Advances in Characterisation Methods and Computational Modelling

Workshop Report

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MatCHMaker
Materials Characterisation & Modelling

EMMC

Funded by
the European Union

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1. Introduction

MatCHMaker[1] was awarded a grant under the call HORIZON-CL4-2022-RESILIENCE-01-19 [2] - Advanced materials modelling and characterisation (RIA) as the future of European industrial manufacturing requires further advances in characterisation methods and computational modelling. This will lead the way to the reliable design of new and sustainable materials and processes, rapid upscaling, and effective quality control. These advances can only be achieved through the development of innovative techniques and a new generation of instrumentation, responding to industrial needs. MatCHMaker and its sister projects AddMorePower,[3] AID4GREEENEST,[4] CoBRAIN,[5] and D-STANDART[6] introduce their approach to this topic.

In this report, we provide a first insight on each of these projects as presented in our joint online workshop "Advances in characterisation methods and computational modelling". (see Appendix) The presenters selected a particular industrial challenge (use case) and introduced their approach on how to advance characterisation methods and materials modelling, respectively.

2. The Needs of Industry and the Solutions Provided

The first use case of the project **MatCHMaker** turned the attention to **cement** and how this product can be optimised. Industry is looking to reduce CO_2 emissions and for low-carbon binders. To decrease its CO_2 emission, the cement of today is already substituted with Supplementary Cementitious Materials (SCM) which are wastes of other industries like: fly ashes and ground granulated blast furnace slags (GGBS). As the EU is on its road to decarbonizing its industries, coal power plants will progressively shut down and the steel industry is changing its processes; this will lead to shortage of fly ashes and GGBS. Among new SCM, calcined clays and limestone are among the most abundant. However, the amounts of each material have to be optimized to minimize CO_2 while keeping durability and mechanical performance identical to current products. It is important to understand the microstructure since the usage of low-carbon binders will have different phase assemblages which in turn influences the macroscopic properties. Conventional X-ray diffraction (XRD) is not sufficient as it only measures precisely the amount of crystalline phases and does not recognise porosity. In MatCHMaker's approach, scanning electron microscopy (SEM) and Transmission Electron Microscopy (TEM) are used to identify and quantify the amount of each phase, i.e., areas of the microstructure with similar element composition. During SEM, the backscattered electrons (BSEs) are analysed, so one can characterise deeper regions of a sample. Energy-Dispersive X-ray Spectroscopy (EDX) is harnessed to obtain the chemical composition.

However, the raw images obtained with SEM and EDX must be processed to identify and quantify the relevant phases that characterise the cement paste. A dedicated processing workflow, depicted in Figure 1, has been developed to perform this analysis, combining Machine Learning (ML) approaches and expert's knowledge. The first step of this workflow consists in the clustering of the

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[1] https://cordis.europa.eu/project/id/101091687
[2] https://cordis.europa.eu/programme/id/HORIZON_HORIZON-CL4-2022-RESILIENCE-01-19/en
[3] https://cordis.europa.eu/project/id/101091621
[4] https://cordis.europa.eu/project/id/101091912
[5] https://cordis.europa.eu/project/id/101092211
[6] https://cordis.europa.eu/project/id/101091409

pixels into the main phases in a Portland cement: pores, hydrates, portlandite and clinker. A second step of the workflow focuses on the clinker to obtain a more detailed phase identification (alite, belite, celite, ferrite).

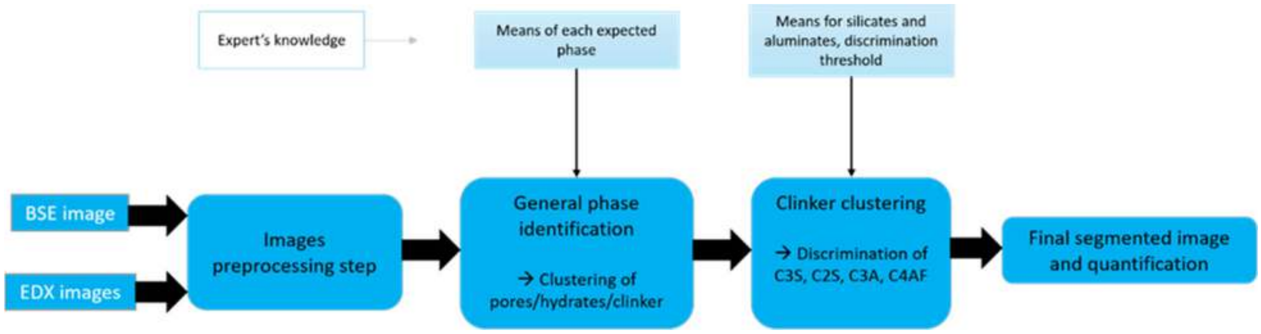

Figure 1. General workflow for the analysis of SEM and EDX images of cement paste

An example of the final result is illustrated in Figure 2. Future work will exploit the uncertainty prediction of the ML models and their propagation to the phase quantification.

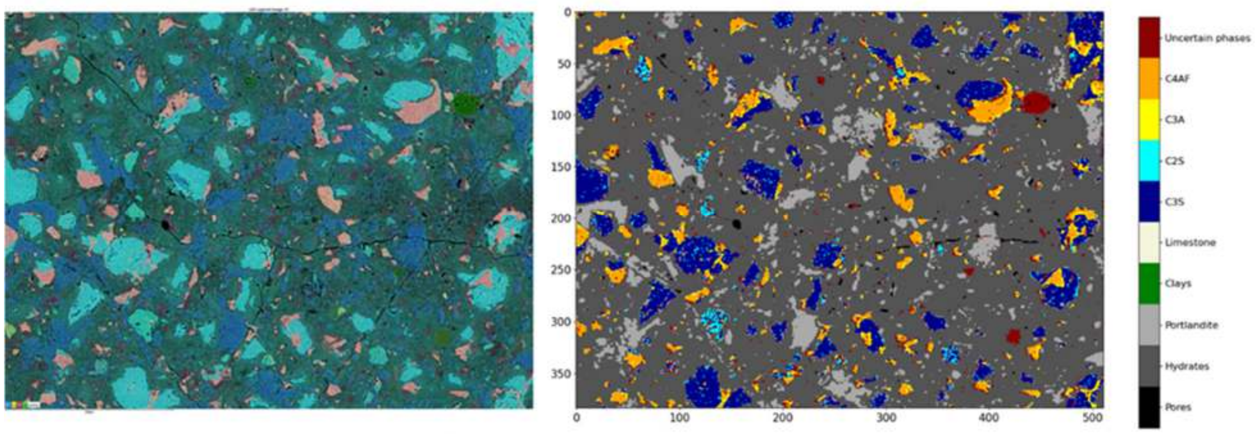

Figure 2. Left: Example of a layered image of Portland cement (2 days). Right: Result of the clustering result using the developed workflow

One of the main challenges of this approach is its full automation. Currently, expert's input is needed to provide an initial guess for ML algorithms. These inputs depend on the image, their acquisition condition (contrast, saturation) and the expected phases to recognise in the sample (presence of limestone, calcined clay, …). As the number of images is expected to be vast, building a seamless workflow will rapidly appear as a necessity.

The challenge for MatCHMaker is to translate a human expert's knowledge into data a machine can correctly process. This data-driven method comes with the necessity to store meaningful data, of which many may be proprietary. The cement manufacturers will profit from this novel characterisation workflow as it enables them to analyse low-carbon binders and understand the microstructures fast.

The Need: Low-carbon binders characterization for the building sector.

The Solution: A seamless robust processing workflow for SEM and TEM images using ML to understand the microstructure in depth as opposed to time-consuming and subjective manual image analysis.

AID4GREENEST is investigating Artificial Intelligence (AI) assisted characterisation workflows to characterise **advanced steels**. They are using SEM techniques such as Electron Backscatter Diffraction (EBSD) and Secondary Electron imaging (SE). The ultimate goal will be to populate an open repository, MicrostructureDB,[7] where users can explore the microstructure space and find links between the structure, processing and properties of steel. The database will support different privacy levels to accommodate industrial users. Advanced AI tools will aid to explore, organise, and make predictions for user's own datasets. The idea is to create maps of metal where similar images cluster together and different materials are located in spatially separated clusters. The maps are constructed from advanced deep learning models both from labelled and unlabelled data. A useful test data set for this development was the UltraHigh Carbon Steel micrograph DataBase (UHCSDB).[8] An overview of the workflow is depicted in Figure 3.

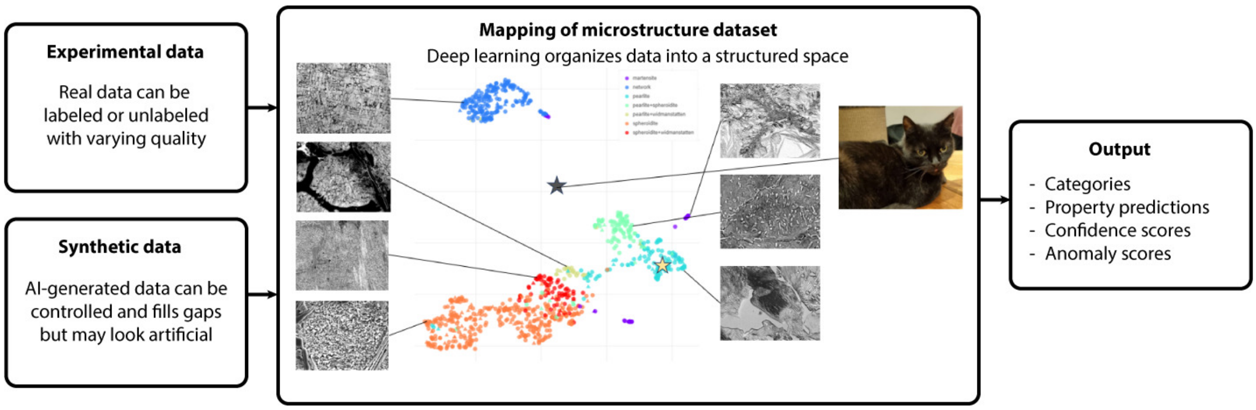

Figure 3. Workflow of the AID4GREENEST methodology

Working with AI-assisted characterisation raises the issue whether enough real data is available; often data are sparse and expensive to obtain. Also, the diversity of available material data is limited which is not beneficial in the quest for novel and advanced materials for the steel industry. This is where synthetic data created by AI can play a role as filling in the knowledge gaps; however, they must ultimately correlate to a physical reality. Microstructures of steels are complex and require novel characterisation workflows. The steel industry will profit from guidance of which process parameters they have to tweak to get the desirable microstructures and thus, a better steel.

The Need: Manufacturing of advanced steels to keep up with contemporary market needs.

The Solution: An open repository with experimental and modelling data and AI tools to assist the knowledge search as opposed to siloed information with low data interoperability.

AddMorePower attempts to develop advanced characterisation workflows for the **power electronics** industry. Copper metallisation is a process that uses copper to improve the performance and reliability of power electronic devices. However, such devices are exposed to high currents and frequencies which may cause cracks and subsequently lead to failure. A synchrotron-based X-ray microscope is used to understand the thermomechanical fatigue processes that incur in copper. As these processes are multi-scale and multi-physics events, Dark-field X-ray microscopy (DFXM) is used as an imaging technique. AddMorePower (Figure 4) is using the experimental structures obtained

[7] https://microstructuredb.com/
[8] https://holmgroup.github.io/publications/uhcs-data.pdf

from characterisation as an input for the Düsseldorf Advanced Material Simulation Kit (DAMASK) [9] with the goal to elucidate the degradation mechanisms and approach life-time modelling. Their data is managed with NOMAD Oasis [10] and they develop parsers and require programming skills for their digital threads.

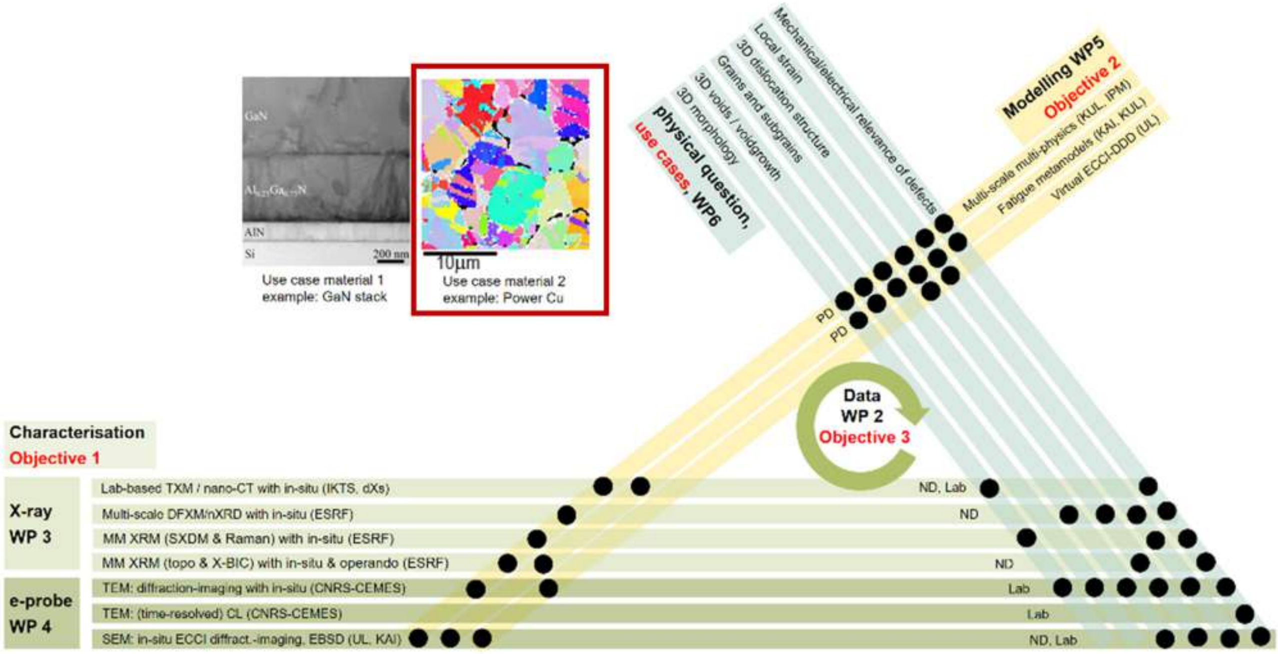

Figure 4. AddMorePower deeply integrates X-ray and Electron probe microscopy techniques (Objective 1) with materials modelling (Objective 2). To successfully enable this, an intensive data management is necessary (WP2). Applied are these characterisation-modelling workflows for power electronics copper metallisation structures as well as wide bandgap semiconductors

These characterisation workflows do require expert knowledge and cannot be wrapped up in user-friendly apps in the near future. However, Europe harbours these experts and some industrial challenges can only be tackled with ingenuity rather than routine, especially when it comes to multiscale characterisation and modelling. The instruments comprised in a synchrotron setting are on a high technology readiness level (TRL) and can be democratised, i.e., made accessible to persons who wish to engage with more challenging characterisation settings.

The Need: Increase the performance and reliability of power electronic devices.

The Solution: Bring complex characterisation and modelling workflows to live by making them accessible to experts and lift them off the drawing board.

The **CoBRAIN** project focus is on the innovation in **coatings** that various industry sectors need to prevent corrosion and combat wear. It is of interest to replace strategic and toxic elements such as cobalt, or processes employing toxic compounds like chromium electroplating. Coatings must perform often under extreme environmental conditions and ideally, be made from sustainable and "green" materials. The interoperability of data obtained from physical models, numerical models and characterisation is at the core of the selection and development criteria for new materials, optimised for each working environment. Characterisation methods must cover the nano-micro-meso scale, be correlated with performance indicators, and give a clear picture of the coatings structure and this,

[9] https://damask-multiphysics.org/
[10] https://nomad-lab.eu/nomad-lab/nomad-oasis.html

ideally very fast. The methods of choice (Figure 5) for CoBRAIN are multi-technique High Speed Nano-indentation and scratch testing, wear testing (sliding, abrasion, etc.) and electrochemical corrosion testing, combined with high-resolution microscopies (e.g. SEM, EBSD, FIB+TEM), X-ray diffraction, and spectroscopies (e.g. micro-Raman).

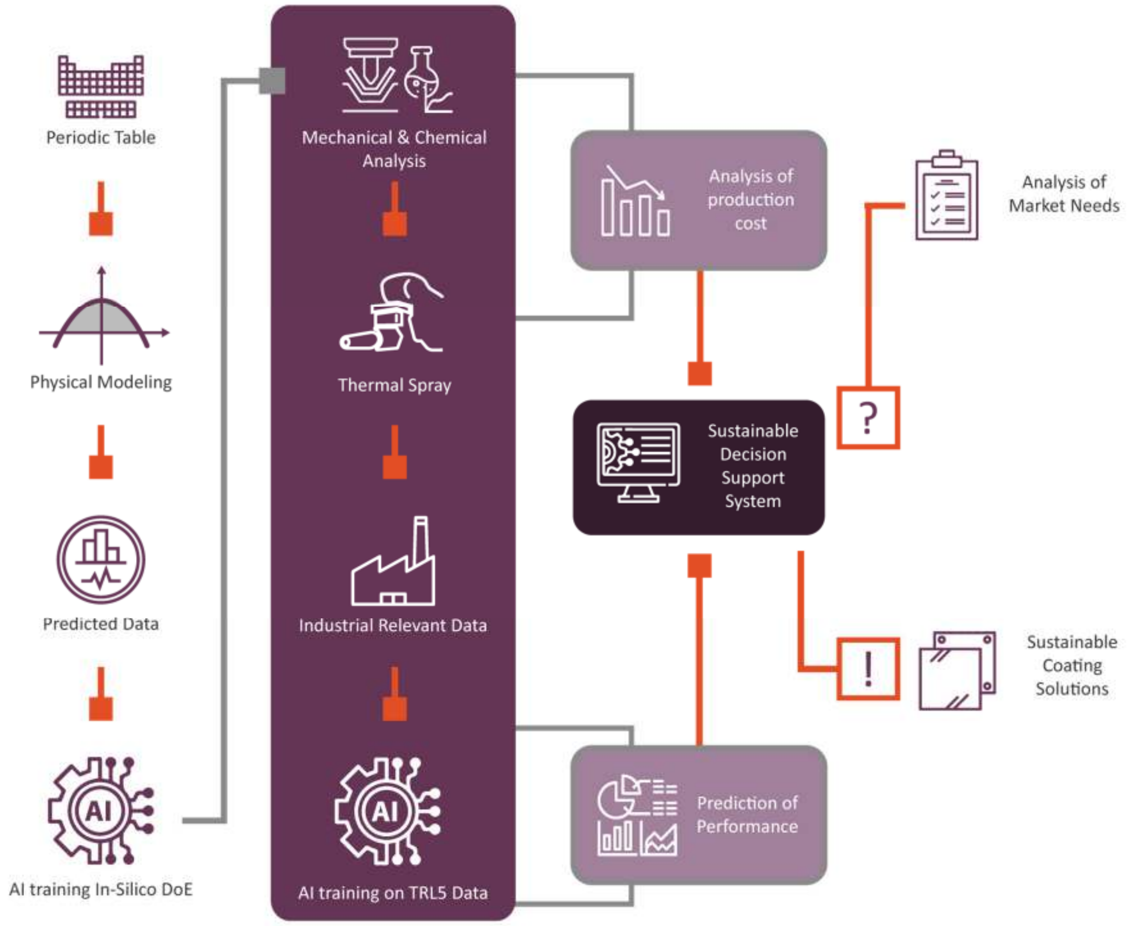

Figure 5. Main idea at the basis of CoBRAIN's project

The partners are also using physics-based materials modelling combined with ML. The alloys are designed in-silico and novel chemistries can be researched. Multiple computational methods, including Computer Coupling of Phase Diagrams and Thermochemistry (CalPhaD), Density-Functional Theory (DFT), Computational Fluid-Dynamics (CFD) and Finite Element (FE) methods, are employed to model the coatings' properties and the materials evolution during the thermal spray process. ML models are then be used to augment the physics-based data to obtain a large dataset and to correlate them with experimental data. Ontologies are used to guarantee semantic interoperability among all experimental and theoretical workflows. The outcome will be a link between process data gathered in the project and the materials performances to carry out a Life Cycle Performance Assessment (LCPA) and thus enable a Sustainable Decision Support System (SDSS).

The Need: Wear and corrosion protective ceramic/metal coatings comprising less toxic and more abundant elements.

The Solution: To design dedicated material solutions for industries using ontologies to guarantee semantic interoperability between state-of-the-art characterisation, materials modelling and ML methods.

Composite structures in the aerospace and renewable energy industries are the topic of project **D-STANDART**. Composites are seen as crucial materials for a sustainable future; however, they must be reliable during and ideally beyond the lifetime of the product. This requires careful evaluation of their fatigue performance. Fatigue testing of composites is very time consuming, since each possible lay-up in a design has to undergo the full cycle from coupon to full-scale testing. Hence if the manufacturers could reduce the number of tests without compromising on safety, the time to market could be reduced. Currently this issue is solved by only qualifying a small set of lay-ups, and restricting designers to only pick lay-ups from this set. This severely restricts the material design space, and sacrifices one of the key advantages of composites: that the lay-up can be optimally tailored to the applied loads. To address this issue, D-STANDART proposes a combination of new experimental methodologies for characterising composite laminates, combined with AI-based surrogate modelling (Figure 6).

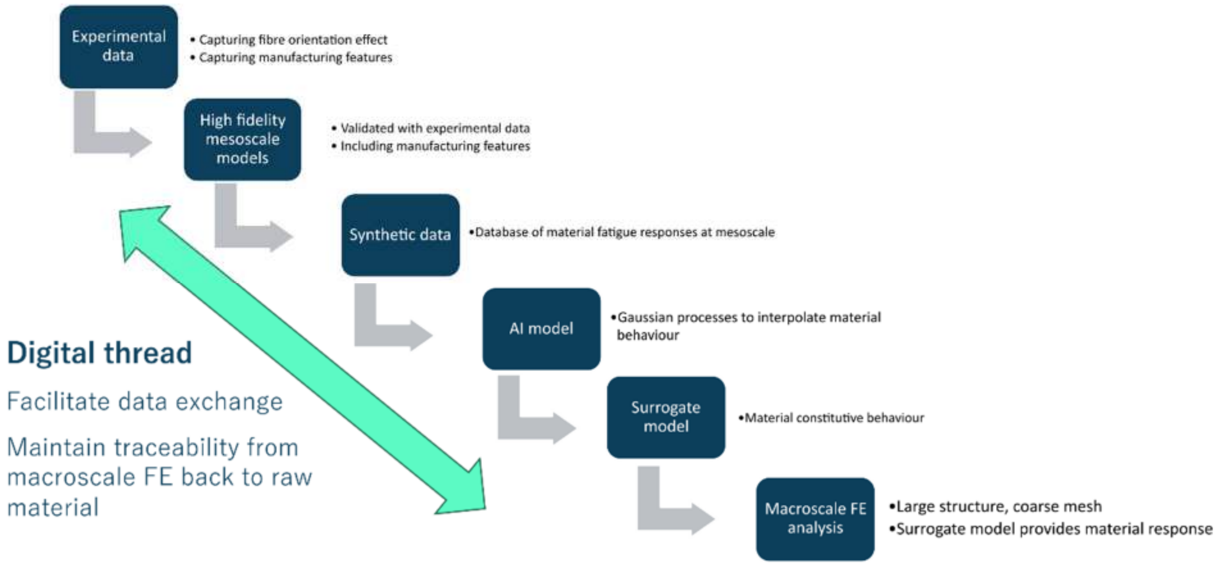

Figure 6. Workflow of the D-STANDART methodology for fatigue evaluation

Experimental data are used to validate high fidelity mesoscale models. These models are then used to create synthetic data, which are the basis for the training of surrogate models that can capture the complexities of the composite material. These surrogate models can then be integrated into numerical analyses of full-scale structures, providing insight into the expected fatigue performance, at comparatively low computational cost. The whole process is geared towards providing confidence in the product's durability to the manufacturer from the start. To further build confidence in the final predictions, and to support digital product certification in a future industrial environment, D-STANDART is developing a digital thread that facilitates the data exchange and maintains traceability throughout the workflows.

The Need: Sustainable composites for the aerospace and renewable energy industries.

The Solution: AI-based surrogate modelling to enable evaluation of composite structures with arbitrary lay-ups, supported by a digital thread to connect experiments, surrogate models and life cycle predictions, with full traceability.

3. Conclusions and Outlook

The five projects cover a wide range of industries and application, and give evidence that workflows comprising both characterisation and modelling data are key on the path to develop advanced materials. ML and AI play a big role in aiding the human-in-the-loop with uncovering the knowledge that is distributed in the available data. The human-in-the-loop plays a big role to provide data in ML/AI ready format and assures that ML/AI is learning the right things. The concept of digital threads will be important so the process of how data were collected becomes transparent and FAIR.

Advances in Characterisation Methods and Computational Modelling will require that practitioners meet in a pre-competitive way and share best practises. Ideally, they agree on data formats and make data available to enlarge the learning space for ML/AI. The latter are software and require verification and validation to be trustworthy. Also here, developers may want to share best practises and procedures where the reliability can be confirmed.

All five projects have impressive case studies and will deliver a proof of concept for their respective methods; however, to establish a workflow in industry will require a manufacturer to add it to their existing roadmap[11] and make it part of their process from a managerial perspective. Hence, the projects aim to clearly demonstrate value, so industry may be open to adopt new workflows.

Hence, our cluster of projects is looking forward to become an enabler for industry to use novel characterisation and modelling workflows to take advantage of it and advance materials for the common benefit.

4. Acknowledgement

This study has received financial support from the European Union: MatCHMaker (GA 101091687), AddMorePower (GA 101091621), AID4GREENEST (GA 101091912), CoBRAIN (GA 101092211), and D-STANDART (GA 101091409).

5. Disclaimer

All statements of fact, opinion, or analysis expressed in the report are those of the authors only and do not necessarily reflect those of the European Union. Neither the European Union nor the granting authority can be held responsible for them. The information used and statements of fact made are not guarantees, warranties or representations as to their completeness or accuracy. The authors assume no liability for any short term or long terms decision made by any reader based on analysis included in this report. In many cases, the opinion expressed in the reports is our current opinion based on prevailing trends and is subject to change.

[11] E.g., for the cement industry, see: https://gccassociation.org/

6. Acronyms, Abbreviations and Elucidations

AI – Artificial Intelligence

BSE - backscattered electron

CFD - Computational Fluid-Dynamics

DAMASK - Düsseldorf Advanced Material Simulation Kit

DFT – Density Functional Theory

DFXM - Dark-field X-ray microscopy

EBSD - Electron Backscatter Diffraction

EDX - Energy-Dispersive X-ray Spectroscopy

FE - Finite Element

GGBS - granulated blast furnace slags

LCPA - Life Cycle Performance Assessment

ML – Machine Learning

SCM - Supplementary Cementitious Materials

SDSS - Sustainable Decision Support System

SEM - Scanning Electron Microscopy

TEM - Transmission Electron Microscopy

TRL - technology readiness level

UHCSDB - UltraHigh Carbon Steel micrograph DataBase

XRD - X-ray diffraction

7. Appendix

The meeting was facilitated by the EMMC and took place online, 24th Oct 2024, 1-5pm CET.

Time/CET	Speaker	Topic
1.00 – 1.05	Ludovic Jason MatCHMaker	Welcome, setting the scene
1.05 – 1.35	Alexandre Ouzia (Heidelberg Materials, D) and Geoffrey Daniel (CEA, F) MatCHMaker	*"ML model for phase assemblage analysis of low carbon cement pastes"*
1.35 -2.05	Rina Jaeken (ePotentia, B) AID4GREENEST	*"Computer vision insights from the development of MicrostructureDB"*

2.05 – 2.35	Andre Clausner (Fraunhofer IKTS, D) AddMorePower	"*Advanced Characterisation and Modelling for Degradation Processes in Copper BEoL Stacks of next generation Power Devices*"
2.35 – 3.05	Anssi Laukkanen (VTT, FI) and Marco Sebastiani (University Roma Tre, I) CoBrain	"*Combining physical modelling, artificial intelligence, and experimental verification for the development of sustainable coating materials based on high-entropy alloys*".
3.05 – 3.15	Break	
3.15 - 3.45	John-Alan Pascoe (TU Delft, NL) D-Standart	"*More efficient fatigue evaluation of composite structures by leveraging machine learning and surrogate modelling*"
3.45 – 4.30	Open Discussion	