 20 mL, were considered for the optimization of counting conditions, with constant counting time of 4 h. Blank samples were either prepared with pure 15 wt.% NH_4OH solutions (instrument blanks), or by subjecting non-irradiated concrete to the whole procedure (sample blanks). Optimal counting window and resulting detection efficiency were determined by measuring ^{36}Cl matrix-matched calibration standards.

Instrumentation

Ion chromatography was employed to quantify Cl^- concentration through the various steps of the procedure. Analyses were performed with a Dionex ICS 1100 equipped with a RFC-30 KOH eluent generator, ion suppression and electrical conductivity detector. Calibration was performed with certified solutions in the Cl^- concentration range between 1 and 100 mg L^{-1}. Prior to analysis, samples were diluted with ultrapure water to a suitable concentration.

Elemental analyses were performed with a NexION 2000 ICP-MS (PerkinElmer). The instrument was calibrated with multielemental certified reference solutions, diluted with ultrapure 1 wt.% nitric acid to the concentration range 0.1—100 µg L^{-1}. Prior to analysis, samples were diluted with 1 wt.% ultrapure nitric acid in order to achieve analytes concentration compatible to the calibration range.

A Hidex 300 SL Super Low Level with cooling set at 16 °C was employed to perform liquid scintillation counting in Cherenkov mode. Gamma spectrometry (for samples traced with ^{152}Eu) was performed with a 2″ NaI(Tl) scintillator (Silena) coupled with a model 926 multichannel buffer (Ortec). The detector was calibrated with certified multielemental solutions, adopting the same geometry (10 mL test tube) as for actual samples. An Octete plus (Ortec) spectrometry system was employed to perform alpha activity measurements for samples traced with ^{226}Ra. Counting samples were prepared by evaporation of 200 µL aliquots on stainless steel planchets. Detection efficiencies were determined via the standard addition method.

Results and discussion

Leaching

The Cl leaching yields (η_{Cl}) performed on Cl-spiked concrete obtained by varying the number and duration of the leaching steps are reported in Table 1. No direct comparison of these values with literature can be carried out, as the specific analyte recovery of the leaching step alone was not assessed by other studies through spiked samples. As shown, the performance of the leaching remains very high, with only marginal improvements in the case of multiple leaching steps. Moreover, the duration of each leaching step appeared to play a minor role, suggesting the possibility for optimization to reduce the time intensiveness of the method while still keeping high Cl recoveries. Therefore, two leaching steps of 1 h were deemed to be the optimal choice in terms of Cl yield and time-intensiveness.

Table 1 Cl leaching yields obtained by varying the number and duration of the leaching steps in 1 M NaOH performed on Cl-spiked concrete

Number of leaching steps	Duration of each leaching step (h)	η_{Cl} (%)
1	2	88.5 ± 3.7
2	2	94.1 ± 3.9
3	2	95.3 ± 6.0
2	1	93.3 ± 5.0

Cl precipitation and re-dissolution

The precipitation step was studied at two conditions: in 1 M NaOH, i.e. the supernatant produced by the leaching step, and at a pH between 4 and 8, obtained by adding a suitable amount of HNO_3 to the basic leachant of the first step. In both cases, 25 mg of stable Cl were initially present in solution. Cl recovery yield of the selective precipitation and re-dissolution steps was $\eta = 92.5 \pm 4.3\%$ for the neutralized sample and $\eta = 50.5 \pm 2.9\%$ for the 1 M NaOH solution. As expected, the chemical yield is affected by the basicity of the solution which, if not corrected, can result in a decreased amount of AgCl precipitate in favor of silver oxide [20]. Hence, neutralization of the leachate was adopted.

Optimization of counting conditions

The spectrum of an instrument blank sample and of a ^{36}Cl standard solution are reported in Fig. 2. The spectrum of a sample blank (deriving from the treatment of 1 g of blank concrete) is not reported in the figure for clarity, as it was essentially identical to that of the instrument blank (2.9 ± 0.1 cpm and 3.1 ± 0.1 cpm for instrument blank and sample blank respectively, under the same counting conditions; see also Table 4).

This fact indicates that matrix constituents do not play a role in the observed blank count rate. Hence, sample blanks can be likely substituted by instrument blanks when dealing with real samples, greatly simplifying the procedure and decreasing its time intensiveness.

As expected for Cherenkov emission, the ^{36}Cl spectrum is strongly peaked at low channel numbers. To define the optimal counting region, the upper limit of the counting window was varied in the channel range 75–200, while the lower limit was set to channel 1. Corresponding values of the detection limit (DL), calculated according to [21], and determined for various sample volumes, are reported in Fig. 3.

It appears that the optimal window spans between channels 1 and 125, irrespective of sample geometry, thus indicating good robustness of the chosen counting conditions. A summary of figures of merit for the optimal counting window is reported in Table 2. Chemical yield of 90% and counting time of 4 h are assumed in the calculations to provide a more representative description of the method performance.

Despite the high chemical recovery, the obtained DL values are quite high if compared with other literature methods ($\approx$ 10 mBq g^{-1} [8, 14]) as a consequence of the low detection efficiency of Cherenkov counting. However, as will be further stressed in the next section, this type of radiometric measure provides for an additional removal of interferences caused by low-energy emitters, thus greatly simplifying the radiochemical procedure. Lower DL may be achieved by longer measuring times or, as will be described in the following, by treating a larger sample mass.

Finally, to preliminarily validate the whole procedure, three blank concrete samples were spiked with a known amount of ^{36}Cl (approximately 1 Bq) and with 25 mg of stable Cl carrier. The leaching was performed as optimized above; the leachates were then filtered and neutralized with concentrated nitric acid, the precipitation was triggered

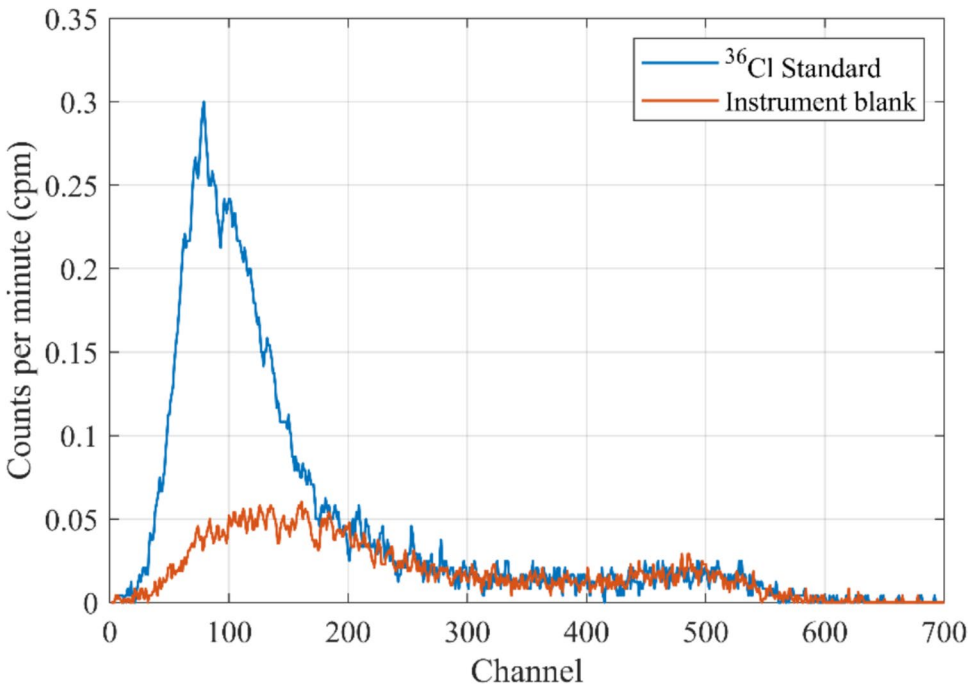

Fig. 2 LSC Cherenkov spectra of an instrument blank solution and of a ^{36}Cl standard. Sample volume 10 mL

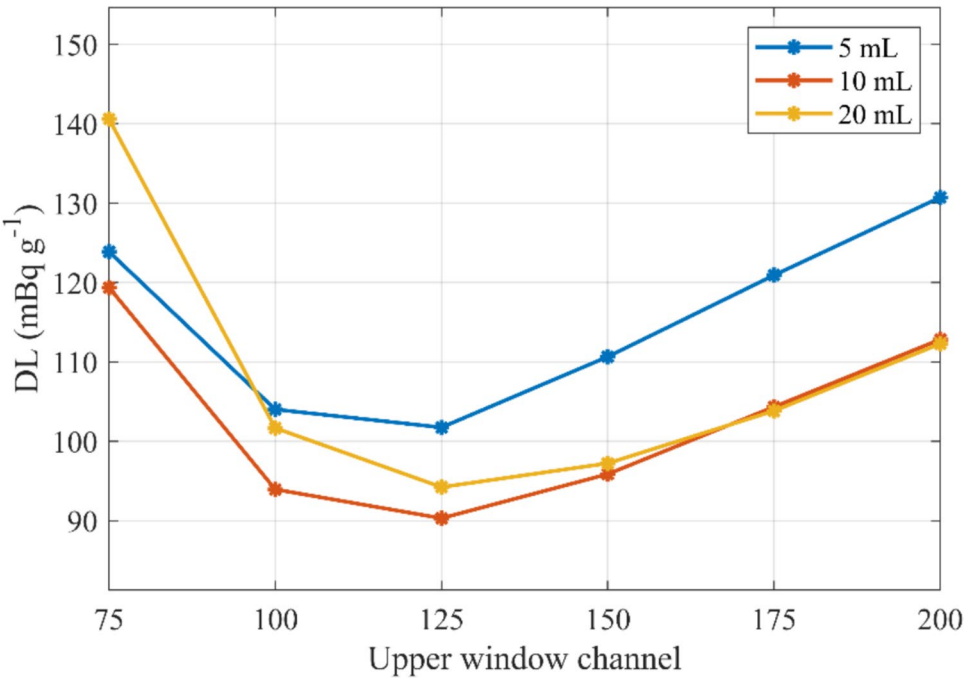

Fig. 3 Detection limit corresponding to various sample volumes and upper limit of the counting window, under constant counting time of 4 h

Table 2 Blank count rate, detection efficiency and characteristic limits for various sample geometries (sample mass 1 g, η_{Cl} 90%, channels 1 – 125)

Sample volume	Blank count rate (cpm)	Efficiency	DL (mBq g^{-1})
5 ml	2.6 ± 0.1	9.2 ± 0.2%	113
10 ml	2.9 ± 0.1	9.6 ± 0.2%	100
20 ml	3.2 ± 0.1	9.8 ± 0.2%	105

and then the precipitates dissolved with 10 mL of 15 wt.% NH_4OH and measured by LSC in Cherenkov mode in the aforementioned optimized counting conditions. The chemical yields for each sample were measured via IC. The average accuracy of the method was determined at 93 ± 5%. To further validate the method in a broader range of analyte concentrations, and to test its reproducibility, an interlaboratory comparison exercise is being planned within the scopes of the EURAD-2 ICARUS Work Package.

Method robustness

The method essentially comprises four purification steps, of which three are *chemical*, and one is *physical*. In particular, interferents are removed when:

1. they are not extracted from the concrete in the alkaline leaching environment;
2. they are not precipitated by AgCl;
3. once precipitated, they are not re-dissolved in 15 wt.% NH_4OH;
4. they do not entail the emission of Cherenkov light.

The fourth aspect represents a physical threshold, which can essentially prohibit the detection of an interferent providing its categorical discrimination. One prime example is iodine: since it has a chemical behavior which is very similar to chlorine, it is known to precipitate as AgI together with AgCl, and is one of the major elements of concern for radiochemical purification of Cl. However, the only I isotope with relevance in decommissioning (^{129}I, $\beta_{max} = 149$ keV, $t_{1/2} = 1.5 \times 10^7$ y) produces no Cherenkov emission and therefore it does not interfere with the measurement of ^{36}Cl, hence its decontamination was not assessed numerically.

Decontamination factors (DFs) were calculated as the mass (or activity) ratio of an interferent before and after a separation step. The decontamination effectiveness of the procedure was studied sequentially for the leaching and for the combination of AgCl precipitation and re-dissolution steps. Keeping in mind the physical consideration about Cherenkov threshold described at the beginning of this section (which, for example, negates the relevance of ^{129}I, ^{55}Fe, ^{63}Ni), the main interferents considered were Co, Ba, Eu, Sr, Cs and Ra. Among these, the first three can be present as activation products (^{60}Co, ^{133}Ba, ^{152}Eu) in the bulk of neutron-exposed structures, and are therefore the main radionuclides to be considered. Sr and Cs (^{90}Sr and ^{137}Cs isotopes) are fission products, and their presence is expected only on surfaces in case of contamination [22, 23]. Finally, decontamination from Ra was assessed since it is a naturally occurring radionuclide, and some of its decay products, in particular Bi isotopes, are Cherenkov positive.

Due to their short half-life (< 20 min) compared to typical sample processing times (few hours) they are not a problem per se. Indeed, they can induce a significant measurement bias only if their first long half-life precursors, i.e. $^{226/224}$Ra, also follow Cl in the procedure. Hence, the decontamination of Ra was quantified as the obtained results can be immediately transposed to Bi. Obtained decontamination factors are reported in Table 3.

Very high decontamination factors—higher than 10^6—were achieved for all interferents. Hence, the proposed method proved to be robust towards the main interfering radionuclides. Only in the case of Cs, a more modest DF of 3.8×10^3 was recorded. Despite being lower than corresponding values reported in literature, this deserves some quantitative contextualization. For Cherenkov counting, detection efficiency of ^{137}Cs is approximately half of that of ^{36}Cl (5.6 ± 0.2%, 10 mL sample, ch. 1–125 counting window). If a maximum acceptable interference of 5% from ^{137}Cs with respect to the ^{36}Cl DL is considered (i.e. ≈ 5 mBq g^{-1}), by applying the reported DF and counting efficiency this corresponds to a maximum acceptable concentration of ^{137}Cs in the original sample equal to ≈ 30 Bq g^{-1}. This essentially defines the upper applicability limit of the current method as-is. Being ^{137}Cs a fission product, its presence in concrete is a consequence of surface contamination, and its concentration in bulk samples is expected to be minimal. Hence, the applicability range of the method is expected to be fully compatible with the great majority of actual samples; to verify such compliance, ^{137}Cs activity can be—and routinely is—easily determined by gamma spectrometry as part of a comprehensive radiological characterization. If the aforementioned activity level is exceeded, two approaches can be adopted:

- the solution obtained from the AgCl re-dissolution can be subjected to a chromatography step as described by Hou [8], since at this stage sample conditions and matrix composition are identical. DF > 10^6 can be reached in this case;
- subtraction of ^{137}Cs contribution from the LSC count rate after having assessed its activity by gamma spectrometry and its Cherenkov detection efficiency.

Table 3 Partial and total decontamination factors for the considered interferents

DF	Leaching	Precipitation + Re-dissolution	Total
^{152}Eu	$> 2.3 \times 10^{3*}$	$> 3.0 \times 10^{3*}$	$> 6.9 \times 10^6$
^{226}Ra	6.8 (2)	$> 5.2 \times 10^{6*}$	$> 3.5 \times 10^7$
Co	$2.72\ (8) \times 10^4$	$6.3\ (2) \times 10^2$	$1.71\ (7) \times 10^7$
Cs	11.8 (3)	$3.26\ (9) \times 10^2$	$3.8\ (1) \times 10^3$
Sr	$4.4\ (1) \times 10^2$	$2.21\ (7) \times 10^4$	$9.7\ (3) \times 10^6$
Ba	$2.93\ (9) \times 10^4$	$4.62\ (1) \times 10^4$	$1.35\ (4) \times 10^9$

*Analyte after decontamination step was below detection limit. N.A.: not assessed

Scalability of the method

The mild leaching conditions, without complete matrix dissolution, the effectiveness in interferent removal, and the absence of resin columns which can impose limits on sample loading, all support the scalability to sample masses at least one order of magnitude greater than 1 g.

To verify the stability of the figures of merit, the method was applied to a blank 30 g concrete sample. Results for an instrument blank solution, and for two sample masses are reported in Table 4. Blank count rates and detection efficiencies (determined by direct addition of ^{36}Cl tracer to the corresponding solutions) all appeared comparable to a pure 15 wt.% NH_4OH instrument blank, thus indicating robustness of the method towards large sample masses. As a result, the DL of the method could be reduced to less than 5 mBq g^{-1}, which might be particularly appealing for materials clearance purposes. The use of a larger mass can also ensure a more representative sampling.

For this larger sample mass, the applicability of the method in terms of maximum ^{137}Cs activity resulting in a 5% bias when ^{36}Cl is at the DL corresponds to ≈ 1 Bq g^{-1}. Despite being lower than the 1 g sample result, this can still be considered reasonable when the scope of analysis is to assess clearance. As an example, Italy adopts clearance levels of 0.1 Bq g^{-1} and 1 Bq g^{-1} for ^{137}Cs and ^{36}Cl, respectively [24]. Therefore, pursuing a DL of < 5 mBq g^{-1} for ^{36}Cl would be uncommon, when the material is already exceeding its clearance limit for ^{137}Cs.

Table 4 Typical blank count rates, detection efficiencies and corresponding DLs obtained for various sample masses

Sample type	Blank count rate (cpm)	Efficiency	Yield (typical)	DL (mBq g^{-1})
15 wt.% NH_4OH (instrument blank)	2.9 ± 0.1	9.6 ± 0.2%	—	—
1 g concrete	3.1 ± 0.1	9.7 ± 0.2%	80–90%	104
30 g concrete	3.3 ± 0.2	9.5 ± 0.2%	80–90%	3.5

Values for 15 wt.% NH_4OH instrument blank are reported as a reference. 10 mL sample volume

Conclusions

Radioanalytical procedures must strike a balance between complexity, cost and attainable performance. In this work, a new, simplified method has been proposed for the determination of ^{36}Cl in concrete, and its efficiency, selectivity and accuracy have been demonstrated. By exploiting physical threshold in Cherenkov emission, less stringent requirements in terms of decontamination had to be faced, making the method more robust versus a large spectrum of interferents while at the same time being simple and fast to implement. When needed, scalability to large sample masses also allows to reach a DL compatible with clearance requirements without modifying the analytical method. Thanks to the simplicity of the procedure, a complete analysis of a sample—counting included—can be completed within a single working day, at the same time achieving a chemical yield and DL in line with other literature methods, i.e. ≈ 90% and around 10 mBq g^{-1} respectively. Within the scopes of the EURAD-2 ICARUS WP, an intercomparison exercise will also be organized across several laboratories to validate the accuracy and reproducibility of the present method in comparison with alternative protocols. Finally, the present method generates no organic effluents requiring dedicated management and disposal by avoiding the use of resins or scintillation cocktails, with the only residues being the treated concrete and the aqueous counting solution. At the same time, the method is very cost efficient: reagents and materials costs for a single analysis amount to less than 1 €, thus making them negligible with respect to personnel and instrumental needs.

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Data availability The authors declare that the data supporting the findings of this study are available within the paper. Should any raw data files be needed in another format they are available from the corresponding author upon reasonable request.

Declarations

Conflicts of interest The authors declare that they have no competing interests to report.

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