マテリアル先端リサーチインフラ利用報告書
ARIM User's Report

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課題データ / Project Data

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利用者と利用形態 / User and Support Type

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利用形態 Support Type	共同研究/Joint Research

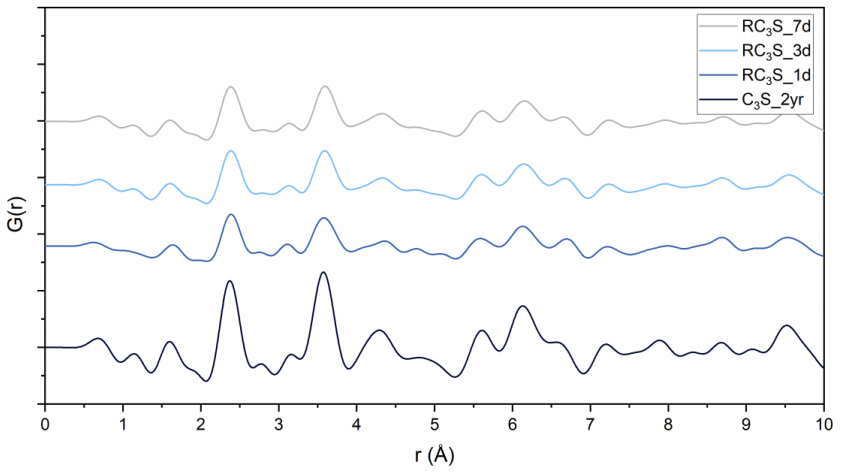
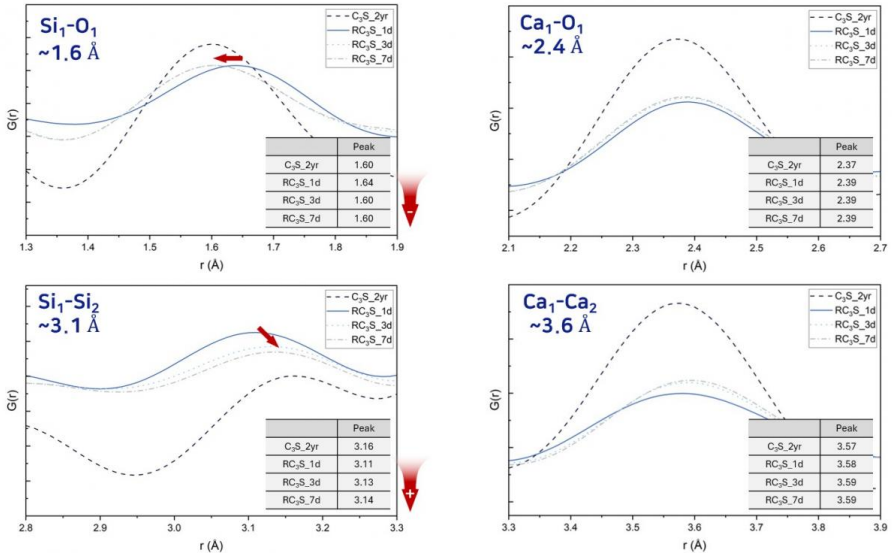
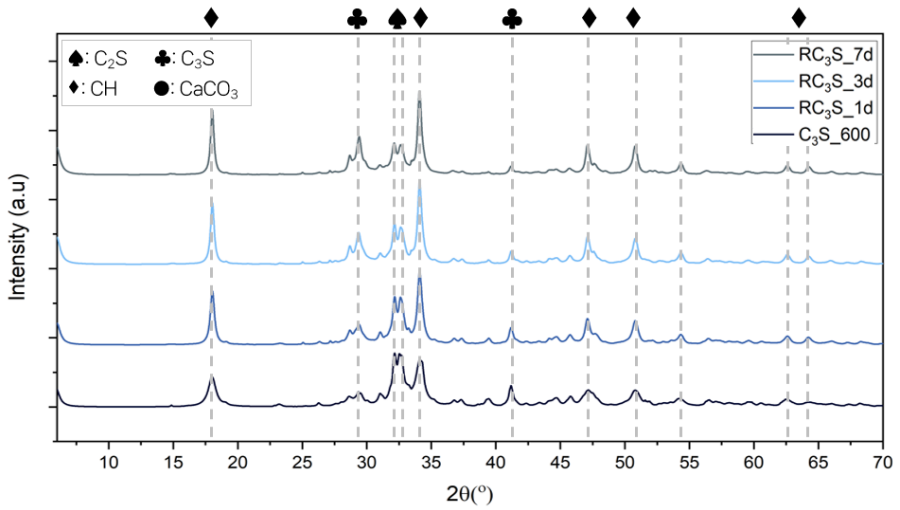
利用した主な設備 / Equipment Used in This Project

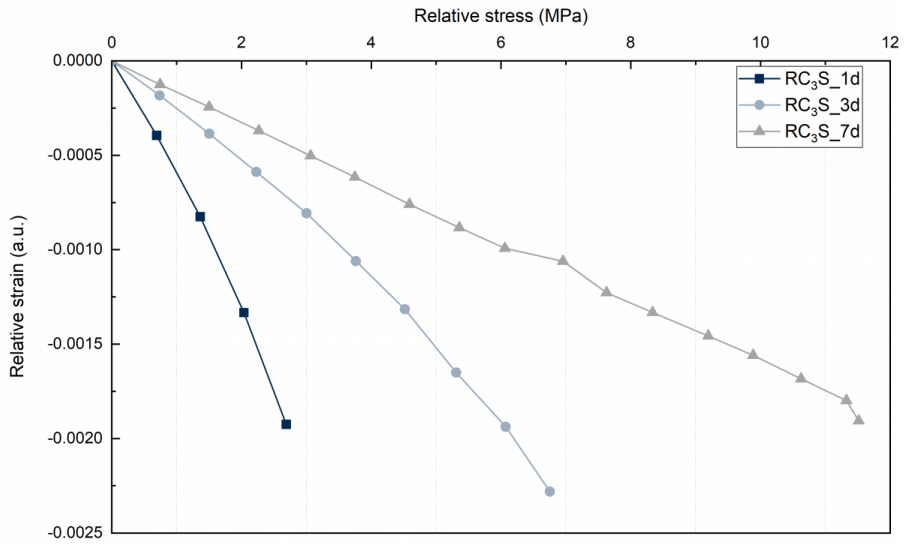
利用した主な設備 Equipment ID & Name	QS-222：高速2体分布関数計測装置

報告書データ / Report

概要（目的・用途・実施内容） Abstract (Aim, Use Applications and Contents)	In concrete industry, significant efforts are being made to achieve a sustainable and circular economy by reducing environmental impacts. Several strategies have been suggested, such as reducing the usage of raw materials, reusing construction and demolition wastes (C&DW), and reducing CO_2 emissions [1]. Reusing C&DW can be an adequate method for achieving a circular economy in concrete industry [2]. In recent years, the recycling and utilization of C&DW have mainly focused on the recycled aggregates obtained from crushed concrete and separated from the mortars, despite hardened cement paste (HCP) accounting for a large proportion of the wastes [1]. The porosity and water adsorption capacity of recycled aggregates are higher than those of natural aggregates due to the adhered hardened cement pastes on the surface [3]. Therefore, the implementation of effective methods for separating the adhered HCP from the recycled aggregate surface, such as mechanical crushing, thermal separation techniques, and gravity separation, is crucial for the reuse of C&DW [1]. Among these methods, thermal separation techniques also can recover the hydraulic properties of recycled cement pastes (RCP), and the rehydration process of thermal-activated RCP can restore compressive strength [1]. As reported in previous studies, the optimum thermal activation temperature, which significantly influences the rehydration ability of RCP, is suggested as 600–800℃ [2]. At thermal activation temperature ranges of 600–800℃, the formation of hydraulic phases—including β-, α-, γ-C_2S, and CaO, is accompanied by the loss of chemically bound water and the decomposition of hydration products—such as amorphous calcium silicate hydrate (C-S-H), calcium hydroxide (CH), and ettringite [1]. Although the thermal phase transition behavior of HCP is well-defined using multiple advanced characterization methods [4], elucidating the rehydration mechanism of thermal-activated HCP remains challenging due to the diverse chemical composition resulting from the complex phases within HCP and variable activation temperatures. To clarify the rehydration kinetics of thermal-activated HCP, it is imperative to uncover both the restoration of hydraulicity and the rehydration mechanism of pure C_3S pastes, which is the primary component of ordinary Portland cement clinker. This study aims to investigate the rehydration kinetics of thermal-activated 2-year-old-pure C_3S pastes and the effect of rehydration degree on the mechanical properties of recycled C_3S pastes at early ages via in-situ synchrotron X-ray scattering experiments combined with pair distribution function (PDF) analysis.

実験 **Experimental**	The C_3S pastes decomposed into amorphous phases such as residual C-S-H, different polymorph of C_2S (α, β, and γ) after heating up to 600℃. Furthermore, as depicted in Fig. 2(B), a drastic increase in amorphous contents, i.e., C-S-H, was observed at an early age of rehydration, which is related to the early strength development of recycled cement pastes due to the high surface area of particle and fast CaO rehydration. Hence, the correlation between early strength development of thermal-activated C_3S pastes and phase evolution can be quantified via utilizing the in-situ synchrotron X-ray scattering under external loading combined with PDF, which is a powerful analytical tool to study short- and intermediate order in amorphous materials. Furthermore, the total X-ray scattering data with a high angular resolution and PDF data can provide the precise phase quantification results using Rietveld refinement method and PDFgui software. As results, the relationship between the phase evolution and the separated elastic behavior of residual C-S-H, regenerated C-S-H and CH, and decomposed amorphous phase, e.g., nesosilicates, in rehydrated C_3S pastes at early age can be elucidated. Moreover, the precise quantification result obtained from Rietveld and PDFgui can be used as input data in GEMS software to investigate the rehydration degree of thermal-activated C_3S pastes. 準備した合成 Thermal-activated C_3S pasteに圧縮負荷をかけながら（常温）XRD実験を行った。事前に準備した試験片は持ち込み装置である小型引張圧縮試験機一式（AC 100 V）を用いて、予備実験の結果より最大15˜18 MPaまで0.5～1 MPa間隔で圧縮応力を負荷した圧縮試験を行った。BL22XUのSi (111)二結晶分光器で単色化した70 keVのX線を使用した。検出器には回折計に搭載された大型デジタル２次元X線検出器を用い、入射ビームサイズは0.5 mm × 0.5 mmとした。試験片からの距離Lを304 mmおよび654 mmでそれぞれPDFおよびXRD解析用のデータを収集した。
結果と考察 **Results and Discussion**	Based on the pair distribution function (PDF) analysis, thermally activated and rehydrated C_3S pastes initially showed distinct atomic pair distributions compared to long-term hydrated C_3S, as shown in Fig. 1. However, as the curing period progressed (1, 3, and 7 days), the differences in atomic distances—particularly a decrease in Si–O and an increase in Si–Si—indicated the formation and stabilization of hydration products (Fig. 2). This trend suggests a recovery in the structural characteristics of calcium silicate hydrate (C–S–H) phases. Furthermore, X-ray diffraction (XRD) and Rietveld refinement confirmed the growth of crystalline $Ca(OH)_2$ and the presence of amorphous C–S–H during rehydration, as shown in Fig. 3. Notably, CH content increased significantly at early curing ages, which correlates with the rapid initial rehydration of CaO and C_2S. In addition, in-situ loading experiments conducted on the rehydrated samples revealed that the formation and polymerization of hydration products contributed to mechanical strength recovery, as evidenced by the elastic stress–strain response observed under compressive loading (Fig. 4). These results collectively demonstrate that thermally treated C_3S paste can recover its hydraulic properties upon rehydration and form hydration products similar to those produced in ordinary hydration processes. Therefore, the findings provide promising evidence for the potential reuse of thermally activated cement powders derived from waste concrete as sustainable cementitious materials.

図・表・数式 1 **Figures, Tables and Equations 1**	 Fig. 1
図・表・数式 2 **Figures, Tables and Equations 2**	 Fig. 2
図・表・数式 3 **Figures, Tables and Equations 3**	 Fig. 3

図・表・数式 4 Figures, Tables and Equations 4	 Fig. 4
その他・特記事項（参考文献・謝辞等） Remarks(References and Acknowledgements)	[1] L. Xu, J.J. Wang, K.F. Li, S.Y. Lin, M.L. Li, T.Y. Hao, Z. Ling, D. Xiang, T.Y. Wang, A systematic review of factors affecting properties of thermal-activated recycled cement, Resour Conserv Recy, 185 (2022). [2] J.A. Bogas, S. Real, A. Carriço, J.C.C. Abrantes, M. Guedes, Hydration and phase development of recycled cement, Cement Concrete Comp, 127 (2022). [3] C.J. Shi, Y.K. Li, J.K. Zhang, W.G. Li, L.L. Chong, Z.B. Xie, Performance enhancement of recycled concrete aggregate - A review, J Clean Prod, 112 (2016) 466-472. [4] S. Im, H. Jee, H. Suh, M. Kanematsu, S. Morooka, K. Taku, N. Yuhei, A. Machida, J. Kim, S. Bae, Temperature effects on local structure, phase transformation, and mechanical properties of calcium silicate hydrates, J Am Ceram Soc, 104 (2021) 4803-4818. [5] A. Abidar, O. Bouchenafa, R. Hamzaoui, S. Mansoutre, M. Paris, L. Barnes-Davin, C. Capra, B. Classen, C. Florence, Investigating the mechanisms of hydraulicity regeneration in pure C_3S paste, Constr Build Mater, 409 (2023). Related Paper:Im Sumin, Jee HyeonSeok, Kanematsu Manabu, Morooka Satoshi, Choe Hongbok, Nishio Yuhei, Machida Akihiko, Tominaga Aki, Jeon Byonghun, Bae Sungchul, "Multi-scale Synchrotron X-ray Scattering Studies on Thermo-induced Changes in Structural and Mechanical Properties of CSH/PCE Composites", Construction and Buildling Materials, 459, 139742 (2025). Doctor Thesis:Im Sumin, "Multiscale Studies on Thermo-Treated Calcium Silicate Hydrates: Its Potential for CO_2 Sequestration", Hanyang University, Ph. D. Thesis (2025). Award:Im Sumin, Bae Sungchul, 2024 Architectural Institute of Korea Excellence Award, 2024/10/24.

成果発表・成果利用 / Publication and Patents

DOI（論文・プロシーディング） DOI (Publication and Proceedings)	
口頭発表、ポスター発表および、その他の論文 Oral Presentations etc.	
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