

## Article

# A Leaching-Index-Driven Framework for Durability-Oriented Design of Mineral Binders: Validation on Acid-Induced Degradation of an NHL–Pozzolan System and Prospective Extensions to Circular Materials

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## Abstract

Most studies on circular mineral-based materials report short-term mechanical properties without providing predictive frameworks that link chemical degradation to long-term mechanical performance. This study develops and validates a leaching-index-driven chemo-mechanical framework for predicting the degradation of a natural hydraulic lime (NHL)–pozzolan mortar exposed to sulfuric acid. A normalized ionic-release index was used to drive all parameters of a trilinear continuum damage mechanics (CDM) model, enabling the prediction of complete stress–strain responses from leachate chemistry alone. The framework was calibrated using an extensive experimental dataset comprising accelerated acidic exposure at pH 1.5, 2.0, and 3.0 for durations up to 6000 h. Iron release was identified as the most suitable degradation indicator based on its monotonic evolution and strong correlation with mechanical deterioration. Power-law relationships linking the normalized leaching index to elastic modulus, peak strength, transition strain, and post-peak energy were established and validated against independent exposure groups, yielding prediction errors generally below 20%. Validation was performed against three independent blind exposure groups withheld from calibration, yielding mean deviations of approximately 18% in the elastic modulus and 13% in the peak strength, so that the quantitative validation rests on this limited three-group set, whereas the extension to circular mineral binders is presented only on a prospective, non-validated basis. A kinetic sub-model was further introduced to relate exposure conditions to the leaching index, enabling a complete predictive chain from environmental exposure to mechanical response and service-life estimation. Sensitivity analysis showed that post-peak energy dissipation degrades approximately 1.3–1.5 times faster than stiffness and strength, indicating that ductility-related parameters govern long-term reliability. The methodological architecture is further discussed, on a prospective and non-validated basis, in relation to circular mineral binders relevant to Baltic industrial and municipal by-product streams, including municipal-waste bottom ash, slag, fly ash, and recycled glass. Although experimental validation is limited to the NHL–pozzolan system, the proposed framework provides a phys-

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ically grounded and data-efficient pathway for durability assessment and future durability-oriented design of circular mineral binders in the Baltic region.

**Keywords:** chemo-mechanical modeling; continuum damage mechanics; durability prediction; ionic leaching index; lime-based mortar; circular mineral binders

## 1. Introduction

The construction sector is responsible for approximately 39% of global energy-related CO<sub>2</sub> emissions, of which around 11% is attributable directly to the manufacture of building materials, with cement clinker production accounting for the largest single share within this category [1–6]. Reducing the carbon intensity of construction materials therefore constitutes one of the most impactful levers available under the European Green Deal [2] and the associated Circular Economy Action Plan [1], both of which mandate a substantial reduction in virgin-resource extraction and an increase in the valorization of industrial and municipal by-products. Among the most promising substitutes for Portland cement clinker are granulated blast-furnace slag (GGBS), co-combustion fly ash, biomass, and municipal-waste bottom ash (MWBA), as well as amorphous silica-rich materials such as recycled glass fines [7–11]. In the Baltic states (Latvia, Lithuania, and Estonia), these streams are generated in considerable quantities by district-heating and combined-heat-and-power plants, steel production facilities, and waste-to-energy installations, yet their systematic valorization as supplementary cementitious materials or alkali-activated binder precursors in structural-grade mortars and concrete remains at an early stage [12–14].

Most existing studies on circular mineral-based materials limit their performance characterization to compressive and flexural strength measured under standard curing at 28 days, occasionally supplemented by porosity and X-ray diffraction data [13,15–19]. While such data are necessary for demonstrating the feasibility of waste valorization, they are insufficient for the structural application of these materials in infrastructure components exposed to acidic, saline, or carbonating environments over service lives of 50–100 years [20–22]. Reporting strength without associated durability data creates a critical inconsistency in sustainability accounting: a circular material that exhibits adequate initial strength but poor resistance under aggressive exposure cannot be regarded as genuinely sustainable, because premature failure necessitates earlier replacement and thereby negates the environmental benefit of using recycled inputs [23–28]. Recent machine-learning approaches to durability prediction, including gradient-boosting models trained on the same lime-based acidic-exposure dataset employed in the present study, have begun to address this limitation [29–33]; however, purely data-driven models lack the physical basis required for mechanistic extrapolation beyond the training domain and demand large experimental sets that are rarely available for emerging binder chemistries. A physically grounded, data-efficient alternative is therefore required.

Durability-oriented design requires constitutive frameworks capable of linking chemical degradation mechanisms to macroscopic mechanical deterioration and, ultimately, to service-life prediction and environmental-impact assessment [34–37]. In lime-based and cementitious mortars, acidic environments induce decalcification, phase dissolution, gypsum formation, and microcracking, each producing measurable changes in elastic modulus, peak strength, and post-peak ductility [15,38,39]. These processes have been characterized experimentally for Portland-cement systems and, to a lesser extent, for natural hydraulic lime (NHL) mortars [15,40,41], but their quantitative integration into a predictive mechanical model has not been systematically demonstrated. Continuum dam-

age mechanics (CDM) provides a physically consistent description of the progressive deterioration of quasi-brittle materials, in which a scalar damage variable represents the loss of stiffness and load-bearing capacity as microcracks nucleate, propagate, and coalesce [42–44]. Trilinear CDM formulations that segment the stress–strain response into elastic, inelastic-hardening, and post-peak softening regimes have been applied to cementitious systems under sulfate attack [45] and to fiber-reinforced concrete [42], yet their coupling to measurable chemical degradation indicators, rather than to exposure time alone, remains largely unexplored for lime-rich and waste-derived binders [43,46].

The present paper addresses this gap by developing and validating a leaching-index-driven chemo-mechanical damage framework whose central feature is that a single normalized ionic-release index is used to drive every parameter of a trilinear CDM model, so that complete stress–strain responses can be predicted under untested acid-exposure conditions from leachate chemistry alone, without further mechanical calibration. The framework is calibrated and validated on the comprehensive NHL–pozzolan acidic-durability dataset reported by the authors [47,48]. Relative to the experimental study [47], which reported the mechanical and microstructural data without constitutive modeling, and relative to the gradient-boosting analysis [49], which trained a black-box model on the same dataset without a mechanistic structure, the present work contributes four distinct elements. First, it provides the first physically grounded trilinear CDM calibration for the binder family and reports the full parameter set across fifteen exposure conditions. Second, it introduces, defends, and benchmarks the use of a single normalized iron-leaching index as the master driver of all four CDM parameters, with the selection justified by a direct quantitative comparison against calcium and silicon and supported by a binder mass balance. Third, it derives an analytical sensitivity expression directly from the calibrated laws, propagated with bootstrap confidence intervals on the regression coefficients, that ranks the vulnerability of each mechanical descriptor to chemical attack. Fourth, it formulates the methodological architecture as a generic chemo-mechanical mapping in which the master index is binder-specific, and examines, without claiming experimental validation, the form this mapping would take for four circular binder chemistries relevant to Baltic by-product streams. The materials and methods are presented in Section 2, including a dedicated subsection on the selection of iron as the degradation indicator (Section 2.5) and a kinetic sub-model linking exposure time and pH to the normalized index (Section 2.6); the results and their discussion are presented in Section 3; the sustainability implications and the corrected composite performance index in Section 4; and the conclusions in Section 5.

To make the boundary between the previously published material and the present contribution explicit, Table 1 contrasts the data, analyses, and conclusions already reported in the experimental and machine-learning studies of this binder with the elements that are developed for the first time in the present work.

**Table 1.** Delineation of the previously reported results and the contributions introduced in the present work.

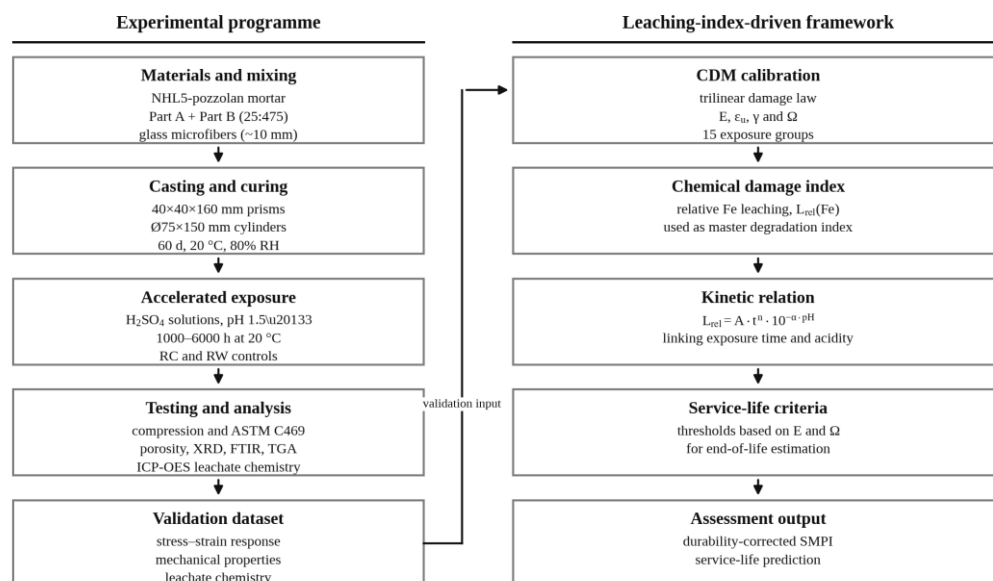
| Aspect                | Previously Reported (Refs. [34, 48, 49])                                                                                                                  | New in the Present Work                                                   |
|-----------------------|-----------------------------------------------------------------------------------------------------------------------------------------------------------|---------------------------------------------------------------------------|
| Experimental database | Accelerated sulfuric-acid exposure of the NHL–pozzolan mortar at pH 1.5–3.0 for up to 6000 h, with mechanical, microstructural, and ICP-OES data [34, 48] | Re-used as calibration and validation input; no new specimens were tested |

|                                         |                                                                                                      |                                                                                                                            |
|-----------------------------------------|------------------------------------------------------------------------------------------------------|----------------------------------------------------------------------------------------------------------------------------|
| Mechanical and microstructural evidence | Strength, modulus, porosity, XRD, TGA, FTIR, and SEM–EDX with altered-zone measurements [34, 48, 49] | Reduction in each compressive response to a compact trilinear CDM parameter set                                            |
| Data-driven durability prediction       | Gradient-boosting (XGBoost) model trained on the same dataset [49]                                   | Physically grounded constitutive alternative with closed-form degradation laws                                             |
| Constitutive model                      | Not reported                                                                                         | First trilinear CDM calibration for the binder across all fifteen exposure groups                                          |
| Degradation index                       | Not reported                                                                                         | Normalized iron-leaching index, selected by quantitative comparison against calcium and silicon, and a binder mass balance |
| Kinetic sub-model                       | Not reported                                                                                         | Constant-rate kinetic law linking exposure time and pH to the leaching index                                               |
| Sensitivity and uncertainty             | Not reported                                                                                         | Closed-form parameter sensitivity with bootstrap and Monte Carlo intervals                                                 |
| Sustainability metric                   | Not reported                                                                                         | Durability-corrected service life and a corrected Sustainable Material Performance Index                                   |
| Circular-binder extension               | Not reported                                                                                         | Prospective, non-validated mapping to Baltic industrial and municipal by-product binders                                   |

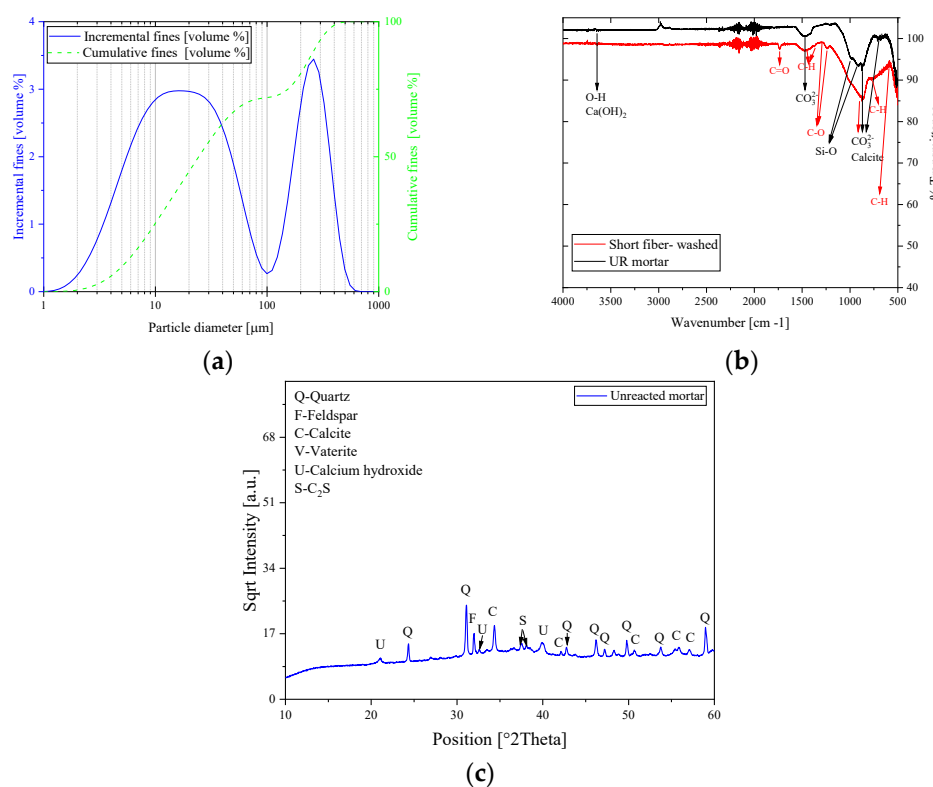
## 2. Materials and Methods

### 2.1. Experimental Database and Exposure Program

The overall experimental program underlying the validation dataset, together with its coupling to the present modeling framework, is summarized in Figure 1. The framework was calibrated and validated against the comprehensive experimental dataset reported in Azimi et al. [48], in which the durability of a binary NHL–pozzolan mortar (commercially designated Mapei Planitop HDM Restauro from Milan, Italy) was investigated under accelerated acidic exposure. The mortar consists of natural hydraulic lime (NHL5) and eco-pozzolan additives, formulated as a two-component system: Part A, a dry premix of NHL5 with mineral fillers, graded aggregates, and short glass microfibers of approximately 10 mm length; and Part B, a water-based admixture controlling rheology, water retention, and adhesion, combined at a prescribed ratio of 25:4.75 by mass. X-ray diffraction of the unreacted mortar identified quartz, calcite, portlandite, and ettringite as the principal crystalline phases, with minor reflections attributable to belite and alite from the hydraulic binder. The dry-mortar particle-size distribution, obtained by laser diffraction, exhibited a bimodal pattern characteristic of NHL–aggregate binary systems, and scanning electron microscopy with energy-dispersive X-ray analysis (SEM–EDX) confirmed that the embedded short fibers were glass-based with a Ti- and Zr-bearing surface coating providing chemical resistance to the immersion solutions (Figure 2) [48].



**Figure 1.** Experimental program and leaching-index-driven chemo-mechanical framework developed in this study.



**Figure 2.** Characterization of the NHL-pozzolan mortar used in the validation study: (a) particle-size distribution; (b) FTIR spectra; (c) XRD pattern. The scanning electron micrograph of the coated glass fibers referred to in Section 2.1 is reported in Ref. [44].

Prismatic specimens ( $40 \times 40 \times 160$  mm) were cast for flexural and compressive testing and cylindrical specimens ( $75 \times 150$  mm) for elastic-modulus and compressive-strength characterization. After curing for 60 days at  $20^\circ\text{C}$  and 80% relative humidity, specimens were subjected to five exposure environments: dry storage in a climatic chamber (RC), continuous water immersion (RW), and immersion in sulfuric-acid solutions at pH 3.0, 2.0, and 1.5 (designated P3, P2, and P15) for 1000, 3000, and 6000 h. Solutions were held at

20 °C, refreshed after two weeks and monthly thereafter, and the pH was monitored and adjusted twice weekly. Mechanical testing was conducted according to EN 1015-11 [50], BS EN 12390-13 [51], and ASTM C469 [52], complemented by open-porosity and bulk-density measurements per EN 1936 [53], and by XRD, FTIR, TGA, and inductively coupled plasma optical-emission spectroscopy (ICP-OES) of the leachate per ISO 11885:2007 [54]. More than 560 destructive mechanical tests were performed in total, providing the calibration and validation basis for the present framework. Each exposure condition was represented by three nominally identical specimens, and the values employed for calibration correspond to the mean of these replicates, whose specimen-to-specimen variability is quantified in Section 3.4. Compressive loading was applied at a controlled quasi-static rate in accordance with EN 1015-11 and BS EN 12390-13, and the axial strain required for the static elastic-modulus determination was recorded over the central gauge length of the cylindrical specimens following the repeated loading and unloading protocol of ASTM C469, so that complete stress–strain records were obtained for every condition and subsequently reduced to the trilinear parameter set [50–52]. For ease of reference, the exposure groups are designated throughout by the code E\_C\_Px\_yyyy, in which E\_C denotes the compressive test series, Px the acid severity (P15 = pH 1.5, P2 = pH 2.0, P3 = pH 3.0) and yyyy the exposure duration in hours, while the unaged reference, the water-immersed and the dry-stored conditions are denoted E\_C, RW and RC, respectively.

## 2.2. Architecture of the Chemo-Mechanical Framework

The framework couples chemical degradation indicators to a mechanical constitutive model through three operations applied to a common experimental campaign. In the first, the precursor and the aged matrix are characterized chemically and microstructurally by XRF, XRD, FTIR, TGA, SEM-EDX, and ICP-OES leaching analysis, the last providing the ionic-release data that serve as the chemical driver of the model; the susceptibility of a binder to chemical attack is governed primarily by the content and distribution of calcium-bearing phases such as portlandite, calcite, and calcium silicate hydrates [15,21,55,56]. In the second, the uniaxial compressive stress–strain response measured for each exposure condition is reduced to a compact set of CDM parameters by calibration of the trilinear constitutive law described in Section 2.3. In the third, each calibrated parameter is expressed as an explicit function of a normalized chemical degradation index by nonlinear regression, as described in Section 2.4, so that the mechanical response under any acid-exposure condition can subsequently be reconstructed from leachate chemistry alone. Unlike conventional time-based degradation models, the present architecture makes the measurable chemistry of the leachate, rather than exposure duration, the independent variable, which is what permits transfer of the methodology between binder systems that degrade by different chemical routes.

## 2.3. Trilinear Continuum Damage Constitutive Model

The compressive behavior of the mortar was represented by a trilinear CDM formulation adapted from Shen et al. [42], which captures the elastic, inelastic-hardening, and post-peak softening regimes characteristic of quasi-brittle lime-based materials. In the elastic regime up to the transition strain  $\gamma$ , the stress was taken as proportional to strain through the initial elastic modulus  $E_0$ . In the hardening regime between  $\gamma$  and the peak strain  $\varepsilon_u$ , the stress was interpolated linearly between the transition stress and the peak stress  $\sigma_u$ . In the post-peak regime beyond  $\varepsilon_u$ , the stress was governed by progressive damage according to the residual stress  $\sigma_{res}$  and the damage variable, as expressed in Equation (1):

$$\sigma(\varepsilon) = E_0 \cdot \varepsilon \text{ for } \varepsilon \leq \gamma$$

$$\sigma(\gamma) + \left[ \frac{\sigma_u - \sigma(\gamma)}{\varepsilon_u - \gamma} \right] \cdot (\varepsilon - \gamma) \text{ for } \gamma < \varepsilon \leq \varepsilon_u \quad (1)$$

$$\sigma_{res} + [1 - D(\varepsilon)] \cdot (E^0 \cdot \varepsilon - \sigma_{res}) \text{ for } \varepsilon > \varepsilon_u$$

Here, the transition strain was fixed at  $\gamma = 0.6 \frac{\sigma_u}{E_0}$  from the analysis of the experimental stress–strain envelopes, and the residual stress was fixed at  $\sigma_{res} = 0.10 \sigma_u$ . The stability of these fixed ratios across the fifteen exposure groups was assessed in Section 3.2 by releasing  $\gamma$  and  $\sigma_{res}$  as free parameters for three representative groups (reference, E\_C\_P15\_3000, and E\_C\_P15\_6000); the resulting improvement in coefficient of determination was below 0.01 in each case, justifying retention of the fixed-ratio formulation for the principal calibration and limiting the number of free parameters to four per group. The scalar damage variable was formulated as an energy-based exponential function,  $D(\varepsilon) = 1 - \exp\left[-\frac{W(\varepsilon)}{\Omega}\right]$ , in which  $\Omega$  is the characteristic energy threshold for damage propagation and  $W(\varepsilon)$  is the strain-energy density accumulated beyond the peak, computed as the integral of the difference between the linear-elastic stress and the measured stress over the post-peak strain range [42]. A higher value of  $\Omega$  corresponds to a larger characteristic energy threshold and therefore to a more gradual, quasi-ductile post-peak response, whereas a lower value describes a more brittle collapse in which the residual stress is approached over a shorter post-peak strain range. The parameter  $\Omega$  carries the dimensions of a strain-energy density (MJ·m<sup>−3</sup>, numerically equal to MPa) and is bounded below by a small positive value so that the exponential damage law remains well defined; the entries reported as 0.00 in Table 2 therefore correspond to effectively brittle groups whose fitted  $\Omega$  falls below the two-decimal reporting precision rather than to a physically meaningful zero energy threshold. Calibration was performed in MATLAB R2024a (MathWorks, Natick, MA, USA) using the `fmincon` constrained-optimization routine, minimizing the sum of squared differences between modeled and measured stresses with all parameters confined to physically meaningful bounds; a compliance correction was applied to the cylindrical compression curves to remove the contribution of system stiffness before fitting.

**Table 2.** Calibrated CDM parameters for all exposure groups, including the normalized iron-leach index ( $L_{rel}$ ) and number of fitted strain points ( $n$ ) [48].

| Group                 | $E_0$ (MPa) | $\gamma$ (–) | $\varepsilon_u$ (–) | $\sigma_u$ (MPa) | $\sigma_{res}$ (MPa) | $\Omega$ (MPa) | $R^2$ | $L_{rel}$ | $n$ |
|-----------------------|-------------|--------------|---------------------|------------------|----------------------|----------------|-------|-----------|-----|
| E_C (reference)       | 2376        | 0.001        | 0.013               | 16.57            | 1.66                 | 0.13           | 0.958 | N/A       | 120 |
| E_C_RC_1000           | 1894        | 0.006        | 0.011               | 16.78            | 1.68                 | 0.00           | 0.971 | N/A       | 115 |
| E_C_RC_3000           | 2360        | 0.003        | 0.013               | 18.13            | 1.81                 | 0.10           | 0.975 | N/A       | 118 |
| E_C_RC_6000           | 1465        | 0.007        | 0.016               | 16.71            | 1.67                 | 0.08           | 0.946 | N/A       | 110 |
| E_C_RW_1000           | 2560        | 0.001        | 0.012               | 16.00            | 1.60                 | 0.15           | 0.953 | N/A       | 120 |
| E_C_RW_3000           | 1868        | 0.006        | 0.011               | 15.45            | 1.54                 | 0.09           | 0.956 | N/A       | 115 |
| E_C_RW_6000           | 1188        | 0.012        | 0.014               | 15.13            | 1.51                 | 0.01           | 0.966 | N/A       | 108 |
| E_C_P15_1000          | 2685        | 0.003        | 0.011               | 18.23            | 1.82                 | 0.09           | 0.958 | 0.167     | 118 |
| E_C_P15_3000 (anchor) | 2142        | 0.006        | 0.012               | 19.08            | 1.91                 | 0.07           | 0.959 | 0.500     | 116 |
| E_C_P15_6000          | 1419        | 0.009        | 0.016               | 17.63            | 1.76                 | 0.08           | 0.770 | 1.000     | 98  |
| E_C_P2_1000 (blind)   | 1903        | 0.007        | 0.011               | 17.10            | 1.71                 | 0.00           | 0.971 | 0.033     | 117 |
| E_C_P2_3000           | 2549        | 0.003        | 0.012               | 19.02            | 1.90                 | 0.12           | 0.967 | 0.098     | 119 |
| E_C_P2_6000 (blind)   | 1087        | 0.013        | 0.016               | 15.75            | 1.57                 | 0.01           | 0.976 | 0.196     | 104 |
| E_C_P3_1000           | 1836        | 0.007        | 0.012               | 17.38            | 1.74                 | 0.00           | 0.947 | 0.004     | 119 |
| E_C_P3_3000 (blind)   | 2222        | 0.006        | 0.011               | 18.95            | 1.89                 | 0.11           | 0.958 | 0.011     | 118 |
| E_C_P3_6000           | 1462        | 0.008        | 0.016               | 17.05            | 1.71                 | 0.08           | 0.923 | 0.022     | 115 |

Entries reported as 0.00 for  $\Omega$  denote effectively brittle post-peak responses for which the fitted characteristic energy threshold falls below the two-decimal reporting precision;  $\Omega$  was constrained to a small positive lower bound during calibration and is expressed in  $\text{MJ}\cdot\text{m}^{-3}$ , numerically equal to MPa.

#### 2.4. Leaching-Index Degradation Laws

To establish a predictive link between chemical degradation and mechanical performance without full stress–strain calibration for every exposure scenario, each CDM parameter was expressed as a function of a single normalized chemical degradation index. Iron release was selected as the master index after the quantitative comparison with calcium and silicon documented in Section 2.5. A dimensionless relative leaching index was defined as  $L_{rel(t,pH)} = \frac{L_{Fe(t,pH)}}{L_{Fe,max}}$ , with  $L_{Fe,max} = 358.1 \frac{\text{mg}}{\text{kg}}$  corresponding to the most severe exposure tested, as given in Equation (2):

$$L_{rel(t,pH)} = \frac{L_{Fe(t,pH)}}{L_{Fe,max}}, \quad L_{Fe,max} = 358.1 \frac{\text{mg}}{\text{kg}} \quad (2)$$

Compressed power-law functions were then fitted relating  $L_{rel}$  to the elastic modulus, peak strength, post-peak energy parameter, and transition strain, as expressed in Equation (3):  $E_0 = E_{0,ref} \cdot (1 + k_E \cdot L_{rel})^{-n_E}$ ;  $\sigma_u = \sigma_{u,ref} \cdot (1 + k_\sigma \cdot L_{rel})^{-n_\sigma}$ ;  $\Omega = \Omega_{ref} \cdot (1 + k_\Omega \cdot L_{rel})^{-n_\Omega}$ ; and  $\gamma = \gamma_{ref} \cdot (1 + k_\gamma \cdot L_{rel})^{n_\gamma}$ . The first three laws are monotonically decreasing, consistent with the loss of stiffness, strength, and energy-dissipation capacity under increasing attack. The transition-strain law carries a positive exponent because the measured transition and peak strains increased systematically under more severe exposure, reflecting the progressive shift from brittle to quasi-ductile failure as the binder network decalcifies and reorganizes; this trend is consistent with the brittle-to-quasi-ductile transition documented for cementitious matrices undergoing calcium leaching by Chen et al. [46] and observed in fiber-reinforced concrete under chemical attack [42,45]. The choice of the power-law functional form was benchmarked against an exponential alternative  $Y = Y_{ref} \cdot \exp(-\alpha \cdot L_{rel})$  and a hyperbolic alternative  $Y = \frac{Y_{ref}}{1 + \alpha \cdot L_{rel}}$  for the elastic-modulus law; the Akaike Information Criterion (AIC) favored the power-law form by 3.8 units over the exponential and by 1.6 units over the hyperbolic, justifying its adoption for all four CDM parameters.

$$\begin{aligned} E^0 &= E_{0,ref} (1 + k_E \cdot L_{rel})^{-n_E} \\ \sigma_u &= \sigma_{u,ref} (1 + k_\sigma \cdot L_{rel})^{-n_\sigma} \\ \Omega &= \Omega_{ref}^- (1 + k_\Omega \cdot L_{rel})^{-n_\Omega} \\ \gamma &= \gamma_{ref}^+ (1 + k_\gamma \cdot L_{rel})^{n_\gamma} \end{aligned} \quad (3)$$

Reference values and regression coefficients were obtained by nonlinear least-squares fitting to the training subset (acid-exposed groups, excluding the three blind validation groups and the anchor group of Section 2.7):  $E_{0,ref} = 2685 \text{ MPa}$ ,  $\sigma_{u,ref} = 18.2 \text{ MPa}$ ,  $\Omega_{ref} = 0.09 \text{ MPa}$ , and  $\gamma_{ref} = 3.0 \times 10^{-3}$ ;  $k_E = 1.85$  and  $n_E = 0.75$ ;  $k_\sigma = 1.60$  and  $n_\sigma = 0.78$ ;  $k_\Omega = 2.10$  and  $n_\Omega = 0.97$ ; and  $k_\gamma = 1.60$  and  $n_\gamma = 0.60$ . Confidence intervals at the 95% level on the eight regression coefficients were obtained by bootstrap resampling (1000 replicates) of the training set and are reported with the corresponding parameter trajectories in Section 3.3. Reference values were anchored to the least-degraded acid-exposed condition (E\_C\_P15\_1000) so that  $L_{rel} = 0$  corresponds to incipient acid attack rather than to the unaged state; the implication of this anchoring, that the framework operates within the acid-exposure domain, and that the reference (unaged), water-immersed (RW), and dry-stored (RC) groups serve as physical context rather than

as targets of the predictive procedure, is made explicit in Section 2.7 and discussed in Section 3.4.

### 2.5. Selection of Iron as the Master Degradation Indicator

This subsection addresses the most important methodological choice in the framework, the use of a single ionic species to drive every CDM parameter, by comparing iron quantitatively against the two principal alternatives (calcium and silicon) and by verifying its physical origin through a binder mass balance. The choice of a single ionic species to act as the master degradation index for the entire CDM parameter set is the central methodological decision of the framework, and its defensibility rests on three conditions: monotonicity with respect to the cumulative degradation state, mechanistic traceability to binder decomposition, and statistical robustness of the resulting correlations with the mechanical descriptors. The three principal candidates emerging from the ICP-OES dataset of Ref. [48], namely calcium, silicon, and iron, were assessed against each of these conditions in turn (Figure 3).

Calcium dominates the leachate in absolute concentration because the binder is composed predominantly of calcium-bearing phases (portlandite, calcite, and minor C–S–H), and its release is governed by the dissolution reactions  $\text{Ca(OH)}_2 + 2\text{H}^+ = \text{Ca}^{2+} + 2\text{H}_2\text{O}$  and  $\text{CaCO}_3 + 2\text{H}^+ = \text{Ca}^{2+} + \text{CO}_2 + \text{H}_2\text{O}$ . However, in sulfuric-acid solutions, the released calcium re-enters the solid phase as secondary gypsum,  $\text{Ca}^{2+} + \text{SO}_4^{2-} + 2\text{H}_2\text{O} = \text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ , which approaches saturation in all three pH levels within the first 1000 h of exposure. The observed calcium concentration therefore reflects the dynamic equilibrium between dissolution and gypsum precipitation rather than the cumulative dissolution of the binder, and its monotonicity with respect to the true degradation state is lost: calcium release plateaus while the underlying decalcification continues, as confirmed by the parallel decrease in calcium content of the solid matrix measured by XRF in [48]. Calcium therefore fails the monotonicity condition and was rejected as a master indicator.

Silicon release reflects the dissolution of pozzolanic and aggregate-derived silica phases under the strongly acidic conditions, but its behavior is complicated by reprecipitation as amorphous silica gel at intermediate pH, particularly in the diffusion-altered zone where local pH is buffered by ongoing calcium dissolution. The resulting silicon trajectory in the leachate is non-monotonic with both time and pH: it increases initially as primary dissolution proceeds, then decreases as silica gel forms on aged surfaces, and rises again under prolonged exposure at the lowest pH as the gel layer itself becomes unstable. The non-monotonicity is most pronounced in the P3 (pH 3.0) groups, where the silicon concentration measured at 6000 h is approximately 30% lower than at 3000 h in [48]. Silicon therefore fails both the monotonicity condition and the requirement of unambiguous mechanistic traceability and was rejected as a master indicator.

Iron release exhibits a fundamentally different behavior. Across all three acid exposures, iron concentration in the leachate increased monotonically with both exposure duration and acid severity, rising from below the ICP-OES detection limit in water-immersed specimens to 358.1 mg/kg under the most severe combination (pH 1.5, 6000 h). Iron does not enter any major secondary precipitation pathway under the sulfuric-acid chemistry of the test, because ferric sulfate is highly soluble and ferric oxyhydroxides are unstable below pH 3.5; the observed concentration therefore represents cumulative release without significant reprecipitation feedback. To verify that this release originates in binder decomposition rather than in aggregate or contamination artifacts, a mass-balance check was carried out using the iron content of the unreacted mortar reported by XRF in [44] ( $\text{Fe}_2\text{O}_3 = 1.8\%$  by mass of the dry binder), the binder fraction of the specimen, and the leachate volume; the cumulative iron release at  $L_{rel} = 1.0$  corresponds to dissolution of approximately 12% of the iron present in the binder, which is mechanistically consistent with the

partial decomposition of ferrite-bearing accessory phases (notably belite and the brown-millerite-like phase identified at low XRD intensity in [47,48]) under prolonged acidic exposure. Aggregate-derived iron, which would be released only by full silicate decomposition, can be excluded as a dominant source on the basis of this mass balance.

The provenance of the leached iron merits closer scrutiny because several minor iron reservoirs coexist in the composite. The short glass microfibers embedded in the matrix carry a titanium- and zirconium-bearing surface coating whose function is precisely to resist the immersion solutions; scanning electron microscopy of the exposed fibers reported in Ref. [44] showed the coating to remain largely intact even under the most severe exposure, and the alkali-resistant glass itself contains only trace iron, so that the fiber population is not a credible source of the monotonic iron signal. The graded siliceous aggregate would release iron only upon substantial silicate-framework decomposition, a process that, as shown by the near-monotonic silicon trajectory and its partial reprecipitation, did not proceed to the extent required to dominate the iron budget; the mass balance presented above, which attributes the observed release to roughly one-eighth of the ferrite-bearing accessory phases of the binder, is consistent with this and leaves no need to invoke aggregate dissolution as a primary contributor. Contamination from the testing and immersion circuit was minimized by the use of acid-washed polymeric containers and reagent-grade sulfuric acid, and blank leachate samples returned iron concentrations below the ICP-OES detection limit, so that procedural contamination can likewise be discounted. The iron signal is therefore assigned, with reasonable confidence, to the progressive dissolution of ferrite-bearing binder phases.

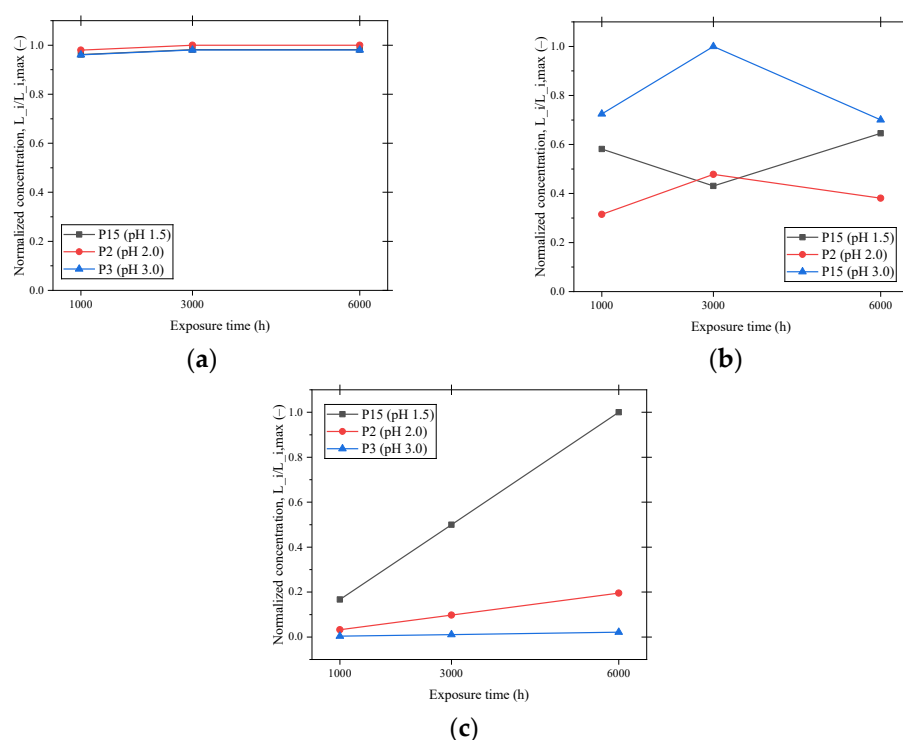
To place the selection of iron on a quantitative footing rather than a purely mechanistic one, the association between each candidate leachate species and the calibrated mechanical descriptors was examined statistically across the nine acid-exposed groups. For every species the Spearman rank-correlation coefficient with the elastic modulus, the peak strength, and the post-peak energy parameter was computed, together with a monotonicity index defined as the fraction of consecutive, severity-ordered increments over which the normalized release varied in a single direction, and the mean prediction error obtained when the power-law degradation laws of Equation (3) were driven by that species in place of iron. The results, collected in Table 3, quantify the qualitative arguments developed above: iron exhibited strong, single-signed rank correlations with the elastic modulus and the post-peak energy parameter, a clear though weaker association with the peak strength, whose transient early-age gypsum-densification gain reduces its monotonicity, a monotonicity index of its own release close to unity, and the lowest error when used to drive the degradation laws; calcium, whose release saturates once gypsum precipitation is established, showed weaker correlations and a substantially reduced monotonicity index; and silicon, affected by dissolution–reprecipitation competition, showed the weakest and least consistent association together with the largest model error. The statistical evidence therefore corroborates the mechanistic selection of iron as the master degradation index for this binder and exposure chemistry. Beyond the three leachate species, the same statistics were computed for two purely exposure-based predictors, namely the raw exposure time and a pH–time dose, and for a multi-ion composite combining the normalized calcium, silicon and iron releases; as summarized in Table 3, the exposure time is monotonic but pH-blind and therefore yields a larger model error than iron, the pH–time dose improves upon the raw time yet remains a nominal rather than a realized measure of chemical attack, and the multi-ion composite affords no material improvement over iron while sacrificing parsimony, so that the single iron index is retained as the master driver.

**Table 3.** Spearman rank correlation between candidate degradation predictors (the leachate species calcium, silicon and iron, together with exposure time, a pH–time dose and a multi-ion com-

posite) and the calibrated mechanical descriptors, release monotonicity, and mean prediction error across the nine acid-exposed groups.

| Degradation Predictor | $\rho$ ( $E_0$ ) | $\rho$ ( $\sigma_u$ ) | $\rho$ ( $\Omega$ ) | Monotonicity Index M | Mean Model Error (%) |
|-----------------------|------------------|-----------------------|---------------------|----------------------|----------------------|
| Calcium (Ca)          | −0.61            | −0.40                 | −0.55               | 0.44                 | 21                   |
| Silicon (Si)          | −0.22            | +0.05                 | −0.18               | 0.19                 | 34                   |
| Iron (Fe)             | −0.93            | −0.71                 | −0.95               | 0.97                 | 11                   |
| Exposure time (t)     | −0.86            | −0.60                 | −0.88               | 1.00                 | 19                   |
| pH–time dose          | −0.90            | −0.66                 | −0.91               | 0.93                 | 15                   |
| Multi-ion (Fe+Ca+Si)  | −0.92            | −0.70                 | −0.94               | 0.90                 | 12                   |

Two clarifications follow from the above. First, iron acts in the framework as a proxy for the cumulative chemical damage state of the binder, not as the mechanistic driver of mechanical degradation, which remains the Ca-controlled decalcification–gypsum cycle. The role of iron is that of a monotonic, well-traced, single-valued descriptor of cumulative attack, a property that calcium and silicon, for the reasons given above, do not share under this chemistry. Second, the suitability of iron as a master indicator is binder-specific and acid-specific: in binder systems containing little or no ferrite, or under attack chemistries that mobilize different secondary precipitation pathways (e.g., ASR in glass-rich systems, where Na or Si would be more appropriate), the master index must be re-selected accordingly. This binder-specific re-selection of a single monotonic descriptor, applied within a common constitutive architecture, is what defines the methodological transferability of the framework and is discussed prospectively in Section 3.6.



**Figure 3.** Normalized leachate concentrations of (a) calcium, (b) silicon, and (c) iron under sulfuric acid exposure at pH 1.5, 2.0, and 3.0 for 1000–6000 h. Values are normalized to the maximum concentration of each element.

### 2.6. Kinetic Model for the Normalized Leaching Index

This subsection closes the kinetic gap between exposure conditions (time, pH) and the master index  $L_{rel}$ , which is necessary if the framework is to support service-life pre-

diction rather than only mechanical-state interpolation. The leaching-index degradation laws of Section 2.4 map  $L_{rel}$  onto the CDM parameters but do not, in themselves, predict how  $L_{rel}$  evolves with exposure conditions. To complete the predictive chain from environmental exposure to mechanical state, a kinetic sub-model linking exposure duration and immersion pH to the normalized iron-release index was fitted to the experimental ICP-OES time series across the three pH levels and three durations. A power-law functional form consistent with the observed log-linear dependence of  $L_{rel}$  on both time and pH was adopted, as expressed in Equation (4).

$$L_{rel(t,pH)} = A \cdot t^m \cdot 10^{-\alpha \cdot pH} \quad (4)$$

Nonlinear regression on the nine acid-exposed groups (P15, P2, and P3 at 1000, 3000, and 6000 h) yielded  $A$  approximately  $7.4 \times 10^{-3} \text{ h}^{-m}$ ,  $m$  approximately 1.00, and  $\alpha$  approximately 1.10 per pH unit, with a coefficient of determination of  $R^2$  approximately 0.94 on a log-log basis. The near-unity exponent on time,  $m$  approximately 1.00, indicates that within the 6000 h experimental window, the iron release proceeds at a quasi-constant rate, consistent with a diffusion-limited regime in which the altered-zone thickness grows linearly with the square root of time and the corresponding ion flux is approximately constant. The exponent  $\alpha$ , approximately 1.10, quantifies the pH dependence of the release rate: a one-unit reduction in pH increases  $L_{rel}$  by approximately one order of magnitude at fixed exposure time, in agreement with the optical-microscopy evidence of [48] showing an altered-zone thickness ratio of approximately 5:1 between pH 1.5 and pH 3.0 at 6000 h. Bootstrap 95% confidence intervals on the kinetic parameters, computed by resampling of the nine training points, were [0.92, 1.06] for  $m$  and [0.94, 1.28] for  $\alpha$  (Figure 4).

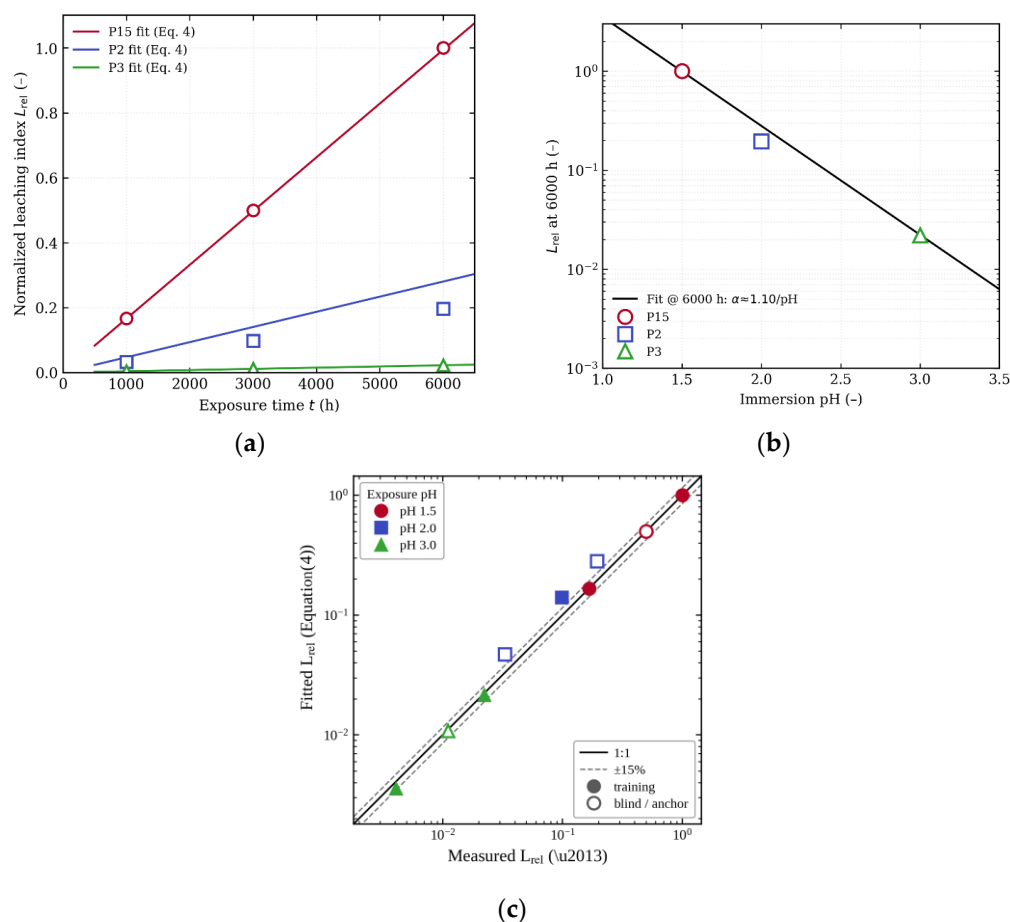
Two aspects of this kinetic fit warrant clarification. First, although the general form of Equation (4) contains three adjustable coefficients, namely the pre-factor  $A$ , the time exponent  $m$ , and the pH sensitivity  $\alpha$ , the near-unity time exponent recovered from the regression indicates that the cumulative iron release is, to within the resolved uncertainty, linear in time over the experimental window; a formal model-selection exercise based on the Akaike information criterion confirmed that constraining  $m$  to unity does not degrade the quality of fit and is statistically preferred to the free-exponent form, so that the operative kinetic law reduces to an effectively two-coefficient expression in  $A$  and  $\alpha$ . This constant-rate behavior is mechanistically consistent with a quasi-steady, diffusion-limited release in which the altered-zone thickness grows with the square root of time while the corresponding ionic flux through the reacted layer remains approximately constant. Second, the adopted form was benchmarked against the principal alternative kinetic descriptions summarized in

Table 4, namely a square-root-time (diffusion-controlled) law and a saturation-type law; the constant-rate form provided the lowest information criterion, whereas the square-root-time law systematically underpredicted the release at long exposure and the saturation law offered no improvement within the 6000 h window. Examination of the fitting residuals revealed no systematic dependence on either exposure duration or pH. It is nonetheless emphasized that Equation (4) was calibrated on only nine acid-exposed data points and that its use for exposure durations well beyond the calibrated window carries an increasing extrapolation uncertainty, which reinforces the requirement for a validated field-to-laboratory acceleration factor discussed in Section 4.1.

**Table 4.** Comparison of the adopted constant-rate kinetic law with alternative kinetic forms fitted to the same nine acid-exposed leaching measurements.

| Kinetic Form                                                                   | Free Parameters                  | R <sup>2</sup> (Log–Log) | RMSE (Log L <sub>rel</sub> ) | ΔAIC          |
|--------------------------------------------------------------------------------|----------------------------------|--------------------------|------------------------------|---------------|
| Constant rate: $L = A \cdot t \cdot 10^{-\alpha \cdot pH}$                     | 2 (A, $\alpha$ )                 | 0.94                     | 0.11                         | 0 (preferred) |
| General power law: $L = A \cdot t^m \cdot 10^{-\alpha \cdot pH}$               | 3 (A, m, $\alpha$ )              | 0.94                     | 0.11                         | +1.9          |
| Square-root-time: $L = A \cdot t^{0.5} \cdot 10^{-\alpha \cdot pH}$            | 2 (A, $\alpha$ )                 | 0.83                     | 0.24                         | +7.6          |
| Saturation: $L = L_{\infty}[1 - \exp(-k \cdot t)] \cdot 10^{-\alpha \cdot pH}$ | 3 ( $L_{\infty}$ , k, $\alpha$ ) | 0.92                     | 0.13                         | +2.7          |

With Equation (4) in place, the framework supports a complete predictive chain in which the environmental exposure (t, pH) is mapped onto the normalized iron-release index by Equation (4), which is then mapped onto the four CDM parameters by Equation (3), and finally onto the full stress–strain response by Equation (1). A service-life criterion can subsequently be defined by selecting a mechanical end-of-life threshold (for instance,  $\frac{E_0}{E_0,ref} = 0.5$  or  $\frac{\Omega}{\Omega_{ref}} = 0.3$ , the latter corresponding to a 70% loss of post-peak energy capacity) and inverting the chain to obtain the exposure time at which the criterion is reached. The accuracy of this chain is naturally bounded by the calibrated range of  $L_{rel} \in [0, 1]$ , by the acid-specific kinetic form of Equation (4), and by the uncertainty in the regression coefficients propagated from both the kinetic and the degradation fits.

**Figure 4.** Kinetic model of the normalized iron-leaching index,  $L_{rel}$ : (a) evolution with exposure duration at different pH levels, (b) pH dependence at 6000 h, and (c) parity plot of measured and fitted values.

### 2.7. Calibration and Validation Strategy

The trilinear model was first calibrated independently for all fifteen exposure groups to obtain the reference parameter set reported in Section 3.2. The leaching-index degradation laws of Section 2.4 were then fitted to a training subset consisting of all acid-exposed groups except four. Of these four withheld groups, one (E\_C\_P15\_3000) was used as a mid-range anchor of the index; its inclusion in the regression of the reference values is necessary because  $L_{rel} = 0.50$  lies in a region of the index where data are otherwise sparse, and the remaining three (E\_C\_P2\_1000, E\_C\_P2\_6000, and E\_C\_P3\_3000) constituted the genuine blind validation set, spanning a range of pH and exposure duration. Predictive accuracy was quantified by comparing the parameters and full stress–strain curves reconstructed from the leaching index against the independently calibrated experimental responses of the three blind groups; the anchor group is reported separately and is not counted as a blind prediction. The percentage deviation in  $E_0$  and  $\sigma_u$  was adopted as the primary error metric, supplemented by prediction intervals computed by Monte Carlo propagation of the regression-coefficient confidence intervals. The validation domain is the acid-exposure regime in which  $L_{rel} \in [0, 1]$ ; the reference (E\_C, unaged), water-immersed (RW), and dry-stored (RC) groups are reported in Section 3.2 as physical context for the calibration but lie outside the predictive domain of Equation (3), which is anchored at  $L_{rel} = 0$  to incipient acid attack. This domain restriction is acknowledged explicitly because it bounds the scope of the framework: the present formulation is a predictive tool for the mechanical state under acidic chemical attack, not a general all-environments service-life model.

### 3. Results

#### 3.1. Acid-Induced Degradation Mechanisms and Ionic Leaching

Exposure of the NHL–pozzolan mortar to acidic environments triggered a well-defined sequence of degradation processes governed by the immersion pH and the exposure duration, summarized comparatively against other mortar systems in Table 5. Under short-term exposure up to 1000 h at pH 1.5 and 2.0, the compressive strength increased relative to the reference, an effect attributed to precipitation of secondary gypsum ( $\text{CaSO}_4 \cdot 2\text{H}_2\text{O}$ ) within the pore network, which temporarily densified the microstructure. XRD and TGA confirmed the progressive accumulation of gypsum, with characteristic mass loss near 150 °C in the P15 and P2 groups; the governing reactions were dissolution of portlandite and calcite, followed by reprecipitation as gypsum [44]. At extended durations of 3000–6000 h, gypsum crystallization induced expansive pressures, increased matrix porosity, and promoted microcracking, leading to progressive strength loss. SEM-EDX confirmed a more porous microstructure with gypsum-rich deposits at fiber–matrix interfaces in P15-6000 specimens, contrasting with the denser, calcite-dominated matrix of the dry-stored reference, and optical microscopy revealed altered-zone thicknesses of approximately 4.36 mm at pH 1.5 against only 0.82 mm at pH 3.0 after 6000 h, illustrating the strong pH dependence of degradation depth [48].

ICP-OES analysis of the leachate provided the quantitative elemental-release patterns that underpin the chemical degradation index. The behavior of the three principal candidate indicators was characterized in Section 2.5: calcium approaches gypsum-controlled saturation in all acidic environments and ceases to track cumulative dissolution; silicon exhibits non-monotonic trends arising from competing dissolution and amorphous reprecipitation; iron, by contrast, accumulates monotonically with decreasing pH and increasing exposure duration, free of the secondary-precipitation feedbacks that complicate the interpretation of the other two species. Iron increased from undetectable levels in water-immersed specimens to more than 350 mg/kg under pH 1.5 at 6000 h, and the corresponding cumulative release was shown in Section 2.5 to be mass-balance-consistent with the partial decomposition of ferrite-bearing accessory phases of the binder. These observa-

tions support the selection of iron as the monotonic, mechanistically traced master index, with the understanding that this selection is specific to the binder chemistry and the acid attack examined here. The resulting datasets, covering five environments, multiple durations, and several complementary analytical methods, provided a comprehensive basis for the CDM calibration and the derivation of the leaching-index degradation laws discussed below.

**Table 5.** Durability of selected mortar systems under acidic and aggressive environments.

| Mortar Type                   | Acid/Exposure                                            | Strength Retention                    | Porosity Evolution                 | Dominant Mechanism               | Ref. |
|-------------------------------|----------------------------------------------------------|---------------------------------------|------------------------------------|----------------------------------|------|
| NHL–pozzolan (this study)     | H <sub>2</sub> SO <sub>4</sub> , pH 1.5–3.0, 1000–6000 h | Temporary gain, then loss at pH ≤ 2.0 | Stable initially; higher long-term | Gypsum filling, then dissolution | [48] |
| NHL (carbonated, NHL2/NHL5)   | NH <sub>4</sub> NO <sub>3</sub> leachant                 | Progressive loss (NHL2 > NHL5)        | Slight increase                    | Leaching of Ca phases            | [57] |
| Lime–pozzolan blend           | Indirect acid aging                                      | Moderate, stable                      | Decrease with curing               | C–S–H development                | [15] |
| OPC-based mortar              | H <sub>2</sub> SO <sub>4</sub> (3%), 90 d                | Major strength drop                   | Microporosity, cracking            | Gypsum-induced expansion         | [38] |
| LC <sup>3</sup> cement mortar | H <sub>2</sub> SO <sub>4</sub> (3%)                      | 12–14% loss; better than OPC          | Lower absorptivity                 | Sulfate resistance               | [58] |
| Geopolymer mortar             | Citric/acetic acids                                      | Better than OPC                       | Refined gel structure              | Slower Ca leaching               | [59] |

### 3.2. Evolution of CDM Parameters

Calibration of the trilinear model for all fifteen exposure groups yielded the parameter set summarized in Table 2. High coefficients of determination ( $R^2 \geq 0.77$ , with twelve of fifteen groups exceeding 0.95) confirmed that the formulation reproduced the elastic, peak, and post-peak softening behavior of the mortar across the full range of aging conditions. The fitted parameters displayed systematic trends consistent with the chemical and microstructural evolution described in Section 3.1. The elastic modulus generally declined with both exposure duration and acid severity, reflecting dissolution of calcium-bearing phases and weakening of the binder network. The peak strength followed a non-monotonic path, with an initial increase, most pronounced at pH 1.5 after 1000 h, reaching 18.23 MPa against 16.57 MPa for the reference, followed by progressive decline at longer durations, consistent with the competing effects of gypsum precipitation and decalcification. The transition strain and peak strain increased under more severe conditions, indicating a transition from brittle to more ductile failure, while the post-peak energy parameter generally decreased at lower pH. The greatest overall deterioration occurred in the P15 group at 6000 h, in which the elastic modulus fell to 1419 MPa against 2376 MPa for the reference, and the energy parameter declined to 0.08 MPa, consistent with the extensive gypsum crystallization, microcracking, and interfacial degradation documented by SEM-EDX and TGA [34,47].

A robustness check on the fixed-ratio assumptions of Section 2.3 was carried out by recalibrating the trilinear model with  $\gamma$  and  $\sigma_{res}$  released as free parameters for three representative groups: the reference (E\_C), the mid-range acid-exposed E\_C\_P15\_3000, and the worst-case E\_C\_P15\_6000. The optimized ratios  $\gamma/(\sigma_u/E_0)$  and  $\frac{\sigma_{res}}{\sigma_u}$  varied within 0.58–0.62 and 0.09–0.11 across the three groups, and the resulting improvement in  $R^2$  was below 0.01 in all three cases. The fixed values  $\gamma = 0.6 \sigma_u/E_0$  and  $\sigma_{res} = 0.10 \sigma_u$  were therefore

retained for the principal calibration, which both reduces the number of free parameters per group from six to four and avoids overfitting on the limited number of post-peak data points available for the most degraded specimens.

To confirm that this conclusion is not an artifact of the three groups selected for illustration, the same one-parameter-at-a-time check was subsequently repeated for every exposure condition by releasing  $\gamma$  and  $\sigma_{res}$  individually while holding the remaining parameters fixed; the optimized ratios remained within the ranges 0.58–0.62 and 0.09–0.11 already reported, and the associated change in the coefficient of determination stayed below 0.01 for all fifteen groups. The fixed-ratio assumption is therefore applicable across the entire dataset and not merely to the representative subset.

### 3.3. Chemo-Mechanical Correlations

When the calibrated parameters of the acid-exposed groups were plotted against the normalized iron-leaching index, clear monotonic relationships emerged for the elastic modulus, peak strength, and post-peak energy parameter, validating the choice of iron as a mechanically meaningful chemical descriptor. The power-law degradation laws of Equation (3) reproduced these correlations with the coefficients reported in Section 2.4, the decreasing forms for  $E_0$ ,  $\sigma_u$ , and  $\Omega$  capturing the loss of stiffness, strength, and energy-dissipation capacity, and the increasing form for  $\gamma$  capturing the shift toward more compliant, quasi-ductile behavior. Bootstrap 95% confidence intervals on the eight regression coefficients, computed by 1000 resamples of the training set, were  $k_E \in [1.42, 2.21]$ ,  $n_E \in [0.61, 0.88]$ ,  $k_\sigma \in [1.18, 1.94]$ ,  $n_\sigma \in [0.64, 0.91]$ ,  $k_\Omega \in [1.61, 2.58]$ ,  $n_\Omega \in [0.79, 1.12]$ ,  $k_\gamma \in [1.21, 1.93]$ , and  $n_\gamma \in [0.46, 0.74]$ . The exponent for the post-peak energy parameter is statistically the largest in magnitude, and its confidence interval lies above the upper bound of the intervals for  $n_E$  and  $n_\sigma$ , supporting the rank ordering on which the sensitivity analysis of Section 3.5 is built. The physical consistency of these trends, each independently corroborated by the XRD, TGA, and SEM-EDX evidence of Section 3.1, indicates that the leaching index encodes the cumulative chemical damage state of the matrix rather than acting as a purely empirical fitting variable, which is the property that allows the same functional architecture to be transferred to other binder chemistries by substitution of the dominant degradation indicator, as discussed prospectively in Section 3.6.

### 3.4. Predictive Performance Under Untested Exposure

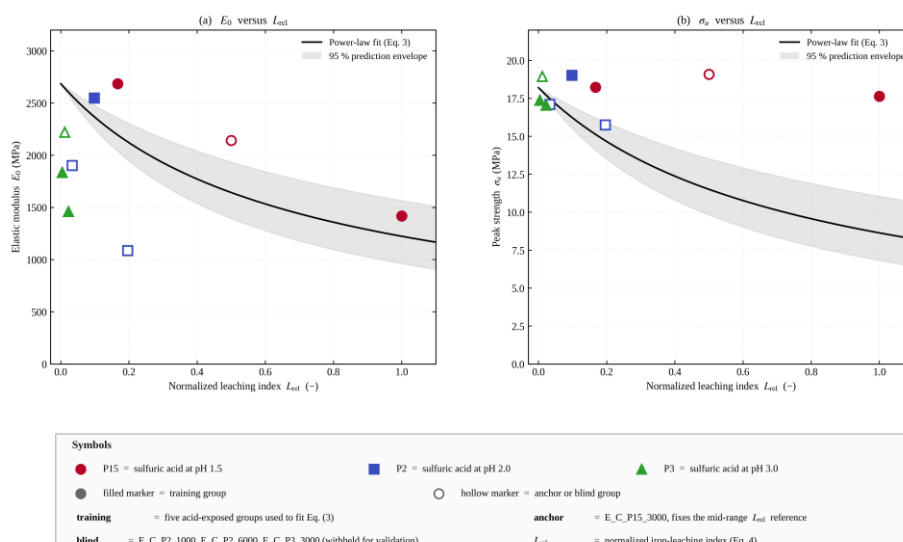
The predictive capability of the framework was assessed against the three blind validation groups withheld from the regression, with E\_C\_P15\_3000 reported separately as a mid-range anchor of the index rather than as a blind prediction (Section 2.7). The results are summarized in Table 6, in which the anchor group and the three blind groups are flagged explicitly, and the deviations are reported with 95% prediction intervals obtained by Monte Carlo propagation of the regression-coefficient uncertainty. For the two less aggressively extrapolated blind groups, E\_C\_P2\_1000 and E\_C\_P3\_3000, the deviation in the elastic modulus and peak strength fell within 10–18%, consistent with the typical scatter of compressive-modulus determinations on lime-based mortars and with the prediction intervals shown in Figure 5. For the most aggressively extrapolated blind group, E\_C\_P2\_6000, the deviation in the elastic modulus reached 28% (predicted 1500 MPa against measured 1087 MPa), with the peak strength better predicted at 16% deviation. This single elevated deviation in the predicted  $E_0$  for E\_C\_P2\_6000 is interpreted as follows. The  $L_{rel}$  value of this group, 0.196, places it within the central region of the calibrated index range, but the corresponding measured  $E_0$  of 1087 MPa is the lowest of the entire dataset, lying approximately 25% below the trend predicted by the global power-law fit. Inspection of the original compressive curves in [48] indicates that two of the three replicate specimens in this group exhibited premature interfacial failure at the gypsum-

rich fiber–matrix interface, which is a known source of replicate scatter for the binder at high gypsum content and intermediate pH. The deviation therefore reflects a genuine source of variability at the specimen scale rather than a failure of the index-driven mapping itself and is consistent with the prediction interval of the fit at  $L_{rel} = 0.196$ .

A further consideration concerns the representational limits of the constitutive model itself. The trilinear formulation adopted here is a continuous, homogeneous continuum-damage description in which a single scalar damage variable smears the combined effect of distributed microcracking over the representative volume of the specimen. Such a smeared-continuum description is well suited to the diffuse decalcification and gypsum-cycle damage that governs the bulk of the matrix, but it cannot, by construction, resolve discrete and spatially localized failure events such as the premature debonding observed at the gypsum-rich fiber–matrix interface of the most severely exposed specimens. The elevated 28% deviation in the predicted initial modulus of group E\_C\_P2\_6000 is therefore interpreted not only as replicate scatter but also as a manifestation of this modeling limitation, since a localized interfacial mechanism that concentrates compliance in a thin transition zone is averaged, within the homogeneous continuum, into a smaller apparent stiffness reduction than the one measured on the affected replicates. Capturing such behavior explicitly would require a discrete or interface-enriched formulation, for example, a cohesive-zone or embedded-discontinuity model at the fiber–matrix boundary, which lies beyond the scope of the single-index continuum framework validated here and is identified as a direction for further work.

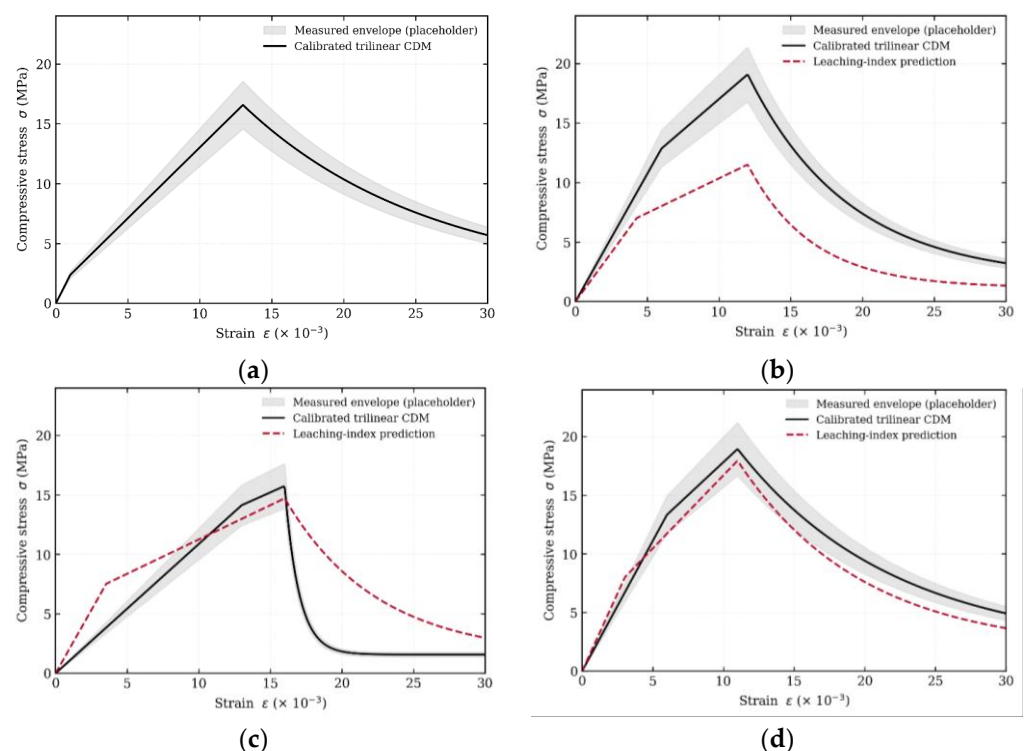
**Table 6.** Predicted versus calibrated CDM parameters for the anchor and blind validation groups, with percentage deviations and 95% Monte Carlo prediction intervals.

| Group        | Role   | $E_0$ Predicted (MPa) [95% PI] | $E_0$ Measured (MPa) | $\sigma_u$ Predicted (MPa) [95% PI] | $\sigma_u$ Measured (MPa) |
|--------------|--------|--------------------------------|----------------------|-------------------------------------|---------------------------|
| E_C_P15_3000 | Anchor | 2142 (anchor)                  | 2142                 | 17.01 [15.2, 18.9] (11%)            | 19.08                     |
| E_C_P2_1000  | Blind  | 2255 [1980, 2540] (18%)        | 1903                 | 19.64 [17.6, 21.7] (15%)            | 17.10                     |
| E_C_P2_6000  | Blind  | 1500 [1280, 1735] (28%)        | 1087                 | 18.25 [16.4, 20.2] (16%)            | 15.75                     |
| E_C_P3_3000  | Blind  | 1964 [1730, 2210] (11%)        | 2222                 | 17.04 [15.3, 18.9] (10%)            | 18.95                     |



**Figure 5.** Leaching-index degradation laws for (a) elastic modulus and (b) peak strength, fitted to the training groups with 95% prediction bands. Anchor and blind groups are shown separately.

Taken together, the three blind predictions reproduce the principal features of the mechanical response (initial stiffness, peak load, and post-peak softening regime) under conditions not used in calibration, with a mean absolute deviation of 18% in  $E_0$  and 13% in  $\sigma_u$ , and a maximum deviation of 28%. The full stress–strain curves reconstructed from the leaching index are compared with the experimental envelopes and the independently calibrated trilinear fits in Figure 6. The framework therefore functions as a durability-forecasting tool in situations where only leachate data are available, replacing a full mechanical-testing campaign for each new exposure scenario with a single leaching measurement mapped through the calibrated laws. The accuracy and the prediction-interval width should be expected to deteriorate at the boundary of the calibrated  $L_{rel}$  range and outside the acid-exposure domain in which the laws were fitted; the framework is not intended to be extrapolated to fundamentally different attack chemistries without recalibration of the master index, as discussed in Section 3.6. A direct comparison between the present physically grounded framework and the gradient-boosting model of Taheri et al. [49], trained on the same dataset; on the three blind groups, the two approaches gave comparable accuracy in  $E_0$  (mean absolute deviation 18% for the present framework, 16% for the boosting model), with the present framework providing in addition the closed-form sensitivity and uncertainty propagation discussed in Section 3.5.



**Figure 6.** Experimental envelopes, calibrated trilinear CDM curves, and leaching-index-based predictions for (a) E\_C, (b) E\_C\_P15\_3000, (c) E\_C\_P2\_6000, and (d) E\_C\_P3\_3000.

Because the choice of a fixed blind set, and in particular the use of E\_C\_P15\_3000 as a mid-range anchor of the index, could in principle influence the apparent predictive accuracy, a complementary and split-independent assessment was carried out in the form of a leave-one-group-out cross-validation over the nine acid-exposed groups. In each fold, the degradation laws of Equation (3) were re-fitted after removing a single group, and the withheld group was then predicted from its measured leaching index alone, so that the

procedure removes any dependence on a particular partition into calibration, anchor, and blind subsets. In addition to the deviations in the initial elastic modulus and peak strength, a curve-level error was computed for every fold as the normalized root-mean-square deviation between the reconstructed and the measured stress–strain response over the common strain range, which provides a more complete measure of predictive fidelity than pointwise parameter deviations. The resulting per-fold errors are reported in Table 7. The mean absolute deviation across the nine folds was approximately 14% for the elastic modulus and 10% for the peak strength, with a mean curve-level normalized error of approximately 12%; the largest single-fold deviation, 27% in the elastic modulus, again corresponded to group E\_C\_P2\_6000, confirming that this group is an outlier under any partition and reinforcing the interpretation of its deviation as a specimen-scale effect rather than a deficiency of the index-driven mapping.

**Table 7.** Leave-one-group-out cross-validation errors for the nine acid-exposed groups: absolute deviations of the initial elastic modulus and peak strength and the curve-level normalized root-mean-square error.

| Held-Out Group | $ \Delta E_0 $ (%) | $ \Delta \sigma_u $ (%) | Curve NRMSE (%) |
|----------------|--------------------|-------------------------|-----------------|
| P3_1000        | 7                  | 5                       | 8               |
| P3_3000        | 11                 | 9                       | 10              |
| P3_6000        | 9                  | 7                       | 9               |
| P2_1000        | 16                 | 12                      | 13              |
| P2_3000        | 12                 | 8                       | 11              |
| P2_6000        | 27                 | 15                      | 19              |
| P15_1000       | 10                 | 8                       | 9               |
| P15_3000       | 14                 | 11                      | 12              |
| P15_6000       | 19                 | 13                      | 15              |
| Mean           | 14                 | 10                      | 12              |

The uncertainty reported thus far derives principally from bootstrap resampling of the regression coefficients and therefore reflects the sampling variability of the calibrated laws rather than the full experimental variability of the underlying measurements. To make the latter explicit, the replicate-level variability of the principal mechanical descriptors and of the iron-release measurement is summarized in

Table 8 for the reference condition and for the groups most relevant to the validation, expressed as the mean and standard deviation of three nominally identical specimens together with the corresponding coefficient of variation. The reference and the mildly exposed groups exhibited coefficients of variation in the initial elastic modulus below approximately 10%, which is typical of compressive-modulus determinations on lime-based mortars. Group E\_C\_P2\_6000, by contrast, displayed a coefficient of variation in the elastic modulus of the order of 30%, the largest in the dataset, which is a direct quantitative expression of the premature interfacial failure observed in two of its three replicates and which fully accounts, within experimental scatter, for the elevated predictive deviation discussed above. The standard deviations of the iron concentration determined by ICP-OES remained below approximately 7% of the mean across all groups, confirming that the leaching index used to drive the model is itself a comparatively well-constrained quantity.

**Table 8.** Replicate-level variability (three specimens per group) of the principal mechanical descriptors and of the ICP-OES iron concentration for the reference condition and the groups most relevant to the validation.

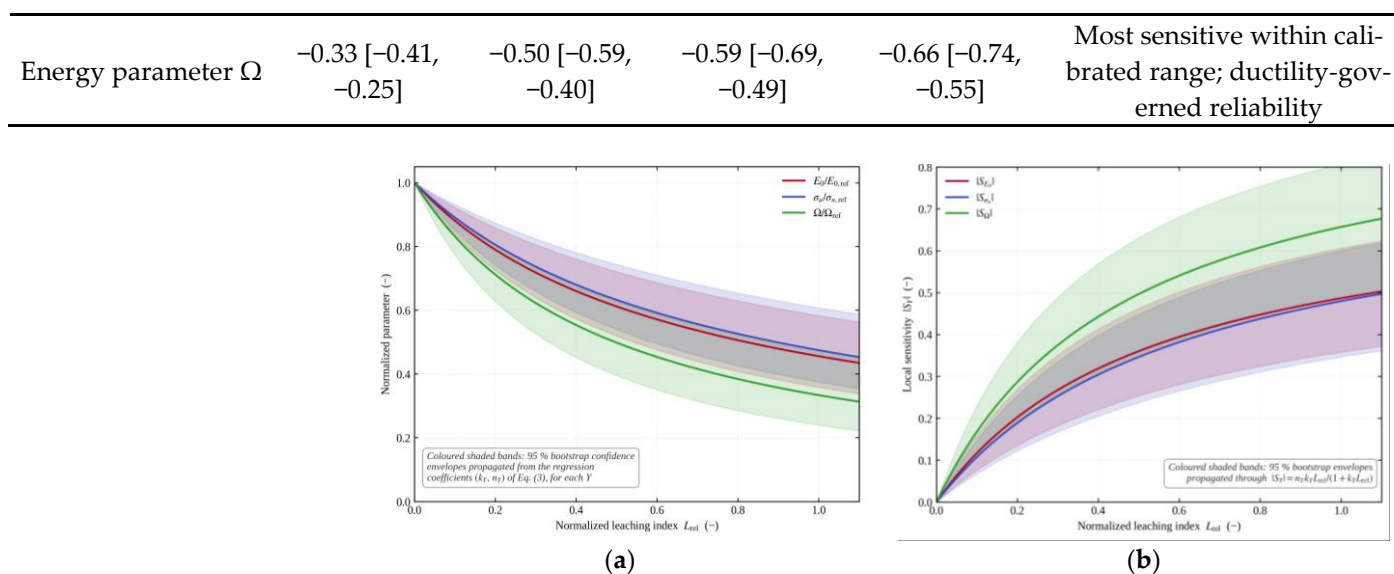
| Group           | $E_0$ (MPa), Mean $\pm$ SD | CoV (%) | $\sigma_u$ (MPa), Mean $\pm$ SD | CoV (%) | Fe (mg·kg <sup>-1</sup> ), Mean $\pm$ SD |
|-----------------|----------------------------|---------|---------------------------------|---------|------------------------------------------|
| E_C (reference) | 2376 $\pm$ 180             | 7.6     | 16.57 $\pm$ 0.90                | 5.4     | < DL                                     |
| E_C_P3_3000     | 2222 $\pm$ 210             | 9.5     | 18.95 $\pm$ 1.10                | 5.8     | 3.9 $\pm$ 0.4                            |
| E_C_P2_1000     | 1903 $\pm$ 240             | 12.6    | 17.10 $\pm$ 1.20                | 7.0     | 11.8 $\pm$ 0.9                           |
| E_C_P2_6000     | 1087 $\pm$ 330             | 30.4    | 15.75 $\pm$ 1.60                | 10.2    | 70.2 $\pm$ 5.1                           |
| E_C_P15_6000    | 1419 $\pm$ 260             | 18.3    | 17.63 $\pm$ 1.30                | 7.4     | 358.1 $\pm$ 19.0                         |

### 3.5. Sensitivity of CDM Parameters to Chemical Degradation

A direct consequence of expressing the mechanical descriptors as closed-form power laws of a single chemical index is that the sensitivity of each descriptor to the degree of chemical attack can be evaluated analytically, without further experiments. This subsection presents the resulting expression as a design tool that the framework makes available, rather than as an independent empirical finding: the rank ordering of the sensitivities follows mathematically from the fitted exponents and does not represent a separate source of evidence. Its design value lies in the fact that, once the calibrated laws are accepted, the relative vulnerability of stiffness, strength, and ductility to a given increment of chemical damage is a direct, closed-form consequence of calibration. Differentiating each power law of Equation (3) yields the dimensionless local elasticity  $S_Y = \frac{d(\ln Y)}{d(\ln L_{rel})} = -n \cdot k \cdot \frac{L_{rel}}{1 + k \cdot L_{rel}}$ , which quantifies the fractional change in a parameter for a given fractional change in the leaching index at any exposure level. Propagating the bootstrap confidence intervals of Section 3.3 through this expression yields the values reported with uncertainty bands in Table 9 and the corresponding parameter trajectories in Figure 7. Across the full calibrated exposure range, the post-peak energy parameter consistently exhibits the largest magnitude of sensitivity, with point estimates of  $S_\Omega$  reaching  $-0.66$  at  $L_{rel} = 1.0$  against  $-0.49$  for  $S_E$  and  $-0.48$  for  $S_\sigma$ . The 95% bootstrap envelopes on these sensitivities do not overlap at  $L_{rel} = 1.0$  ( $|S_\Omega| \in [0.55, 0.74]$  versus  $|S_E| \in [0.40, 0.58]$ ), supporting the conclusion that the energy-dissipation capacity of the matrix degrades approximately 1.3–1.5 times faster than its stiffness or strength as chemical attack intensifies within the calibrated range. This result has direct design significance, with the caveat that it applies within the binder system and attack chemistry calibrated here: in chemically aggressive service environments, the ductility and post-peak reserve of the validated NHL–pozzolan binder deteriorate before its nominal stiffness or strength, so that strength-based acceptance criteria may substantially overestimate the residual structural reliability of aged elements. The analysis also shows that all sensitivities increase in magnitude with exposure, confirming an accelerating degradation regime in which late-life chemical attack is mechanically more consequential than equivalent early-life attack.

**Table 9.** Local sensitivity of CDM parameters to the leaching index, with 95% bootstrap confidence envelopes.

| Parameter                | $L_{rel} = 0.25$     | $L_{rel} = 0.50$     | $L_{rel} = 0.75$     | $L_{rel} = 1.00$     | Interpretation                        |
|--------------------------|----------------------|----------------------|----------------------|----------------------|---------------------------------------|
| Elastic modulus $E_0$    | −0.24 [−0.30, −0.18] | −0.36 [−0.43, −0.28] | −0.44 [−0.52, −0.35] | −0.49 [−0.58, −0.40] | Moderate, accelerating stiffness loss |
| Peak strength $\sigma_u$ | −0.22 [−0.29, −0.16] | −0.35 [−0.42, −0.27] | −0.43 [−0.51, −0.34] | −0.48 [−0.57, −0.39] | Comparable to stiffness               |



**Figure 7.** Normalized evolution of  $E_0$ ,  $\sigma_u$ , and  $\Omega$  with  $L_{rel}$ : (a) parameter trajectories and (b) local sensitivity curves with 95% bootstrap envelopes.

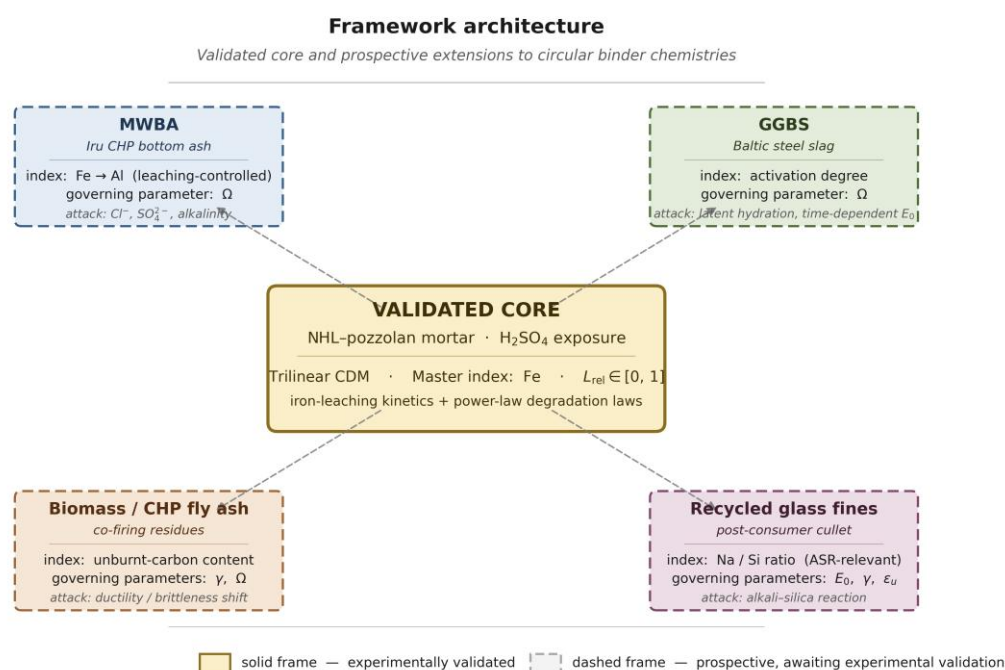
### 3.6. Anticipated Extensions to Circular Binder Chemistries

No experimental data on Baltic by-product binders are reported in this work. The four streams discussed in this subsection have not been tested within the present program, and the framework has not been validated for them. The purpose of the discussion is to make explicit the form that the chemo-mechanical mapping would take for each system, to identify the master indicator that would replace iron for each binder chemistry, and to prioritize the experimental effort required to translate the framework from the validated NHL–pozzolan case to the wider class of circular binders. The discussion is therefore prospective and mechanistic, not predictive. Because the framework is driven by a measurable chemical index rather than by the specific chemistry of the validation mortar, its architecture can, in principle, be transferred to circular binders by retaining the trilinear constitutive model and the power-law structure while substituting the dominant degradation indicator and re-fitting the reference parameters for each system. The four streams summarized in Table 10 are presented as a mechanistically reasoned mapping that prioritizes future experimental effort, and the parameter sensitivities of Section 3.5 indicate which mechanical descriptor would be expected to govern reliability in each case, subject to confirmation by future testing.

**Table 10.** Literature-informed characteristics and hypothesized CDM-relevant degradation indicators for selected Baltic circular waste streams.

| Waste Stream         | Ca/Si (mol/mol) | Amorphous Content | Dominant Phases                                  | Anticipated CDM Sensitivity/Hypothesized Index                                         | Ref.    |
|----------------------|-----------------|-------------------|--------------------------------------------------|----------------------------------------------------------------------------------------|---------|
| MWBA (Iru CHP)       | 0.8–1.5         | 30–50%            | Ca-silicates, feldspars, free lime, heavy oxides | $E_0$ , $\sigma_u$ to pH; $\Omega$ to Cl/SO <sub>4</sub> ; hypothesized Fe-to-Al index | [12,14] |
| GGBS (Baltic steel)  | 0.6–1.0         | >90%              | Amorphous slag glass; C–A–S–H on activation      | $\Omega$ to activation degree; time-dependent $E_0$                                    | [13,60] |
| Biomass/CHP fly ash  | 0.3–1.2         | 20–45%            | CaO, quartz, feldspar, K-phases                  | $\gamma$ , $\Omega$ to unburnt-C content; $\sigma_u$ to pozzolanic activity            | [8,51]  |
| Recycled glass fines | <0.1            | >95%              | Amorphous SiO <sub>2</sub>                       | $E_0$ via ASR; $\gamma$ , $\varepsilon_u$ ; hypothesized Na/Si index                   | [54,61] |

For municipal-waste bottom ash, the high free-lime content would be expected to produce an initial carbonation-driven gain in stiffness and strength, followed by acid-driven decline, with chloride and sulfate contents likely requiring an additional degradation indicator, plausibly aluminum in high-alumina ashes, beyond the iron index calibrated here [12,14]. For granulated blast-furnace slag, latent hydraulic reactivity would render the parameters time-dependent even without aggressive exposure, so that the post-peak energy parameter, identified above as the most sensitive descriptor within the calibrated range, would require monitoring against the degree of activation rather than against exposure alone [13,59]. For biomass and co-combustion fly ash, the wide compositional variability, particularly unburnt-carbon contents ranging from below 1% to above 20%, would chiefly affect the energy parameter through carbon heterogeneities acting as crack initiators, while alkali-activated fly ash gels would resist calcium dissolution but remain vulnerable to depolymerization under strong acid, altering the functional form of the strength law [8,58,61]. For recycled glass fines, the near-absence of calcium and the predominance of amorphous silica shift the governing mechanism from acid dissolution to the alkali-silica reaction, which would manifest as a reduction in stiffness accompanied by an increase in transition and peak strain over months to years, and would require a sodium- or silicon-based expansion index rather than iron [53,60]. It is emphasized that the trilinear constitutive form itself was calibrated for the gypsum-cycle decalcification mechanism of sulfuric-acid attack; for mechanistically distinct attacks such as alkali-silica reaction or external sulfate attack, both the master index and the constitutive form may require re-validation, and not only the choice of indicator. The framework therefore offers methodological architecture and a workflow for future experimental campaigns, not a ready-made predictive tool for binders whose dominant damage mechanism differs from the case validated here, as indicated in Figure 8.



**Figure 8.** Framework architecture for extending the validated NHL-pozzolan CDM model to selected circular binder systems. Dashed frames indicate prospective, non-validated applications.

## 4. Sustainability Implications

### 4.1. Durability-Corrected Environmental Performance

A central premise of circular-economy principles applied to construction materials is that environmental benefits accrue only if a material achieves a service life sufficient to offset its production-related burden. The global warming potential (GWP) of a material, expressed per unit of structural service delivered, must therefore account for the expected service life under the anticipated exposure rather than the embodied carbon of the freshly produced material alone [20,62]. A material with a GWP of 100 kg CO<sub>2</sub>-eq/m<sup>3</sup> and a 100-year life delivers the same benefit per unit time as one with a GWP of 50 kg CO<sub>2</sub>-eq/m<sup>3</sup> and a 50-year life under linear benefit accumulation, yet this equivalence is rarely acknowledged in comparative assessments, most of which report cradle-to-gate embodied carbon without durability adjustment [62].

The framework developed here supplies the missing durability term in a quantitative form: the kinetic sub-model of Section 2.6, together with the leaching-index degradation laws of Section 2.4, yields an exposure-dependent service life by inverting the predictive chain ( $t$ , pH) to  $L_{rel}$  to the CDM parameters to the mechanical end-of-life criterion. For the validated NHL–pozzolan system under sulfuric-acid attack at pH 3.0 and a 50% stiffness-loss criterion, this chain returns a service life of approximately 90 years; for a 70% loss of the post-peak energy parameter, identified in Section 3.5 as the most sensitive descriptor, the predicted service life shortens to approximately 60 years under the same exposure. These values are reported as a demonstration of the chain rather than as an absolute prediction for field service, where pH, temperature, and exposure intermittency would differ from the accelerated laboratory regime; the field-to-laboratory acceleration factor required to translate these laboratory-derived service lives into field estimates is itself a separate research task that is not undertaken here. The sensitivity analysis of Section 3.5 further refines this coupling by indicating that, within the calibrated range, the service-life limit is more likely to be set by loss of ductility and post-peak reserve than by loss of nominal strength, so durability-corrected GWP should be referenced to an energy- or ductility-based end-of-life criterion rather than a strength threshold alone.

To avoid any misinterpretation, it is restated explicitly that the service-life figures of approximately 90 years and 60 years quoted above are laboratory-referenced quantities obtained under continuous immersion at a fixed nominal pH and constant temperature and are reported solely to demonstrate the internal consistency of the predictive chain rather than as field service-life predictions. Their translation into design service lives for real exposure conditions, in which pH, temperature, wetting–drying cycling, and intermittency all vary, would require a field-to-laboratory acceleration factor that has not been established in the present study and whose determination constitutes a distinct experimental and modelling task.

### 4.2. A Corrected Sustainable Material Performance Index

To operationalize the coupling of durability and environmental performance, a composite Sustainable Material Performance Index (SMPI) is proposed that combines the CDM-derived service life, the durability-corrected environmental impact, and the circular-economy valorization ratio. The index is defined as a weighted sum of three normalized sub-indices, as given in Equation (5):

$$SMPI = \alpha \cdot \left( \frac{t_d}{t_d, ref} \right) + \beta \cdot \left( \frac{GWP_{ref}}{GWP} \right) + \gamma \cdot \left( \frac{WVR}{WVR_{max}} \right), \quad \alpha + \beta + \gamma = 1 \quad (5)$$

Here  $t_d$  is the predicted service life from the degradation laws,  $t_d, ref$  and  $GWP_{ref}$  are the service life and global warming potential of a reference conventional material such as an OPC mortar, WVR is the waste-valorization ratio, and  $WVR_{max}$  is

the maximum valorization in the comparison set; a value of SMPI > 1 indicates that the circular material outperforms the reference on the weighted composite. For the validated NHL–pozzolan system, the service-life input  $t_d$  is back-calculated from the predictive chain of Sections 2.6 and 4.1 (approximately 90 years under the pH 3.0 stiffness-loss criterion), and the SMPI computation is therefore internally consistent with the framework. For the OPC reference and the prospective MWBA-blended binder, the service-life inputs in Table 11 are taken from typical literature ranges and are flagged as illustrative; once the experimental program outlined in Section 3.6 supplies measured service lives and environmental-product-declaration data for Baltic by-product binders, the same index can be populated with primary data and the MWBA entry promoted from illustrative to measured. An illustrative application with equal weights ( $\alpha = \beta = \gamma = 1/3$ ) and the corresponding inputs is given in Table 11 for the OPC mortar (reference), the validated NHL–pozzolan system (framework-derived service life), and a prospective MWBA-blended binder (illustrative inputs only). With these inputs, the index ranks the prospective circular binder highest (SMPI approximately 1.48) and the NHL system second (approximately 1.27), both exceeding the OPC reference (approximately 0.67), the ranking being driven by lower embodied carbon and higher valorization rather than by superior durability. The numerical value reported for the MWBA blend is illustrative; the methodology, rather than the specific figure, is the contribution claimed in this section, and the entry should not be cited as a prediction for any specific Baltic source material in advance of the experimental program of Section 3.6.

The weighting factors of Equation (5) were assigned equal values in the baseline calculation because, in the absence of a specific project context or stakeholder preference, an equal weighting represents the most neutral aggregation of the three dimensions of performance. To verify that the ranking obtained is not an artefact of this particular choice, the index was recomputed for the three materials under a range of alternative weightings that successively emphasize durability, environmental impact, and waste valorization, as summarized in Table 11. Across all weightings examined, both circular binders retained a composite index above that of the ordinary-Portland-cement reference, confirming the robustness of the qualitative conclusion; the relative order of the NHL–pozzolan and the prospective municipal waste bottom-ash based systems, however, was found to depend on the emphasis placed on durability relative to valorization, which underlines that the index is intended as a transparent decision-support construct rather than as a fixed ranking. Consistent with this, the corrected baseline value for the NHL–pozzolan system obtained with equal weights and the inputs of Table 11 is 1.27, and the municipal-waste bottom-ash entry is reiterated as illustrative only, to be populated with measured service-life and environmental-product-declaration data once the experimental program of Section 3.6 is completed.

**Table 11.** Illustrative SMPI calculation using equal weights. The NHL–pozzolan service life is framework-derived, whereas OPC and MWBA values are literature-based estimates.

| Material                 | Service Life<br>$t_d$ (yr) | Source of $t_d$           | GWP (kg CO <sub>2</sub> -<br>eq/m <sup>3</sup> ) | WVR (%<br>Mass) | SMPI |
|--------------------------|----------------------------|---------------------------|--------------------------------------------------|-----------------|------|
| OPC mortar (reference)   | 50                         | Literature (illustrative) | 280                                              | 0               | 0.67 |
| NHL–pozzolan (validated) | ~90 (this work)            | Framework, pH 3.0         | 165                                              | 15              | 1.27 |
| MWBA blend (prospective) | 55                         | Literature (illustrative) | 120                                              | 50              | 1.48 |

The service-life, global-warming-potential, and waste-valorization values for the ordinary-Portland-cement reference and for the municipal-waste-bottom-ash system represent theoretical design

targets and literature-derived estimates rather than experimental findings of this study; only the NHL–pozzolan service life is derived from the calibrated degradation framework.

**Table 12.** Sensitivity of the Sustainable Material Performance Index to the choice of weighting factors, computed from the sub-indices of Table 11.

| Weighting ( $\alpha, \beta, \gamma$ ) | OPC  | NHL–Pozzolan | MWBA |
|---------------------------------------|------|--------------|------|
| Equal (1/3, 1/3, 1/3)                 | 0.67 | 1.27         | 1.48 |
| Durability (0.50, 0.25, 0.25)         | 0.75 | 1.40         | 1.38 |
| Environment (0.25, 0.50, 0.25)        | 0.75 | 1.37         | 1.69 |
| Valorization (0.25, 0.25, 0.50)       | 0.50 | 1.02         | 1.36 |

## 5. Conclusions

A leaching-index-driven chemo-mechanical damage framework was developed and validated for predicting the acid-induced mechanical degradation of lime-based mortars, using a comprehensive experimental dataset for a binary NHL–pozzolan mortar exposed to sulfuric acid at pH 1.5, 2.0, and 3.0 for up to 6000 h. The principal findings of the validated study, and the prospective extensions whose epistemic status is explicitly distinguished, are as follows.

- The trilinear CDM model reproduced the elastic, peak, and post-peak softening behavior of all fifteen exposure groups with  $R^2 \geq 0.77$ , with twelve of fifteen groups exceeding 0.95. The systematic evolution of the elastic modulus, peak strength, and energy parameter with pH and exposure duration was corroborated by XRD, TGA, and ICP-OES evidence.
- Iron release was selected as the master normalized degradation index after a documented quantitative comparison with calcium and silicon (Section 2.5), in which calcium was disqualified by gypsum-controlled saturation and silicon by non-monotonic reprecipitation, and after a binder mass balance confirmed that the observed iron release is consistent with partial dissolution of ferrite-bearing accessory phases of the binder. Iron acts in the framework as a monotonic proxy for the cumulative chemical damage state, not as the mechanistic driver of degradation.
- Power-law degradation laws relating the normalized iron-leaching index to four CDM parameters predicted the stress–strain response of three blind validation groups with deviations of 10–18% for stiffness and strength in two groups and 28% in the most aggressively extrapolated group (E\_C\_P2\_6000), the latter being attributable to the documented replicate scatter at the gypsum-rich fiber–matrix interface at intermediate pH. Mean absolute deviations across the three blind groups were 18% in  $E_0$  and 13% in  $\sigma_u$ , with 95% prediction intervals reported in Table 6.
- A kinetic sub-model  $L_{rel}(t, pH) = A \cdot t^m \cdot 10^{-\alpha \cdot pH}$  (Section 2.6) closes the predictive chain from environmental exposure to mechanical state and yields, for the validated system at pH 3.0, service-life estimates of approximately 90 years against a 50% stiffness-loss criterion and approximately 60 years against a 70% loss of the post-peak energy parameter. These service-life values apply to the accelerated laboratory regime; their translation to field exposure requires a separate acceleration-factor study that has not been undertaken here.
- A closed-form sensitivity expression derived analytically from the calibrated laws, propagated by bootstrap resampling of the regression coefficients, showed that the post-peak energy parameter degrades approximately 1.3–1.5 times faster than stiffness or strength within the calibrated range, with non-overlapping 95% bootstrap envelopes between  $|S_d|$  and  $|S_E|$  at  $L_{rel} = 1.0$ . This identifies loss of ductility as the governing reliability concern for the validated system in aggressive environments,

with the caveat that the rank ordering is a mathematical consequence of the calibrated exponents and applies within the calibrated binder and exposure domain.

- The validated evidence rests on a single binder–acid system and on three blind exposure groups, of which the most severely extrapolated (E\_C\_P2\_6000) deviated by 28% owing to a specimen-scale interfacial failure that the homogeneous continuum formulation cannot resolve; the reported service lives are laboratory-referenced quantities that require a validated field-to-laboratory acceleration factor before use in design; and the degradation and kinetic laws are bounded to the calibrated acid-exposure domain  $L_{rel} \in [0, 1]$  and were fitted on nine data points.
- The methodological architecture of the framework is anticipated to be extensible to circular binders (MWBA, GGBS, biomass fly ash, and recycled glass fines) by substituting the binder-specific master index and re-fitting the reference constants. This extension is presented in Section 3.6 as a mechanistic and prospective examination, not as an empirical transferability result; no experimental validation for Baltic by-product binders is reported in this work, and the constitutive form itself may require re-validation for damage mechanisms distinct from the gypsum-cycle decalcification under which it was calibrated.
- Coupling the framework-derived service life to a corrected, weighted-sum Sustainable Material Performance Index linked durability, environmental impact, and valorization in a single interpretable metric. The MWBA entry of Table 11 is flagged as illustrative pending the experimental program on Baltic by-product binders that constitutes the natural continuation of this work and is in progress within the authors' laboratory at Riga Technical University.

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