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Cross-Ratio Transferability of Cs-137 Diffusion Models in Cement Paste: A Secondary Quantitative Reanalysis and Evidence-Qualification Framework

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Cross-Ratio Transferability of Cs-137 Diffusion Models in Cement Paste: A Secondary Quantitative Reanalysis and Evidence-Qualification Framework

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Abstract

Background: Apparent radionuclide diffusion in cement paste is frequently invoked in discussions of containment performance, although diffusion, hydraulic leakage and assembly integrity represent different endpoints. Objective: To test whether a parsimonious Cs-137 diffusivity relation transfers between water-to-cement (W/C) ratios within one published dataset, and to determine which external observations can legitimately support a barrier-qualification claim.

Methods: Seven paired porosity observations were reanalyzed using published data . Twenty-four reported Cs-137 diffusivity values from Takiya, comprising two displayed series at three W/C ratios and four temperatures, were assessed by leave-one-W/C-group-out prediction, a temperature-matched comparator, paired-cell aggregation and an exploratory thermal-interaction sensitivity test .

Independent published studies were screened by material, transport endpoint and test-condition compatibility. Results: Mean paired porosity reduction was 16.63 percentage points.

Grouped-holdout mean absolute percentage error was 54.32% for the 24-entry regression versus 119.76% for the comparator, but the regression won only 11 of 24 individual comparisons. At 12 aggregated cells, corresponding errors were 53.82% and 118.59%; the thermal-interaction model yielded 53.92%.

A two-point, figure-digitized OPC comparison with Mihara et al. (2019) showed approximately 3.06-fold and 13.64-fold overprediction at W/C 0.50 and 0.30 under different measurement protocols; no dataset met the criteria for formal external validation.

Conclusions: The apparent pooled advantage does not establish reliable extrapolation across W/C groups or external validity.

We propose a non-compensatory evidence-qualification rule separating hydraulic, radionuclide and assembly-integrity claims. The study does not experimentally qualify a tank, Sr-90 barrier performance or liner durability.

Keywords: Cs-137; cement paste; water-to-cement ratio; grouped holdout; transferability; radionuclide transport; evidence qualification.

1. Introduction

Cementitious materials are used in water-retaining and containment structures, but the ability to retain water hydraulically does not establish radionuclide retention. Conversely, a laboratory apparent diffusion coefficient for an intact cement paste cannot demonstrate the watertightness of a reinforced tank with joints, cracks, penetrations and protective liners. The distinction matters because the underlying observables, exposure conditions and failure mechanisms are different. This paper addresses a narrower, testable question: how far can a published Cs-137 transport relation be transferred between cement-paste water-to-cement ratios, and which types of independent evidence would be admissible for broader barrier claims? Relevant cement-paste, mortar and sorption evidence has been reported in the following studies: .

Cement-based materials are also used in radioactive-waste immobilization, where retention mechanisms depend on chemistry and material state . Atomistic modelling indicates that C-S-H composition can influence Cs retention and mobility .

The quantitative starting point is Takiya's (2015) temperature-by-W/C table of apparent Cs-137 coefficients. Mihara et al. (2019) provide paired cement-paste porosity observations and separate radionuclide diffusion results, while Yamada et al. (2021) report Cs and Sr penetration in mortar. These studies are scientifically complementary but cannot be pooled as interchangeable observations: they differ in matrix, chemistry, boundary conditions and measured endpoints. External evidence is therefore screened for compatibility before, rather than after, any predictive score is calculated.

This study evaluates cross-ratio transferability of a published Cs-137 diffusion model using complete W/C-group holdout, tests the sensitivity of predictive error to paired-value aggregation and temperature interaction, and assesses the compatibility of independent published transport evidence. It is a secondary quantitative reanalysis; no tank-level experiments or containment qualification are claimed.

Table 1. Contribution–evidence–claim boundary matrix

Contribution	Direct evidence	Permitted claim
Paired porosity	Seven formulation pairs, Mihara Table 5	Within-sample association; not permeability or retention
Grouped predictive stress test	Takiya Cs-137, 3 W/C groups, 24 entries / 12 cells	Within-source transferability only; not external validation
Evidence compatibility / H-R-I rule	Endpoint and method screening of independent literature	Auditable decision framework; not tank qualification

2. Methods

2.1 Evidence selection and provenance

The primary quantitative dataset comprised seven formulations transcribed from Table 5 of Mihara, Harasawa and Torii (2019): OPC; fly ash at 15% and 30%; blast-furnace slag at 40% and 60%; silica fume at 10%; and a hybrid mixture containing 40% fly ash and 20% silica fume. Each formulation had reported total porosity at water-to-cement ratios 0.50 and 0.30 . Values were treated as published specimen-level formulation summaries, not independent raw replicate measurements. The available dataset does not include original replicate-level uncertainty. The original Cs apparent-diffusion observations were considered qualitatively in the primary evidence synthesis. The only numerical figure-derived Cs values used in this reanalysis were the two OPC observations digitized from Mihara et al. (2019), Figure 6, for the explicitly exploratory comparison described in Section 3.3.1.

A distinct Cs/Sr ingress evidence stream was drawn from Yamada et al. (2021), whose mortar specimens and concentration-depth endpoints are not interchangeable with the cement-paste diffusivity coefficients in Mihara or Takiya. No numerical coefficients were inferred from the Yamada et al. (2021) concentration-depth figures for the present predictive analysis because the available endpoints and experimental conditions were not directly comparable with the Takiya dataset.

Takiya (2015), Table 3-4 (thesis p. 82), tabulates two displayed Cs-137 values at each w/c (0.36, 0.45, 0.60) and temperature (293, 303, 313, 323 K), in units of 10^{-11} m²/s. A cell-by-cell audit confirmed all 24 values against the primary-source table. The thesis reports an instantaneous-source, non-steady diffusion experiment and depth-profile fitting (Section 3.2.3), but the table itself does not explicitly identify whether the two lines are independent replicate specimens; the analysis therefore labels them “displayed series” rather than independent observations. For each held-out W/C, fit $\ln(Dapp) = a + \frac{b}{T} + c\left(\frac{w}{c}\right)$ on the other

two ratios; all observations at the held-out ratio are excluded together. The baseline is the temperature-matched geometric mean across training ratios. Report all-series, each-series, and paired-geometric-mean results, recognizing that the effective number of independently varied w/c conditions is three.

2.2 Paired porosity and grouped Cs-137 validation

For formulation i, the paired porosity difference was defined as $\Delta\varphi_i = \varphi_i(0.50) - \varphi_i(0.30)$, in percentage points; the relative reduction was $100\Delta\varphi_i/\varphi_i(0.50)$. Mean, median and range were calculated descriptively for the seven reported formulation pairs. No population-level statistical inference was attempted because formulations were selected rather than randomly sampled and original replicate-level uncertainty was unavailable.

Let $D_\mathrm{i} > 0$ denote the reported apparent coefficient (in the source's $10^{-11}\ m^2/s$ units), and $\widehat{D}_\mathrm{i}$ its held-out prediction. Absolute percentage error is $APE_\mathrm{i} = 100\left|\frac{\widehat{D}_\mathrm{i}}{D_\mathrm{i}} - 1\right|$; MAPE is the unweighted mean APE across the specified held-out cells. Signed log error is $e_\mathrm{i} = log^{10}\left(\frac{\widehat{D}_\mathrm{i}}{D_\mathrm{i}}\right)$; mean absolute log error is mean $|e_\mathrm{i}|$. The baseline predicts the geometric mean of the two training W/C ratios at the same temperature; when analyzing both displayed series, the matching series is retained. For the paired-cell analysis, the geometric mean within each temperature–W/C cell is computed first and the same grouped cross-validation is rerun. Model fit is ordinary least squares on natural-log coefficients; no measurements at the held-out W/C are used in training.

There are three distinct validation units: (1) 24 displayed numerical entries, (2) 12 geometric-mean temperature–W/C cells, and (3) three held-out W/C groups. Neither 24 nor 12 is the number of independent mixture groups. The primary interpretation rests on the three grouped folds, with the pooled MAPE shown for descriptive transparency. No confidence interval or hypothesis test of comparative predictive superiority is claimed.

The candidate models were the shared-temperature specification $ln(Dapp) = a + b/T + c(W/C)$, and an exploratory interaction $ln(Dapp) = a + b/T + c(W/C) + d(W/C)/T$. For each held-out W/C group, all its values were excluded from fitting; the baseline was the geometric mean of the two training W/C values at the same temperature. Model selection was descriptive, not externally validated.

Robustness and inference boundary: only three W/C groups are available. Consequently, row-level errors and 12 paired cells are correlated within groups; the reported MAPE values are

descriptive diagnostics, not estimates of out-of-population predictive accuracy. No significance test, calibrated confidence interval, or external-validation claim is attached to the model ranking. The simple baseline is temperature-matched and computed using training groups only. The 24-entry and 12-cell analyses are alternative representations of the same source, not independent validations.

2.3 Endpoint definitions and evidence qualification

For a homogeneous saturated matrix with linear equilibrium sorption, a simplified conservation relation is $\alpha\partial C/\partial t = (D_e\partial\hat{}2\ C)/(\partial x\hat{}2\ )$, with $\alpha = \theta + \rho_b\ K_d$ when aqueous concentration is defined per pore-water volume and units are consistent. Here θ is volumetric water content, ρ_b dry bulk density, K_d a linear solid–liquid distribution coefficient, and D_e the effective diffusion coefficient in a bulk-area flux formulation. Under these assumptions, the apparent diffusion coefficient is given by $D_{app} = \frac{D_e}{\alpha}$. The fitted apparent coefficient already incorporates retardation and must not be paired with an additional independent retardation factor without rederivation. This representation is a conceptual benchmark: changing pore chemistry, nonlinear sorption, drying, cracking and finite solution reservoirs can invalidate its assumptions. Sorption and retardation can depend on cement degradation and pore-water chemistry; they should not be inferred from porosity alone .

Three evidence classes were distinguished: (a) inward concentration–depth profiles; (b) through-diffusion flux and reservoir concentration histories; and (c) outward leaching of initially loaded cement. The reported coefficients are not pooled across these classes without matching definitions and boundary conditions. Hydraulic conductivity, capillary sorptivity, and radionuclide diffusion remain separate endpoints. Distinct experimental endpoints have been documented in . Published leaching and waste-form diffusivity experiments further illustrate why experimental endpoints must be distinguished .

Evidence was organized into three non-compensatory domains: hydraulic containment (H), radionuclide-specific transport and retention (R), and assembly integrity and durability (I). The R domain distinguishes between Cs- and Sr-specific transport evidence, while the I domain includes the integrity and durability of joints, cracks, liners, and other relevant assembly components.

For any specified application, PASS requires measured compliance with each applicable criterion; FAIL requires measured noncompliance with at least one criterion; and missing or incompatible evidence leads to INSUFFICIENT EVIDENCE. Acceptance thresholds must be

defined for the intended service and regulatory context. This framework classifies evidentiary support; it is not a tank certification method.

3. Results

3.1 Porosity and radionuclide evidence

In the seven formulations reported by Mihara et al. (2019), mean total porosity was 34.14% at W/C 0.50 and 17.51% at W/C 0.30. The mean paired difference was 16.63 percentage points ($median$ 17.3; $range$ $13.1 - 19.3$), and all seven differences were positive. These are selected formulation-level comparisons, not randomized independent replicates or evidence of a universal tank-scale effect.

Mihara et al. (2019) reported that blast-furnace slag reduced Cs apparent diffusivity at W/C 0.50 and silica fume contributed to a reduction at W/C 0.30. The corresponding total-porosity values do not establish a universally monotonic ranking of Cs diffusivity across the mixtures. For example, at W/C 0.30 the silica-fume-10% formulation had 16.5% total porosity compared with 12.3% for OPC, while the published study reported a favourable silica-fume effect on Cs apparent diffusion at this water ratio.

This is a within-study qualitative contrast, not a calculated Cs diffusivity ratio. The pore-network topology, chemical binding and sorption mechanisms remain plausible explanatory variables, but their separate causal contributions were not estimated in this reanalysis. Related work on pozzolan-modified cement pastes has also examined Cs and Sr immobilization, without providing a directly interchangeable dataset for the present regression .

Yamada et al. (2021) measured both Cs and Sr ingress and reported lower Sr penetration than Cs and improved resistance with a fly-ash-containing mortar under their investigated conditions. Their results support radionuclide-specific assessment and challenge the assumption that one universal diffusion or sorption coefficient describes both species. However, their mortar ingress results cannot be converted directly into pressure-driven leakage rates, radionuclide transfer through an entire reinforced wall, or performance of an untested liner. Mixed-signal profiles that do not distinguish Cs from Sr are not used to assign species-specific coefficients. This ranking is endpoint- and specimen-specific: independent outward-leaching/migration studies have reported a different Cs/Sr ordering; the present manuscript does not assert a universal radionuclide ranking.

3.2 Internal transferability and sensitivity

Across the 24 displayed Takiya (2015) values, leave-one-W/C-group-out regression MAPE was 54.32%, compared with 119.76% for the temperature-matched geometric-mean comparator. Fold-specific regression MAPEs were 50.46% (W/C 0.36), 21.01% (0.45), and 91.48% (0.60); corresponding comparator MAPEs were 279.05%, 8.16%, and 72.06%. Regression yielded lower individual APE in 11 of 24 comparisons. The first- and second-series-only regression MAPEs were 41.06% and 68.51%. When each displayed pair was aggregated to a geometric-mean cell, regression MAPE was 53.82% and comparator MAPE 118.59% (12 cells). These are sensitivity analyses of the same source, not independent validations. The middle W/C holdout tests interpolation, whereas both extreme holdouts test extrapolation.

Across all 12 paired-cell outcomes, median APE was 37.49%, maximum APE was 120.53%, and mean signed $log10(predicted/observed)\ was\ +0.114$. The extrapolated high-$w/c$ fold had the greatest mean positive log error; average improvement over the simple comparator is therefore not evidence of reliable predictions for new mixtures.

To test the source-indicated variation in activation energies, the shared-temperature model $ln(Dapp) = a + b/T + c(w/c)$ was compared with an interaction model $ln(Dapp) = a + b/T + c(w/c) + d(w/c)/T$. Both were fitted to the same 12 geometric-mean cells and evaluated by leaving each entire w/c group out. Training and testing observations were identical across models; the interaction adds one parameter. Predictors were centered and scaled for numerical stability. The pooled held-group MAPE was 53.82% for the three-parameter model and 53.92% for the four-parameter interaction; mean absolute log10 errors were both 0.18343. Fold-specific MAPE for the shared versus interaction models was 49.20% versus 48.12% (w/c 0.36), 21.08% versus 20.98% (0.45), and 91.18% versus 92.66% (0.60). The interaction did not improve overall transfer and increased the maximum held-out error at w/c 0.60 from 120.53% to 138.24%.

The identical pooled mean absolute log10 errors to five decimal places do not establish physical equivalence. The alternative was examined after recognizing w/c-specific activation energies; the model comparison is therefore exploratory. The grouped holdout has only three test folds, and no independent test set was reserved for final model selection.

The simpler shared-temperature specification is retained as an interpretable descriptive benchmark. The four-parameter interaction is not selected for prediction because its pooled

held-group MAPE was not lower and the extrapolation fold remained unstable. Neither specification should be used to predict tank-scale radionuclide breakthrough, certify a liner, or extrapolate to Sr-90.

Table 2. Model comparison under grouped W/C holdout. Error metrics are descriptive and do not constitute external validation.

Model	MAPE (%)	Median APE (%)	Max APE (%)
W/C + temperature	53.82	37.49	120.53
Thermal interaction	53.92	40.29	138.24
Temperature only	118.70	72.15	390.64
Matched geometric baseline	118.59	71.60	390.71

3.3 External predictive eligibility

Target estimand: error of the frozen Takiya regression $ln(Dapp) = a + b/T + c(w/c)$ on independent Cs-137 apparent-diffusivity measurements from cement paste under compatible source chemistry and measurement protocol. A valid external prediction requires a documented test temperature, w/c, binder, and matching apparent-diffusion endpoint. The external source must not be used for fitting or model selection.

External evidence was evaluated for compatibility of material, tracer, transport endpoint, temperature, W/C and measurement protocol. Mihara et al. (2019) report Cs and I apparent-diffusion evidence in cement pastes incorporating mineral admixtures, but its tracer chemistry and concentration-profile estimation protocol differ from the Takiya (2015) Cs-137 dataset, precluding a strict like-for-like external validation score. A separate exploratory OPC comparison is reported below. García-Gutiérrez et al. (2018) report Cs diffusion in mortars of different cement types; the accessible specimen-level conditions do not establish compatibility with the Takiya cement-paste regression. Yamada et al. (2021) report Cs and Sr ingress in mortars rather than a matching Cs-137 apparent-diffusion endpoint. Accordingly, these studies inform transport mechanisms and domain shift but do not supply a defensible independent predictive test set.

Outcome: no independent observations satisfied the strict comparability requirements for a formal external validation score. Consequently, external validation MAPE, RMSE, calibration slope and interval coverage remain NOT ESTIMABLE. However, the separately reported two-

point OPC comparison in Section 3.3.1 quantifies cross-study disagreement under explicitly different protocols; it is not pooled with the internal holdout results.

3.3.1 Exploratory cross-study OPC comparison from Mihara Figure 6

A restricted, non-validation comparison was performed using the two ordinary Portland cement (OPC) bars in Mihara et al. (2019), Figure 6, rather than admixture-bearing mixtures. The original user-supplied raster (330 × 295 pixels) was digitized from its logarithmic ordinate: approximately y = 31, 77, 123, 169 and 214 pixels correspond to 10^{-10}, 10^{-11}, 10^{-12}, 10^{-13} and 10^{-14} m²/s, respectively. OPC central bar tops were read at approximately y = 81 (W/C 0.50) and y = 139 (W/C 0.30). The reading envelope of ±2 pixels correspond to a multiplicative factor of about 1.105 on each side; it reflects raster digitization only, not the experimental error bars. The reported experimental temperature was 25°C (298.15 K) .

The ±2-pixel envelope is an assumed digitization sensitivity range, not a statistically estimated measurement uncertainty. It excludes uncertainty associated with the original experiment, image compression, axis calibration, and differences between the two experimental protocols.

For a frozen full-source fit, the shared-temperature regression $\ln\left(\frac{Dapp}{\left[\frac{10^{-11}m^2}{s}\right]}\right) = a + \frac{b}{T} + c\left(\frac{W}{C}\right)$ was fitted once to the 12 pair-geometric-mean Takiya cells (T in K), without any Mihara values. This full-source refit is distinct from the leave-one-W/C-group-out models used for internal error estimates. Its coefficients were a = 6.999967, b = -2863.153754 K and c = 7.043285. Predictions were then calculated without adjustment at the two Mihara conditions.

The frozen full-source model was used exclusively for the exploratory cross-study comparison. Its coefficients were not used to calculate the leave-one-W/C-group-out errors reported in Section 3.2. Thus, the internal holdout results and the exploratory external comparisons represent distinct evaluation procedures.

Table 3. Exploratory digitization-based comparison of OPC apparent Cs diffusion coefficients. Ranges represent only ±2-pixel reading uncertainty; ratios do not constitute a validated external predictive accuracy statistic.

OPC W/C	Mihara Figure 6 Dapp (m²/s)	Digitization range (m²/s)	Frozen Takiya prediction (m²/s)	Prediction / observed
0.50	8.19e-12	7.41e-12–9.05e-12	2.506e-11	3.06× (2.77–3.38×)
0.30	4.49e-13	4.06e-13–4.96e-13	6.126e-12	13.64× (12.35–15.08×)

The model overpredicted the figure-derived OPC coefficients by factors of 3.06 (W/C 0.50) and 13.64 (W/C 0.30), with digitization-only ratio envelopes 2.77–3.38 and 12.35–15.08, respectively. W/C 0.50 is within the source training range but is a cross-study protocol shift; W/C 0.30 is additionally outside that range. The tracer chemistry and concentration-profile estimation protocol in Mihara differ from the Takiya Cs-137 radiotracer experiment. The comparison therefore identifies substantial unadjusted numerical disagreement across these specific protocols, not a statistically validated external failure rate or a test of radionuclide tank containment.

3.4 Sr-90, hydraulic leakage and liner evidence

García-Gutiérrez et al. (2018) reported Cs in-diffusion in mortars made from different cement types, including apparent coefficients of approximately $3 - 6 \times 10^{-14}\ m^2/s$ for CEM IV, $1 \times 10^{-13}\ m^2/s$ for CEM I, and up to $7.5 \times 10^{-13}\ m^2/s$ for CEM II. These values are not out-of-sample predictions from the Takiya (2015) cement-paste model. Differences in matrix, cement type and experimental protocol prevent interpreting the numerical contrast as model error or a transferable correction factor.

Goo et al. (2021) reported mean effective leaching coefficients for Cs and Sr of 2.75×10^{-8} and $7.62 \times 10^{-11}\ cm^2/s$, respectively ($2.75 \times 10^{-12}\ and\ 7.62 \times 10^{-15}\ m^2/s$). Their ratio, approximately 361, describes the particular waste-form leaching experiment and must not be interpreted as a Cs-137/Sr-90 through-wall diffusion ratio for a tank. Leaching from an initially loaded solid is a different endpoint from external radionuclide ingress or through-diffusion.

Atkinson and Nickerson (1988) directly examined diffusion and sorption of Cs, Sr and iodine in saturated sulphate-resisting Portland cement, highlighting fast and slow pore networks and ion-specific transport. Their work supports mechanism heterogeneity, not a direct numerical validation of the Takiya temperature–W/C regression.

ACI SPEC-350.1-22 addresses tightness testing of environmental engineering concrete containment structures, subject to its stated scope and exclusions; it is not evidence of radiological containment performance in an untested structure . ASTM C1308-21 addresses accelerated contaminant leaching from solidified waste and is not a hydraulic tightness test . Neither document substitutes for application-specific measurements or applicable regulatory requirements.

3.5 Consolidated visual results and evidence compatibility

Figures 1 and 2 summarize the grouped-holdout predictive diagnostics using the 12 pair-geometric-mean temperature–W/C cells from Takiya (2015). These cells represent three distinct W/C groups and should not be interpreted as 12 independent mixture groups. Figure 3 presents the separate exploratory cross-study comparison between frozen Takiya-model predictions and the two OPC apparent Cs diffusion coefficients digitized from Mihara et al. (2019). Its uncertainty bars represent raster-digitization uncertainty only, and the comparison does not constitute formal external predictive validation.

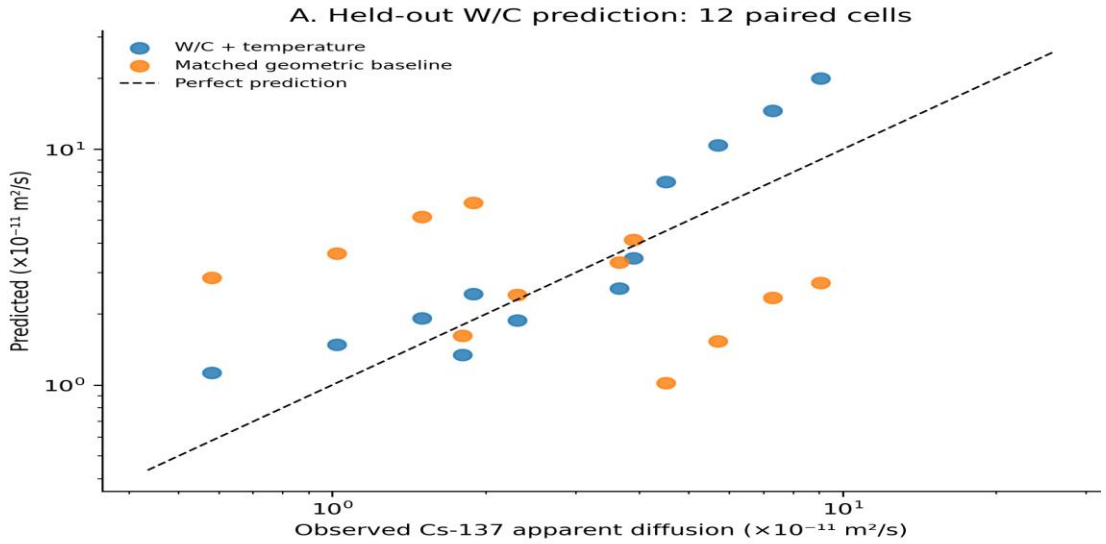


Figure 1. Observed versus out-of-group predicted Cs-137 apparent diffusion coefficients. Dashed line denotes equality; both axes are logarithmic. Data: Takiya (2015), secondary reanalysis.

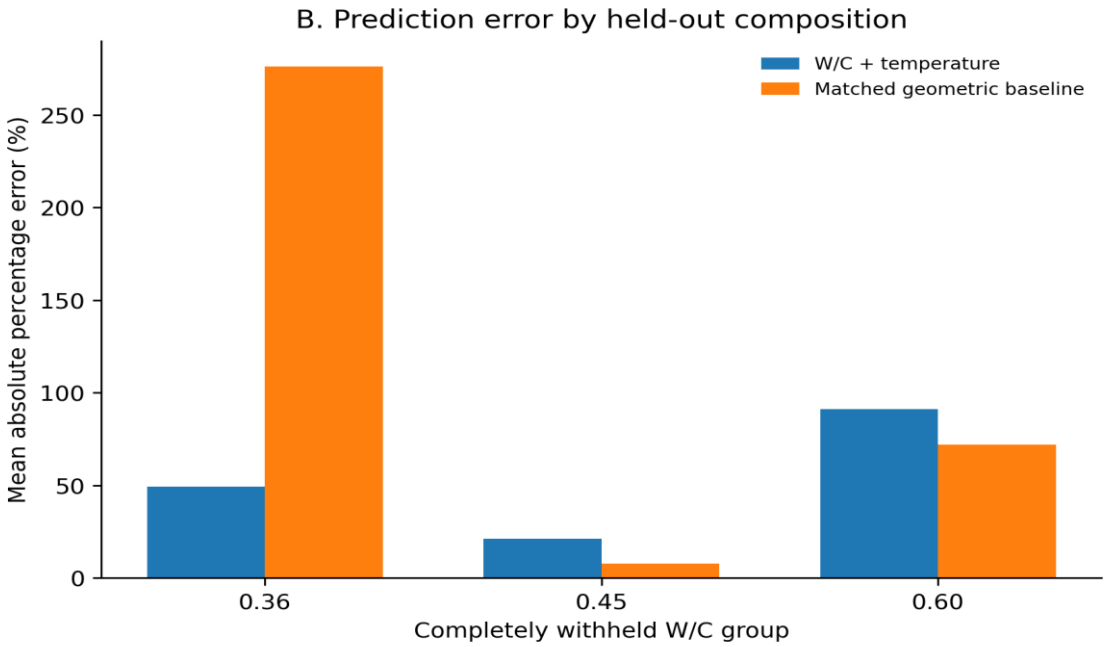


Figure 2. Fold-specific mean absolute percentage error for the parsimonious model and matched-temperature geometric baseline. Three withheld W/C levels provide limited between-group evidence.

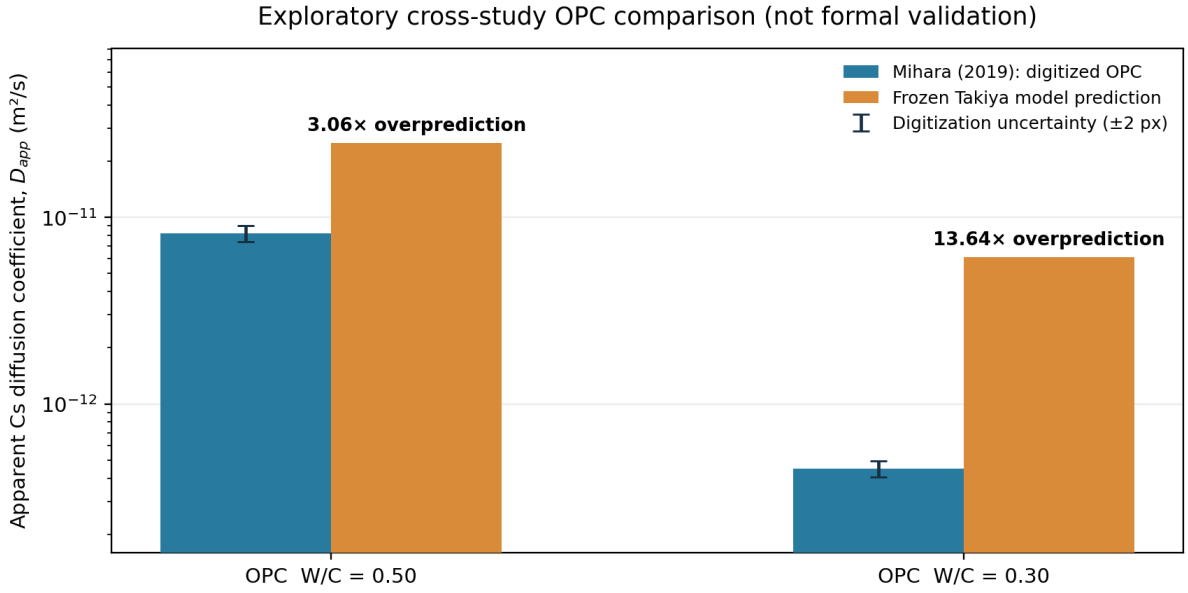


Figure 3. Exploratory comparison of apparent Cs diffusion coefficients for OPC at W/C = 0.50 and 0.30: frozen Takiya-model predictions versus values digitized from Mihara et al. (2019), Figure 6. Error bars represent an assumed ±2-pixel digitization sensitivity range, not experimental confidence intervals. Predictions exceed the digitized observations by factors of 3.06 and 13.64, respectively. Differences in experimental protocols preclude formal external predictive validation.

4. Discussion

4.1 What the evidence supports

In the seven cement-paste formulations reported by Mihara et al. (2019), the lower W/C level was associated with lower total porosity. The same study indicates that radionuclide transport also varies with mineral admixture and experimental conditions. Porosity alone is therefore not a sufficient proxy for Cs or Sr retention. Strong sorption may reduce outward migration under some conditions but may also retain contamination within a cementitious component, a distinct consideration for subsequent management .

Pressure-driven water flow depends on hydraulic conductivity, cracks, joints, head and contact conditions. Molecular diffusion may continue in saturated material even without a measurable bulk water leak. Capillary sorptivity is informative about unsaturated uptake but is not equivalent to saturated hydraulic permeability. Liner performance further depends on adhesion, seams, penetrations, defects and aging. These distinct mechanisms motivate mandatory qualification domains rather than a compensatory weighted score. These mechanisms are distinct from the species-specific diffusion and sorption processes investigated by Atkinson and Nickerson (1988).

Binding mechanisms and changes in cement chemistry further limit transfer between distinct specimen and exposure conditions . The broader multi-barrier interpretation of cementitious retention is also discussed in the literature .

Mihara et al. (2019) investigated hardened cement pastes; Yamada et al. (2021) investigated mortars. Neither directly represents reinforced tank walls with construction joints, reinforcement interfaces, cracks, water pressure and service-specific chemistry. Cs and Sr results depend on concentration, ionic strength, carbonation, calcium leaching and moisture history. The manuscript therefore presents a constrained evidence-analysis method, not a certified material specification for potable-water or wastewater containment.

All 24 Cs-137 entries were checked against Takiya (2015), Table 3-4 (p. 82). The table displays two series per temperature–w/c cell; the current audit does not establish whether these correspond to independently prepared specimens, repeat analyses, or other source-specific reporting conventions. The effective design spans three w/c groups, and no inference treats 24 rows as independent mixtures. The separate Takiya et al. water-tracer article is not a source for these Cs-137 values. Mihara et al. (2019) provide independent, material-specific Cs evidence, but differences in tracer specification, binder, curing, temperature and protocol preclude a

defensible numerical external validation of the Takiya model. Yamada et al. (2021) provide mortar Cs/Sr ingress evidence, not directly comparable Cs-137 paste diffusion coefficients.

The shared-temperature-slope model is a parsimonious descriptive fit, whereas Takiya (2015) reports Cs-137 activation energies of 31.3 ± 4.0, 22.0 ± 2.3 and 18.7 ± 1.7 kJ mol^{-1} for w/c 0.36, 0.45 and 0.60, respectively. This suggests possible temperature–composition interaction. However, a direct leave-one-w/c-out comparison did not favor an added interaction term (paired-cell MAPE 53.92% versus 53.82%). These are three grouped folds, not 12 independent external tests. The end-member folds extrapolate beyond the training w/c range; pooled MAPE can mask their instability. No meaningful population confidence interval or claim of external predictive validity is made. The alternative model comparison was exploratory; model selection and performance estimation on the same folds may be optimistic.

The lower pooled MAPE for the W/C-plus-temperature regression was influenced by the poor performance of the geometric-mean comparator in the lowest withheld W/C group. The regression improved absolute percentage error in only five of the twelve aggregated cells. Therefore, the pooled mean does not establish consistent model superiority or reliable transfer to an untested cement composition. The exploratory temperature interaction did not improve pooled grouped-holdout MAPE.

The Mihara OPC cross-study check provides a useful quantitative stress test but does not resolve whether disagreement arises from tracer identity, pore chemistry, sample preparation, estimation method or the W/C relationship. The plotted experimental error bars were not used as quantitative confidence intervals, and ±2-pixel bounds capture only the uncertainty of reading the supplied low-resolution raster. The result therefore warrants reporting as evidence of limited direct portability, not as a validated external error statistic. This interpretation is consistent with broader reviews emphasizing chemical retention and material-specific behavior in cementitious waste forms . Evidence on Cs exchange and removal from contaminated concrete further underscores the sensitivity of measured outcomes to the chemical environment and treatment protocol .

4.2 Non-compensatory decision rule

The H/R/I rule does not permit favorable results in one domain to compensate for missing evidence in another. The present literature reanalysis cannot establish a tank-level PASS or FAIL: it includes no pressure-leakage testing of an assembled tank, no through-wall Sr-90 breakthrough measurement, and no joint, crack or liner-aging qualification. These absent measurements are not interpreted as measured failures.

4.3 Unresolved scientific and engineering gaps

No independent Cs-137 dataset meeting strict protocol and endpoint compatibility was identified for formal external validation. The exploratory Mihara OPC comparison showed overprediction under different tracer and analysis conditions, and cannot isolate the cause of the disagreement. The two displayed Takiya series cannot be assumed to represent independent experimental specimens. Published Sr ingress and cement-waste leaching results do not establish Sr-90 through-wall breakthrough, and no hydraulic leakage, joint, crack or liner-aging measurements were available for a complete tank assembly. These are limitations of the evidence base, not negative experimental test results.

5. Conclusions

Across seven published cement-paste formulations, lower W/C was associated with lower total porosity, but porosity alone did not establish radionuclide retention. In the three-W/C-group Cs-137 dataset of Takiya (2015), a simple regression had lower pooled grouped-holdout MAPE than a temperature-matched comparator; however, errors differed substantially between held-out groups and the regression did not improve most individual predictions. Pair aggregation and an exploratory temperature interaction did not remove this limitation. A two-point figure-digitized OPC comparison with Mihara et al. (2019) indicated approximately 3.06-fold and 13.64-fold overprediction at W/C 0.50 and 0.30, respectively, under differing measurement protocols; it is not a strict external validation test. The proposed H/R/I evidence-qualification framework separates radionuclide transport, hydraulic containment, and assembly integrity into distinct, non-compensatory evidence domains. The present secondary analysis does not establish compliance with application-specific containment requirements, and no tank-level qualification is inferred from the published cement-paste observations.

Declarations

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Conflicts of interest: The author declares no conflicts of interest.

Ethics approval: Not applicable. This study reanalyzes published, non-personal experimental data and involved no new human or animal research.

Data availability: The experimental data used in this secondary analysis were obtained from publicly available published sources. The Cs-137 apparent diffusion coefficients were transcribed from Takiya (2015), Table 3-4, and the paired porosity values from Mihara et al. (2019), Table 5. The exploratory OPC comparison uses approximate values digitized from Figure 6 of Mihara et al. (2019), as described in Section 3.3.1. No new experimental data were generated. The model specifications, grouping procedures, and error definitions are reported in the Methods section.

Use of AI-assisted tools: AI-assisted tools supported language editing and computational workflow development. The author is responsible for the accuracy of the analysis, citations and scientific interpretation.

Author Contributions

Conceptualization, Methodology, Formal Analysis, Framework Development, Writing – Original Draft, Writing – Review and Editing.

References

American Concrete Institute, 2022. *ACI SPEC-350.1-22: Tightness Testing of Environmental Engineering Concrete Structures—Specification,* s.l.: American Concrete Institute.

ASTM International, 2021. *ASTM C1308-21: Standard Test Method for Accelerated Leach Test for Measuring Contaminant Releases From Solidified Waste,* s.l.: ASTM International.

Atkins, M. & Glasser, F. P., 1992. Application of Portland cement-based materials to radioactive waste immobilization. *Waste Management,* 12(2–3), p. 105–131.

Atkinson, A. & Nickerson, A. K., 1988. Diffusion and Sorption of Cesium, Strontium, and Iodine in Water-Saturated Cement. *Nuclear Technology,* 81(1), pp. 100-113.

Bar-Nes, G., Peled, Y., Shamish, Z. & Katz, A., 2017. Cesium and strontium immobilization in Portland cement pastes blended with pozzolanic additives. *Journal of Nuclear Engineering and Radiation Science,* 3(3), p. 030907.

Duque-Redondo, E., Yamada, K. & Manzano, H., 2021. Cs retention and diffusion in C-S-H at different Ca/Si ratio. *Cement and Concrete Research,* Volume 140, p. 106294.

Evans, N. D. M., 2008. Binding mechanisms of radionuclides to cement. *Cement and Concrete Research,* 38(4), p. 543–553.

Gaona, X. et al., 2021. Cement as a component of the multi-barrier system: radionuclide retention in cementitious environments. *Safety of Nuclear Waste Disposal,* Volume 1, p. 151–152.

García-Gutiérrez, M. et al., 2018. Cesium diffusion in mortars from different cements used in radioactive waste repositories. *Applied Geochemistry,* Volume 98, p. 10–16.

Goo, J.-Y.et al., 2021. Leaching behavior of cesium, strontium, cobalt, and europium from immobilized cement matrix. *Applied Sciences,* 11(18), p. 8418.

Jain, S., Onuaguluchi, O., Banthia, N. & Troczynski, T., 2025. Advancements in immobilization of cesium and strontium radionuclides in cementitious wasteforms—A review. *Journal of the American Ceramic Society,* 108(1), p. e20131.

Jang, J. G., Park, S. M. & Lee, H. K., 2016. Physical barrier effect of geopolymeric waste form on diffusivity of cesium and strontium. *Journal of Hazardous Materials,* Volume 318, p. 339–346.

Kim, J. H., Anwer, H., Kim, Y. S. & Park, J.-W., 2021. Decontamination of radioactive cesium-contaminated soil/concrete with washing and washing supernatant–critical review. *Chemosphere,* Volume 280, p. 130419.

Mihara, M., Harasawa, S. & Torii, K., 2019. Apparent diffusion coefficients of Cs and I in hardened cement paste using mineral admixtures. *Journal of Nuclear Fuel Cycle and Environment,* 26(1), pp. 15-23.

Ochs, M., Mallants, D. & Wang, L., 2016. *Radionuclide and Metal Sorption on Cement and Concrete.* 1 ed. Cham, Switzerland: Springer International Publishing.

Ochs, M., Pointeau, I. & Giffaut, E., 2006. Caesium sorption by hydrated cement as a function of degradation state: Experiments and modelling. *Waste Management,* 26(7), p. 725–732.

Papadokostaki, K. G. & Savidou, A., 2009. Study of leaching mechanisms of caesium ions incorporated in ordinary Portland cement. *Journal of Hazardous Materials,* 171(1–3), p. 1024–1031.

Takiya, H., 2015. *Diffusion mechanism of water and ions in hardened cement pastes with different water to cement ratios.* Sapporo: Hokkaido University.

Yamada, K. et al., 2021. Experimental study investigating the effects of concrete conditions on the penetration behaviors of Cs and Sr at low concentration ranges. *Journal of Advanced Concrete Technology,* 19(6), p. 756–770.