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Cements and concretes materials characterisation using machine-learning-based reconstruction and 3D quantitative mineralogy via X-ray microscopy

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Abstract

3D imaging via X-ray microscopy (XRM), a form of tomography, is revolutionising materials characterisation. Nondestructive imaging to classify grains, particles, interfaces and pores at various scales is imperative for our understanding of the composition, structure, and failure of building materials. Various workflows now exist to maximise data collection and to push the boundaries of what has been achieved before, either from singular instruments, software or combinations through multimodal correlative microscopy. An evolving area on interest is the XRM data acquisition and data processing workflow; of particular importance is the improvement of the data acquisition process of samples that are challenging to image, usually because of their size, density (atomic number) and/or the resolution they need to be imaged at. Modern advances include deep/machine learning and AI resolutions for this problem, which address artefact detection during data reconstruction, provide advanced denoising, improved quantification of features, upscaling of data/images, and increased throughput, with the goal to enhance segmentation and visualisation during postprocessing leading to better characterisation of samples. Here, we apply three AI and machine-learning-based reconstruction approaches to cements and concretes to assist with image improvement, faster throughput of samples, upscaling of data, and quantitative phase identification in 3D. We show that by applying advanced machine learning reconstruction approaches, it is possible to (i) vastly improve the scan quality and increase throughput of 'thick' cores of cements/concretes through enhanced contrast and denoising using DeepRecon Pro, (ii) upscale data to larger fields of view using DeepScout and (iii) use quantitative automated mineralogy to spatially characterise and quantify the mineralogical/phase components in 3D using Mineralogic 3D. These approaches significantly improve the quality of collected XRM data, resolve features not previously accessible, and streamline scanning and reconstruction processes for greater throughput.

KEYWORDS
automated mineralogy, cement, concrete, image analysis, machine learning, X-ray tomography

1 | INTRODUCTION

3D imaging via X-ray microscopy (XRM) is revolutionising materials characterisation and the understanding of grains, particles, interfaces and pores. Detailed and high-quality imaging and analyses are imperative for our understanding of the composition, structure, lifetime transformation and failure of a variety of materials, including building materials such as cements and concretes.[1] Imaging via XRM has an advantage over more traditional 2D methods because it enables the full analyses in all axes, it is nondestructive and it allows imaging at the multiscale, allowing for a more holistic interpretation of samples. Therefore, it is an obvious choice for cements and concretes.

However, there is necessitated improvement of the data acquisition process of samples that are fundamentally challenging to image, usually because of their large size, density (high atomic number) and/or the resolution they need to be imaged at. This is a particular challenge for cements and concretes because the samples are usually heterogeneous and diverse cores varying greatly in their size/diameter (resulting in long scan times because of the need to increase the number of projections collected, or long exposure times), chemical composition, and size of features (e.g., grains, pores, cracks), where observations of small regions of interest alone are not suitable for complete characterisation. There is a need to acquire XRM data at the multiscale, where higher resolution scans complement the lower resolution scans (and vice versa), which is often constrained by high-resolution interior tomographies having a much smaller field of view and a lack of X-ray penetration, appearing noisy and suffering from artefacts, making many small features unobservable. Further, there is generally a contrast overlap between different components of interest that have a similar chemical composition and structure (i.e., cements and concretes being dominated by calcium-based phases) which are difficult to differentiate from one another via standard reconstruction or segmentation techniques. Ideally, improved quantification of features, upscaling of data/images, and increased throughput for repeat samples is required, with the goal to enhance reconstructed data for more authentic segmentations, leading to a better and more holistic characterisation of samples, whatever the problem and application.

Here, we apply various aspects of the ZEISS Advanced Reconstruction Toolbox (ART) to address these issues: DeepRecon Pro, a machine-learning-based reconstruction approach,[2,3] to show that it is possible to vastly improve scan quality through enhanced contrast and denoising, with the additional advantage of increasing throughput of 'thick' cores through shorter scan times. Additionally, we 'upscale' higher resolution targeted regions of interest scans to larger fields of view using DeepScout,[4,5] resulting in resolution recovery. Finally, we apply Mineralogic 3D, a quantitative machine learning approach to spatially characterise and quantify the mineralogical/phase components in 3D. These approaches significantly improve the quality of collected XRM data, resolve features and compositional data not previously accessible, and streamline the scanning and reconstruction process for greater throughput.

2 | SAMPLES AND METHODS

Cements and concretes are versatile materials used for a variety of applications, ranging from civil building materials to the nuclear industry. Of the former, there is a drive to produce cements that are more environmentally friendly, reducing the need for raw materials extraction by using waste materials (e.g., fly ash, blast furnace steel industry and iron-making slag) and that have a lower carbon footprint during their production (e.g., those that cure faster at room temperature). Of the latter, the drive is to produce cements that aid in potential nuclear waste containment (i.e., do not contain pathways for leakage), and concretes that attenuate gamma radiation and can cope with elevated temperatures (i.e., when used in nuclear reactors).[6,7] Therefore, enhanced characterisation of these materials from advanced imaging and reconstruction are vital for their characterisation (Table 1).

The samples studied here were cores of variable thickness ranging from 5 to 30 mm diameter. The thicker cores were particularly challenging to image because of limited X-ray penetration. First, the samples were scanned non-destructively in 3D using a ZEISS Xradia 620 Versa X-ray Microscope (XRM). Two types of scans were collected: lower magnification, larger field of view scans to image as much of the sample as possible; and region of interest (ROI) interior tomography scans at higher magnification (and resolution), using the Versa's bespoke proprietary optics to target specific internal regions to image via the Scout and Zoom approach (Figure 1).

Finally, the resulting scan data were run through a variety of ART software options: DeepRecon Pro,

TABLE 1 Sample information for cements and concretes used in this publication.

Sample number	Sample type	Special features	Used for	Processes
1	**Blended Portland cement**	Contains recycled materials (slag from the iron-making process) 5 mm thick core	General construction	DeepRecon Pro; Mineralogic 3D
2	**Geopolymer cement**	Cure at room temperature, low CO_2 process; contains aluminosilicate precursor (metakaolin) geopolymer and Fe agglomerates for simulated nuclear waste 10 mm thick core	Potential nuclear waste containment and immobilisation	DeepRecon Pro
3	**Dolerite 'high-density' concrete**	Dolerite aids in attenuation of gamma radiation, also contains recycled materials (fly ash) 15 mm thick core	Nuclear industry vessels for AGR reactors	DeepRecon Pro; DeepScout
4	**Standard building concrete**	'Normal' cement minerals (Ca silicates, hydroxides) and aggregates 30 mm thick core	General construction	DeepRecon Pro; Mineralogic 3D

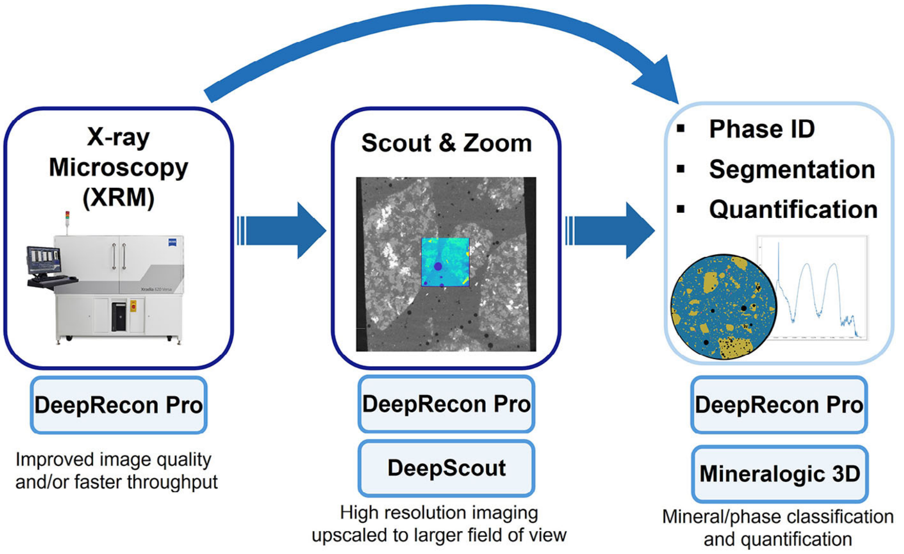

FIGURE 1 Schematic illustrating the workflows used in this publication.

a machine-learning-based reconstruction approach for advanced denoising, enhanced contrast, improved identification of small features and increased throughput; DeepScout, which provides the ability to 'upscale' lower resolution data over a larger field of view; and Mineralogic 3D, which allows us to spatially characterise and quantify mineralogy/phases in 3D. A summary of the workflow used here can be seen in Figure 1.

3 | RESULTS

3.1 | Improving data quality during reconstruction (DeepRecon Pro)

DeepRecon Pro advanced reconstruction provides an alternative to the more traditional (and most common) filtered back-projection or FDK reconstruction technique. In FDK, the entire volume is reconstructed in one single step but has sensitivity to both noise and artefacts, often needing a large number of projections, or long scan times through long exposure times, to overcome these issues. Following this, the FDK reconstructed data is usually run through a series of filters in postprocessing software to try and reduce the noise (e.g., nonlocal means, Gaussian, deconvolution), improve image quality, and deal with artefacts. This can be a long process and does not always result in the desired outputs, sometimes over-smoothing and digitally 'erasing' features of interest.

DeepRecon Pro offers an alternative. It is a deep-learning-based artificial intelligence technique which reconstructs raw projection data as soon as it is generated from the XRM. A trained neural network is generated for image improvement, interpretation and retrieval, allowing

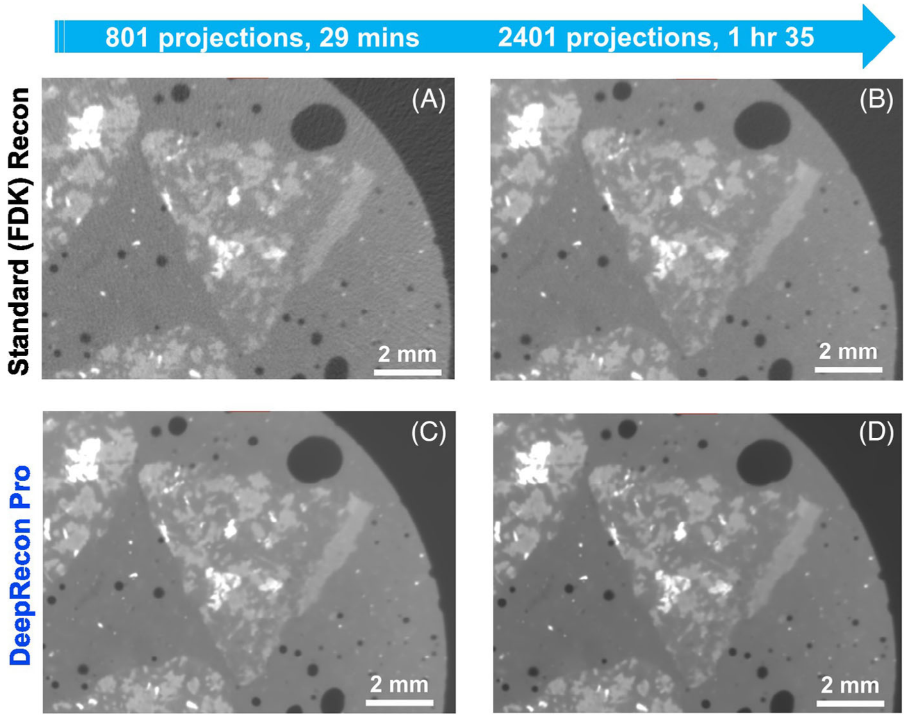

FIGURE 2 2D slice comparison of a zoomed section of a sample 3 concrete core reconstructed using standard methods (FDK) and DeepRecon Pro.

for high-quality reconstructed data even when performing rapid acquisitions using a small number of projections or short exposures (Figure 2). Once a model has been generated, it can be applied to as many similar samples in repetitive experiments as required, further streamlining and speeding up the scanning process. For cements and concretes, these factors are particularly useful because (a) it reduces scan times, (b) it allows for greater throughput, and (c) it does not compromise scan time for scan quality, resulting in true, useable data that can then be utilised for more accurate segmentation, analysis, and modelling. Moreover, the DeepRecon Pro approach is universally valuable for all cement and concrete types and applications, rather than one alone (Table 1).

Examples presented here (Figure 2) shows that scan times can be 3× faster using DeepRecon Pro over standard FDK reconstructions with improved image quality and superior identification of phases and features (Figures 2–4). Figure 4 shows the advantage of using DeepRecon Pro to identify aggregates that make up the 'grains' of the concrete and separate them from the lower contrast cement matrix. Further, the unique ability to use multiple objectives in the Versa XRMs to target internal tomographies of high-resolution regions have traditionally been problematic for large, dense samples, often being noisy and lacking in contrast; results presented here (Figure 4C and D) show that DeepRecon Pro assists greatly with improved contrast, denoising, and general image enhancement, offering a unique insight into the micrometre and nanometre scale relationships in cements and concretes.

3.2 | Resolution recovery: upscaling of high-resolution interior tomographies to larger fields of view (DeepScout)

Like most other methods of microscopy, whether in 2D or 3D, XRM has field of view (FOV) limitations for high-resolution imaging. In most situations, multiple scans on the XRM are collected at the multiscale, often using successively higher objectives (0.4× > 4× > 20× > 40×) to improve the voxel size and spatial resolution, with the field of view decreasing with each scan (Figure 5A). Smaller FOV interior tomographies also often suffer with limited X-ray penetration resulting in noise and artefacts, and consequently require long scan times.

DeepScout is a novel deep learning reconstruction method to address the challenge of achieving high-resolution at larger FOVs. This AI powered technique and workflow can be used to restore the high-resolution scan data to a large FOV scan, or, resolution recovery of that data. It works by replacing the traditional deconvolution step with a convolutional neural network trained specifically on a spatially registered low- to high-resolution feature map. The workflow then performs a spatial

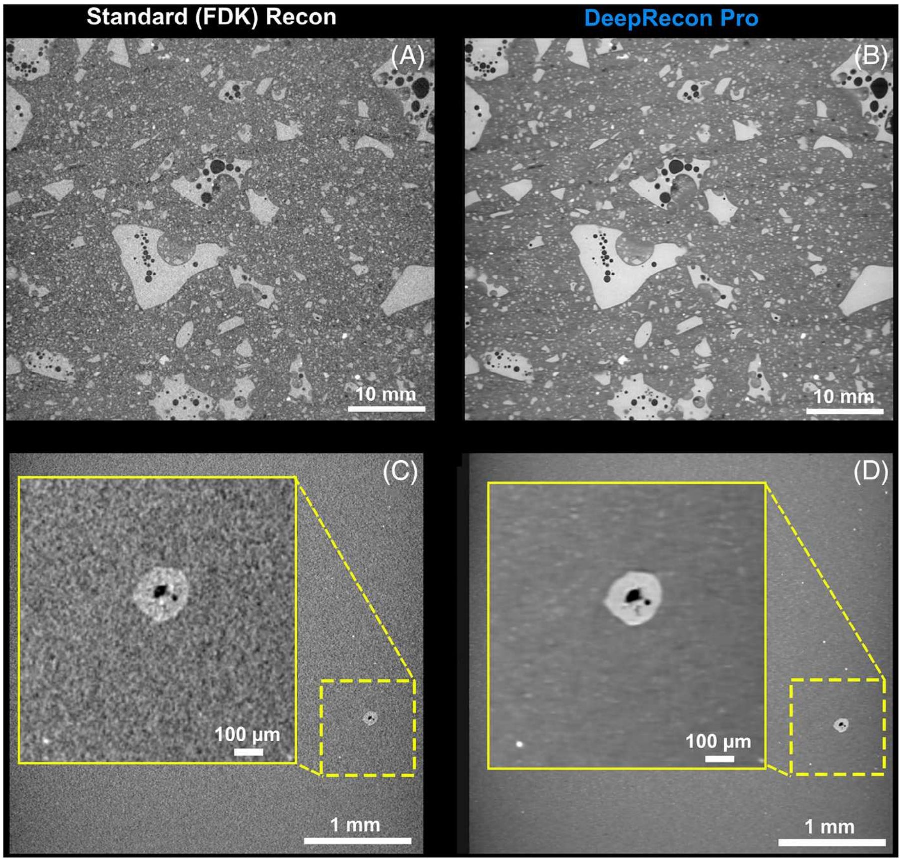

FIGURE 3 2D slice comparison of standard FDK reconstruction and DeepRecon Pro results for Blended Portland cement (sample 1; A and B) and a geopolymer (sample 2; C and D). Denoising and general image improvement observed in DeepRecon Pro data. A and B show porous residual blast furnace slag particles in a blended Portland Cement. Insert in C and D shows an encapsulated particle of Fe-rich sludge waste embedded in an otherwise fairly featureless geopolymer matrix; however, these grains were difficult to identify and determine a robust shape analysis using FDK reconstruction alone, and we are able to identify much smaller Fe-rich grains, otherwise obscured by noise, within the 'featureless' geopolymer matrix.

registration of the multiscale datasets, and generates a model based on the high-resolution data.[5]

This applies to cements and concretes by upscaling high-resolution data to larger fields of view to provide more information over a larger area, more accurate segmentations, and results that are more meaningful and representative of the whole samples rather than studying one small FOV alone. In Figure 5, we have applied this to nuclear reactor concrete; we have upscaled a 4× objective 5 µm high-resolution scan collected at 5.9 mm^3, to 27 mm^3 of the original 0.4× objective low-resolution scan. This is an almost 5× increase in FOV. This approach provides more information about the sample and scan over a larger area: we have improved the contrast between different constituents, the visibility and identification of small grains within the matrix, and the outline and features within high-density grains, which will lead to a more accurate segmentation and analyses of the sample and the ability to spot areas of weakness (Figure 5C and D).

3.3 | 3D distribution and quantitative phase analysis of components with similar contrasts (Mineralogic 3D)

Mineralogic 3D uses XRM data and deep learning algorithms to execute automated mineralogy analyses in three dimensions that provide particle identification, mineral/phase classification and data outputs including segmentation and quantified component analyses. Collecting mineralogical/phase data in 3D has benefits over more traditional 2D chemical acquisition methods (e.g., SEM-EDS); there is no complicated sample preparation, every grain/particle is viewed fully in 3D so there are no stereological assumptions and the time taken to generate useable data is greatly reduced.[8] Mineralogic 3D works by first using DeepRecon Pro to enhance the data through advanced reconstruction, and subsequently takes into account the scanning conditions (kV), X-ray attenuation, the 'real life' density and the chemical composition

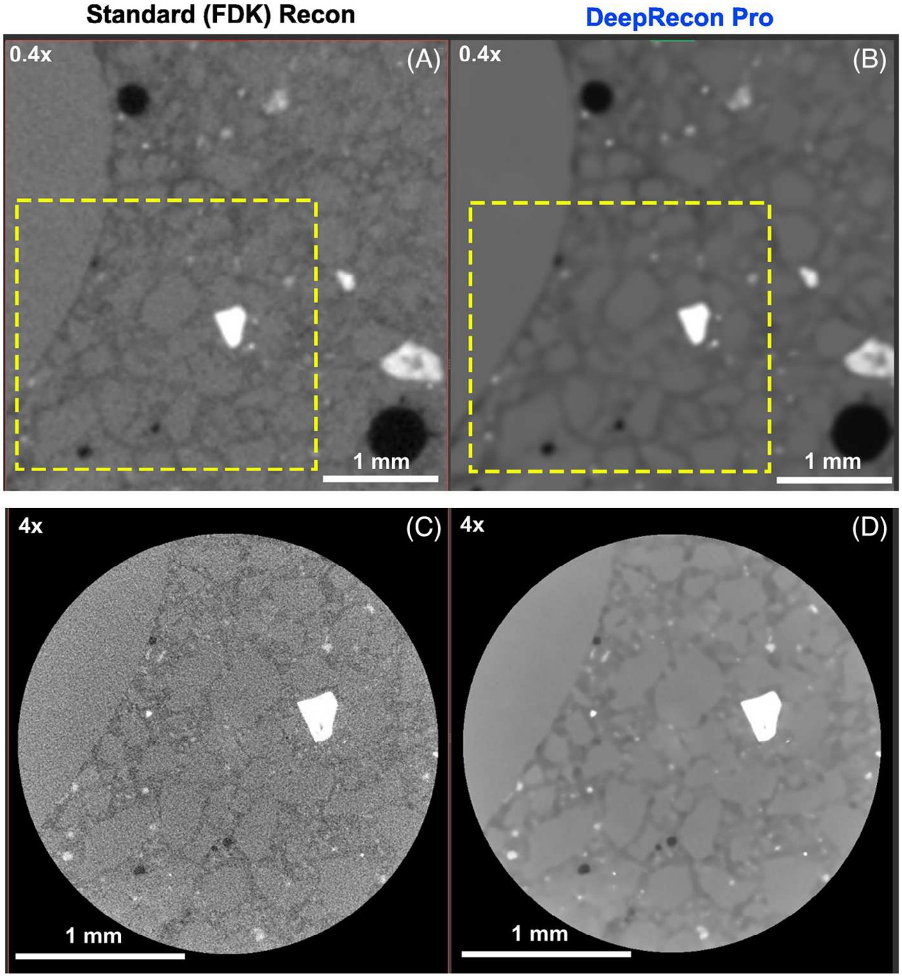

FIGURE 4 2D slice comparison of standard FDK reconstruction and DeepRecon Pro results for 0.4× (A and B) and 4× (C and D) Scout and Zoom interior tomography scans of a 20 mm diameter standard building concrete (sample 4) core. Improvement of the data is observed in both examples. 0.4× scans collected at 27.21 µm voxel size, while 4× ROI scan was collected at 5 µm voxel size. Spatial resolution improvements exemplified from 0.4× to 4× scans, highlighting the need to scan at the multiscale to obtain the required details.

of minerals/phases to apply deep learning algorithms for accurately segmented and quantified data. Moreover, use of a 'mineral library' ensures repeat or batch samples can be run accurately, repetitively and quickly.

Applying this approach to cements and concretes has multiple benefits. First, it allows us to ascertain what minerals/phases are in the samples and to segment them accordingly at multiple scales for a variety of sample sizes. Second, DeepRecon Pro allows us to enhance the contrast, allowing minerals/phases of similar attenuation values to be segmented. Figure 6 shows the plot of four groups of phases that we have successfully segmented in Figure 7, which includes the separation of calcium silicates, the main constituents of the cement powder, which also contains some calcium carbonate. When these are mixed with water, undergoing hydration, they form calcium silicate hydrate and calcium hydroxides, with the calcium carbonate being slightly reactive. This process is essential for the hardening and setting of the cements, and is incredibly dependent on the quantities and proportions of each added component. Here, we can differentiate the lime – an expanding agent which is added and important for overcoming contraction during hydration – from the Ca silicates, which tells us how much 'free lime' has survived reaction/the curing process. Moreover, we can segment and quantify recycled waste materials which are commonly added, such as the aforementioned fly ash and steel/iron industry blast furnace slag. Using these materials not only provides an alternative calcium source and lowers the carbon footprint between industries, but imaging and analysing in this manner allows users to quantify specific constituents, such as the amount of silica and aluminium that is added for more resistance to chemical attack.

Third, we can improve the segmentation of different components through this advanced analysis method. Figure 7 shows the difficulty in seeing the difference between the green and yellow groups in regularly FDK

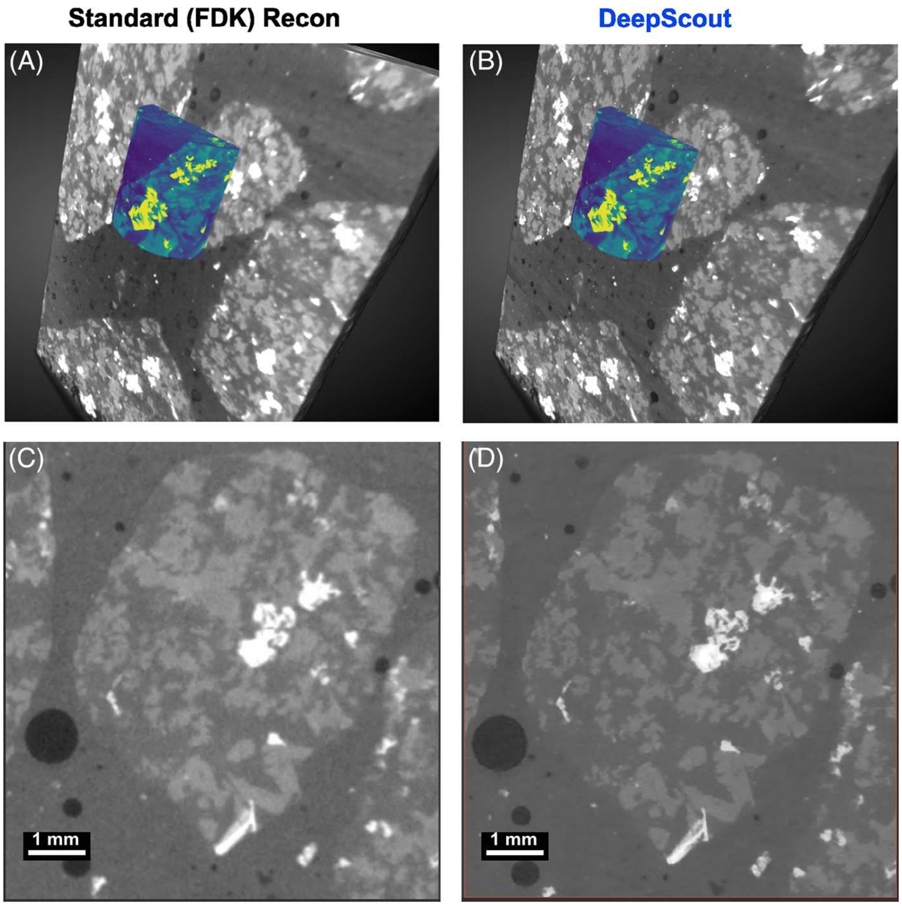

FIGURE 5 Upscaling of internal 4× ROI 5 μm voxel size scan data to larger field of view 0.4 × 28 μm voxel size scan in dolerite concrete (sample 3). Internal tomography of targeted ROI scan shown in bright colours in A and B. Using DeepScout improves the contrast between different constituents, the visibility and identification of small grains within the matrix, and the outline and features within high-density grains (C and D).

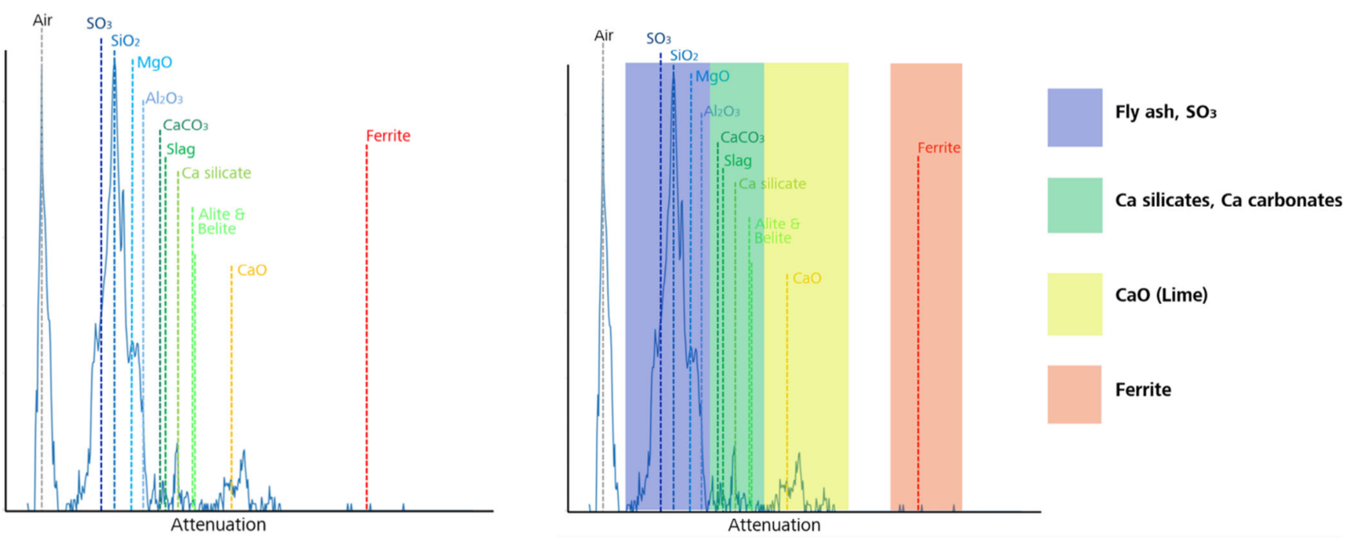

FIGURE 6 Mineralogic 3D histogram of sample 4 (standard construction concrete). Attenuation histogram is smoothed in the first step of Mineralogic using DeepRecon Pro; following look up using the mineral library, phases and minerals are automatically plotted to histogram peaks. We are able to divide the phases/minerals into groups, and are able to tell the difference between Ca phases.

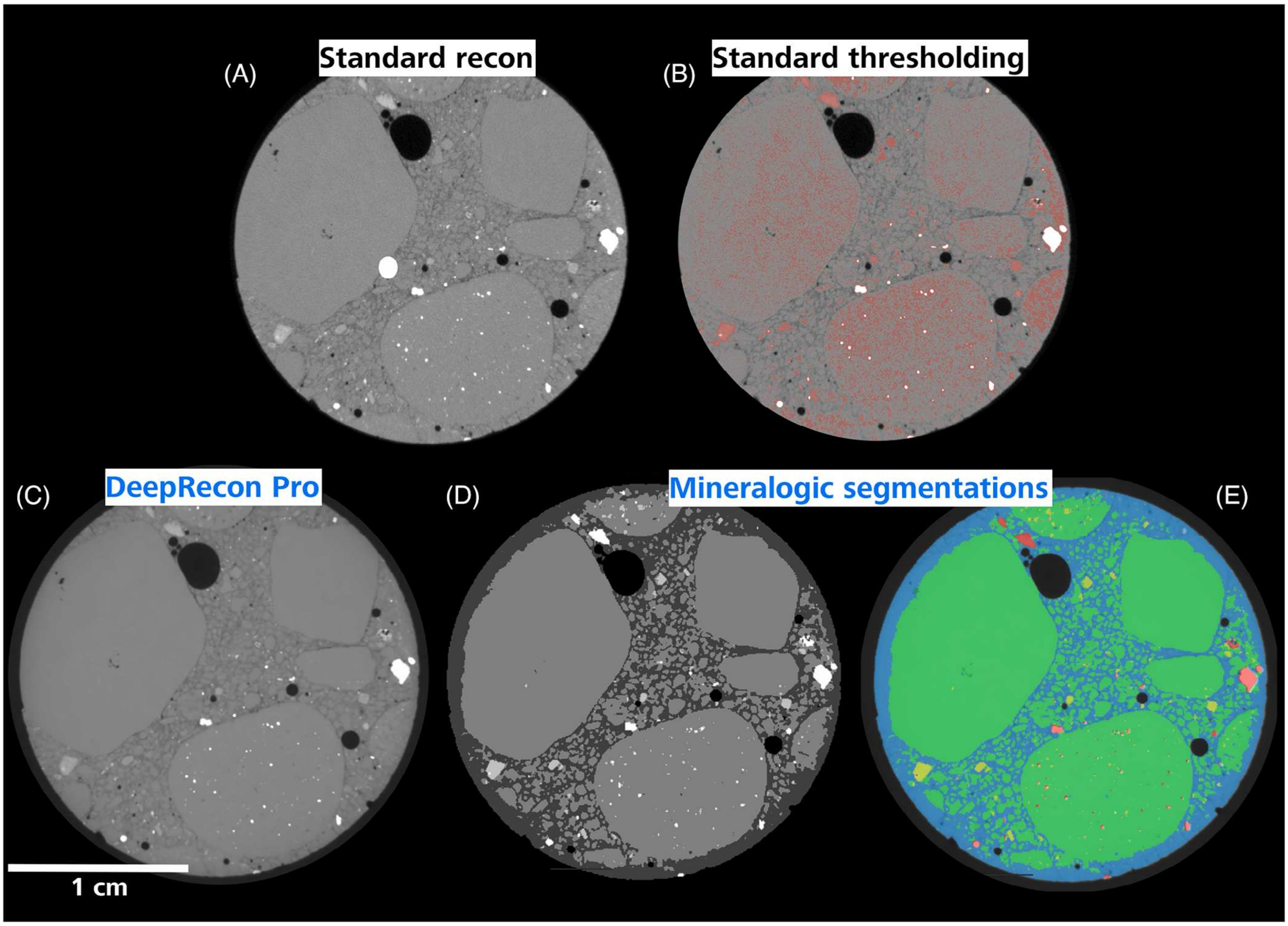

FIGURE 7 Comparison of standard FDK reconstruction (A), standard thresholding (B), DeepRecon Pro (C), Mineralogic 3D phase segmentation (D) and coloured Multi ROI segmentation (E) for standard building concrete (sample 4). Four phases are segmented (not including air), including phases with very similar contrasts that would be difficult to segment with conventional thresholding techniques as in B (yellow phase in E).

reconstructed greyscale images of the data; however, Mineralogic 3D can spot the differences. This is in comparison to attempting a manual threshold of the yellow group using the regularly reconstructed FDK data, which does not segment the data correctly and selects areas of other groups that have fluctuating greyscales/noise (Figure 7B).

4 | CONCLUSIONS

Traditional methods of tomographic reconstruction, including filtered back projection (FDK), are prone to noise and aliasing artefacts and require many projections to be collected, which can lead to long scan times and sample drift. By applying aspects of the Advanced Reconstruction Toolbox (ART), in conjunction with nondestructive X-ray microscopy (XRM), we can improve the quality of cement and/or concrete scans. In this study, we have shown that we are able to improve the image quality of large samples, reduce scan times and increase sample throughput, improve contrast between similar phases of similar chemistry and resolve features and compositional data not previously accessible for a variety of cements and concretes. This has allowed for a quantified and automated mineralogical (phase) analysis of cements and concretes in 3D using Mineralogic 3D. These results provide us with segmentation results that lead to better and a more holistic characterisation of samples, whatever the problem and application of various building materials.

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REFERENCES

1. du Plessis, A., & Boshoff, W. P. (2019). A review of X-ray computed tomography of concrete and asphalt construction materials. *Construction and Building Materials*, *199*, 637–651. ISSN 0950-0618, https://doi.org/10.1016/j.conbuildmat.2018.12.049
2. Harris, W., Gu, A., & Terada, M. (2022). Putting AI to work: A practical and simple application to improve 3D X-ray FA. *2022 IEEE International Reliability Physics Symposium (IRPS)*, 10C.1-1–10C.1-6, https://doi.org/10.1109/IRPS48227.2022.9764574
3. Andrew, M., Thompson, W., Kumar, A., & Hornberger, B. (2018). Iterative reconstruction for increased imaging throughput in lab-based 3D XRM. *Microscopy and Microanalysis*, *24*, 116–117.
4. Gu, A., Andreyev, A., Terada, M., Rodgers, T., & Viswanathan, V. (2023). A deep learning reconstruction technique and workflow to enhance 3D X-ray imaging resolution and speed for electronics package failure analysis. In *2023 International Conference on Electronics Packaging (ICEP)*, Kumamoto, Japan, pp. 69–70. doi: 10.23919/ICEP58572.2023.10129701
5. Andrew, M., Andreyev, A., Yang, F., Terada, M., Gu, A., & White, R. (2022). Fully automated deep learning-based resolution recovery. In Proceedings of the SPIE 12242, Developments in X-Ray Tomography XIV, 122420 M, https://doi.org/10.1117/12.2633095
6. Obaid, S. S., Gaikwad, D. K., & Pawar, P. P. (2018). Determination of gamma ray shielding parameters of rocks and concrete. *Radiation Physics and Chemistry*, *144*, 356–360. https://doi.org/10.1016/j.radphyschem.2017.09.022
7. Khan, M. N. A., Hussain, M. A., Muhammad, Y. M., Muhammad, U., Muhammad, N., Muhammad, A., Hisham, A., Abdullah, M., & Ahmad, Z. S. S. (2023). Development of high-temperature heavy density dolerite concrete for 4th generation nuclear power plants. *Frontiers in Materials*, *10*, https://doi.org/10.3389/fmats.2023.1057637
8. Hill, E. (2021). Whole particle liberation studies (ZEISS Mineralogic 3D), ZEISS microscopy application note, EN_42_013_348 | CZ 11–2021 |.

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