Effect of the surface roughness of fibres on the bonding capacity of the interfacial zone between the fibres and cementitious matrix

Anna Antonova
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Abstract

As fibre-reinforced cementitious composites (FRCC) are multi-scale materials, their structural performance depends on the micro-scale properties of the fibre-matrix bond. However, the development and utilisation of FRCC are restricted due to the limited knowledge of the micro-scale phenomena that influence the bond between the fibres and the cementitious matrix and its response to loading.

The focus of this research was the definition of the properties of the fibre surface and the cement paste surrounding it, which affect the formation and performance of the fibre-matrix bond. The examination involved analysing the effect of the surface roughness of steel fibres on the microstructure of the interfacial transition zone (ITZ) and the degradation of the fibre-matrix bond under repeated loading. The micro-scale properties were explored by employing a different approach to applying the existing experimental techniques to the FRCC.

The utilisation of scanning electron microscopy (SEM) and phase-contrast micro-computed tomography (µCT) enabled the identification of the changes in the distributions of calcium hydroxide, calcium silicate hydrate, pores and unhydrated cement grains within the ITZ. The application of phase-contrast µCT allowed access to the three-dimensional microstructure of the cement paste around the fibre. The importance of the surface roughness of steel fibres for the packing of cementitious grains was examined by estimating the average height and wavelength of surface irregularities using an SEM image analysis and directly measuring the parameters using an atomic force microscope and stylus profilometer. The effect of the fibre surface roughness on its wettability was evaluated through contact angle goniometry. The decrease in the mobility of the water along the fibres that was observed with an increase in fibre surface roughness facilitated the reduction in the porosity near these fibres, which was confirmed using SEM. The resulting mechanical response of the bond between the cement paste and fibres with different types of surface roughness was examined under direct tension cycles with gradually increasing amplitudes. The outcomes of the fibre pull-out tests indicated that the detected micro-scale changes in the properties of the fibres and the cement paste surrounding them influenced the maximum capacity of the fibre-matrix bond and its deterioration. The development of the residual slip was identified from the beginning of loading with the three stages of evolution: deceleration, steady stage and acceleration.

This study points out that the properties of the fibre surface and the cement paste surrounding it clearly affect the performance of the fibre-matrix bond by introducing novel insights about the fibre-matrix interaction that advance the development and modelling of FRCC.

Keywords fibre-matrix interface, fibre-reinforced concrete, phase-contrast tomography, surface roughness, bond degradation
Preface

Looking back over the past few years, I do not regret a single day I have spent in the world of research. There were hard and devastating days, but they were fading at the moment of discovering novel, expected or unexpected answers and phenomena. I am grateful to Aalto ENG Doctoral Programme and Betonitekniikan alan yhteistyöopimust for funding this research and providing a great opportunity to become a part of the scientific community, learn new skills, and explore different materials. The financial support from Auramo Säätiö, Kerttu, and Jukka Vuorinen Fund is also very appreciated.

I am deeply grateful to my supervisor, Professor Jari Puttonen, who encouraged and motivated me during my doctoral studies through fruitful discussions and always reasonable advice on experimental or writing issues. This work would be impossible to complete without your constant guidance throughout these years, especially during COVID times. I would like to thank the advisor of my research work, D. Sc. Marika Eik, for the tremendous input she made to help me with the beginning of my steps in research and a lot of her free time that she dedicated to helping me finalise my research work and scientific publications. I would also like to thank, Professor of Practice, Jouni Punkki, who commented on my research from an industrial perspective and supported me at my very first conference presentation.

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Preface

going through the same years of doctoral studies as you do.
  Last but not least, I would like to thank my family and friends for all the encouragement, support, and patience they provided during this long journey of mine.

Espoo, March 17, 2022,

Anna Antonova
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References

Publications
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This thesis consists of an overview and of the following publications which are referred to in the text by their Roman numerals.


IV Anna Antonova, Marika Eik, Jari Puttonen. Importance of the surface roughness of a steel fibre pulled out from cement paste by slowly increasing load cycles. *Submitted to be evaluated for the publication in Cement and Concrete Composites*, 30 pages November 2021.
Author’s Contribution

Publication I: “Roughness of steel fibre and composition of cement paste close to fibre surface”

A.A did the sample preparation, performed all experimental measurements, part of the analysis and visual representation of the results, did literature review, and contributed to manuscript writing. M.E wrote the manuscript, performed part of the analysis and visual representation of the results and was responsible for the manuscript submission to the journal. J.P. provided constant supervision of the entire working process and feedback during the preparation of the manuscript for the submission.

Publication II: “Phase contrast tomography to study near-field effect of polypropylene fibres on hardened cement paste”

A.A did the sample preparation, processed and analysed the received results, did literature review and the visual representation of the results, and contributed to manuscript writing. M.E processed and analysed the data, wrote the manuscript and was responsible for the manuscript submission to the journal. J.P. provided constant supervision of the entire working process and feedback during the preparation of the manuscript for the submission.

Publication III: “Effect of the roughness of steel fibre surface on its wettability and the microstructure of the cement paste close to the fibre surface”

A.A. designed the experiment plan with the help of the co-authors, performed the main part of the experimental measurements, analysed the re-
Author's Contribution

received results and wrote the manuscript with the revision of co-authors. M.E. did a comprehensive revision of the text several times, assisted and consulted the development of image segmentation algorithm with Python, and commented the corresponding text. V.J. performed the measurements of contact angles, gave consistent feedback and comments regarding the corresponding text and data analysis. J.P. provided constant supervision of the entire working process, revision of the manuscript and feedback during the preparation of the manuscript for the submission.

Publication IV: “Importance of the surface roughness of a steel fibre pulled out from cement paste by slowly increasing load cycles”

A.A. designed the experiment plan and the pull-out setup with the help of the co-authors, performed all experimental measurements, analysed the received results and wrote the manuscript with the revision of co-authors. M.E. revised the manuscript multiple times and gave objective feedback during the preparation of the manuscript for the submission. J.P. provided constant supervision of the entire working process, revision of the manuscript and gave valuable feedback during the preparation of the manuscript for the submission.

Language check

The language of my dissertation has been checked by Scribendi. I have personally examined and accepted/rejected the results of the language check one by one. This has not affected the scientific content of my dissertation.
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Abbreviations

FRCC  Fibre-reinforced cementitious composite
ITZ   Interfacial transition zone
CH    $Ca(OH)_2$, calcium hydroxide, Portlandite
w/c   Water-to-cement
SCM   Supplementary cementitious materials
SEM   Scanning Electron microscope
C-S-H Calcium silicate hydrate
$\mu$CT micro-Computed tomography
UH    Unhydrated
BSE   Backscattered electron
PP    Polypropylene
EDX   Energy-dispersive X-ray
Z     Atomic number
MS    Mean-shift
at. % Atomic percentage
$C_3S$ Alite
$C_2S$ Belite
$C_4AF$ Ferrite
AFM   Atomic force microscope
LVDT  Linear variable differential transformers
AFt   Aluminium trisulphate
AFm   Aluminium monosulphate
PSD   Particle size distribution
Symbols

$h_{av}$  Average height of the rough surface profile  \([\mu m]\)

$l_{av}$  Average length of the rough surface profile  \([\mu m]\)

$R_q$  Root mean square roughness  \([\mu m]\)

$\theta_a$  Advancing contact angle  \([^{°}]\)

$\theta_r$  Receding contact angle  \([^{°}]\)

$\Delta s_{res}^i$  Residual slip per cycle, where \(i\) is the number of cycles  \([\mu m]\)

$\Delta s_{rev}^i$  Reversible slip per cycle, where \(i\) is the number of cycles  \([\mu m]\)

$\Delta k_{asc}^i$  Stiffness of the ascending part of load-slip curve per cycle  \([\mu m]\)

$\Delta k_{des}^i$  Stiffness of the descending part of load-slip curve per cycle  \([\mu m]\)
1. Introduction

1.1 Background

Fibre-reinforced cementitious composites (FRCCs) have found broad application and use in the building industry, such as for flooring, tunnel linings, lightweight structures and heavy-duty pavements for airports, docks and harbours. The latest Model Code 2010 recognises fibre reinforcement as an alternative to the shear reinforcement in beams for restraining inclined cracks [1]. The addition of fibres to the concrete facilitates the design of the thin structures with complex geometries in situations where the installation of the conventional reinforcement is challenging and time consuming. The disconnected distribution of steel fibres in concrete also limits the extent of corrosion by the length of a corroded fibre in contrast to a network of connected reinforcement bars [2]. The fibres are introduced into the concrete mainly to restrict crack propagation due to the mechanisms of fibre debonding and pull-out that increase the energy absorption of the composite. This improves ductile behaviour and the post-cracking toughness of the cementitious composite [3]. The narrow widths of the cracks increase the durability of FRCC by restricting the ingress of moisture, carbon dioxide, chloride and other chemicals. However, FRCC is a heterogeneous material with a multi-scale nature. At the structural level, its performance depends on the micro-scale properties of its constituents, which are often neglected or simplified in structural design, reducing the reliability of FRCC. In order to effectively exploit the capacity of FRCC for novel types of application, such as structures of thin or/and complex geometries, it is crucial to understand the characteristics of the interface between the fibre and the cementitious matrix on a micro-scale and to indicate their effects on the load-bearing capacity of FRCC on a macro-scale.

The interaction between the fibres and cementitious matrix on a micro-scale governs the stress-transferring mechanisms, such as adhesion and friction. The properties of the fibre surface and the microstructure of the
surrounding cement paste define the response of the fibre–matrix bond to the loading. The inclusion of fibres in the cement matrix affects the microstructure of the surrounding cement paste, which is called the interfacial transition zone (ITZ), similar to the aggregates and conventional reinforcement. The size of the cement grains varies between 1 μm and 100 μm; this is smaller than the diameter of the fibre, which is typically in the order of millimetres. As a result, the fibre disturbs the packing of large cement grains near its surface, leading to the agglomeration of small cement grains. The small cement grains get completely hydrated, saturating the water solution near the fibre with silicon and, predominantly, calcium ions that precipitate as calcium hydroxide (CH) [4], which forms in the empty spaces around the fibres that are vacated by the hydrated cement grains. Therefore, the microstructure of the ITZ differs from that of the bulk cement paste by a higher amount of CH phases and higher porosity [3, 5–7]. The increased porosity and permeability of the ITZ can facilitate the ingress of gases and chemicals. This leads to the leaching of the calcium from the soluble precipitations of CH at the fibre–matrix interface, the reduction of the bond capacity between the cement paste and the fibre or/and the corrosion of steel fibres [8]. The size and the network of the pores also define the strength of the cementitious materials [8, 9]. An increased porosity and concentration of CH weaken the ITZ, as compared to the bulk cement paste, which can trigger the failure of the fibre–matrix bond and lead to the poor performance and durability of the FRCC. The assumed microstructure of the ITZ is illustrated in Fig. 1.1. In reality, the microstructure of the ITZ around the inclusions is variable without any stratification [10]. The ITZ can be affected by the water-to-cement (w/c) ratio of the concrete mix [5], the addition of supplementary cementitious materials (SCM) such as silica fume, metakaolin or slag [5, 11, 12] and the orientation [13, 14] and properties of the inclusions [15, 16], which complicate the characterisation of the ITZ. Although the ITZ around the fibres has already been studied using different micro-scale techniques, such as scanning electron microscopy (SEM) and nanoindentation, the origins of the bond formation between the fibres and the cementitious matrix and the effects of their properties on the process of interface development are not well understood.

Several researchers have highlighted the importance of having a strong interface between the fibres and matrix for the efficient mechanical performance of FRCC on the structural level [15, 17, 18]. The formation of the calcium silicate hydrate (C-S-H) near the fibre surface is preferable for the improvement of the fibre–matrix interface, as the C-S-H is a better binding material and has lower solubility than CH. The microstructure and the load-bearing capacity of the fibre–matrix interface were modified by adding silica fume to the mortar mix to produce more C-S-H [5, 6, 19] or by treating the surface of the steel fibres either chemically [20–23] or
mechanically [24–26]. However, the studies in which the surface of steel fibres was modified mechanically evaluated only the response of these fibres to monotonically increasing pull-out loads without considering the changes caused by the fibre in the matrix surrounding it. Studies that modified the matrix concentrated on the properties of the cement matrix and its microstructure while ignoring the effects of the fibre characteristics. Understanding the micro-scale properties and behaviour of the fibre–matrix bond is required for improving the performance of FRCC [27]. Recent findings, such as the ones reported in [15, 16], have addressed the connection between the properties of the fibre surface and the ITZ microstructure to explain the nature of the fibre–matrix bond. However, the empirical outcomes that characterise the fibre–matrix interface and the relationship between the properties of the fibre and the cement paste are limited, and the connection of the micro-scale properties of the fibre–matrix interface to the mechanical performance of the material has been assumed without experimental support [15].

1.2 Objectives and outlines of this research

The main objective of the present work was to define the properties of fibres and the changes caused by the fibre in the cement paste surrounding it that can affect the formation and performance of the fibre–matrix bond. These goals were achieved in Publication I and Publication II, which characterised the main parameters of the ITZ microstructure and fibre surface that should be considered for modifying the fibre–matrix bond. The second part of the main objective was to study the effect of fibre surface roughness on the micro-scale properties of the fibre–matrix interface and
the load-bearing capacity of the fibre–matrix bond, which were the topics of Publication III and Publication IV. The main research questions considered to be within the scope of this objective are illustrated in Fig. 1.2.

The scale of the objective defined that of the experimental techniques employed. The lack of knowledge about the fibre–matrix bond entailed that a novel use of empirical approaches was necessary. Therefore, another objective of this research was to advance the micro-scale investigations of the fibre–matrix interface by applying modern methodologies, such as micro-computed tomography (μCT) or a pull-out test with loading cycles, that had not yet been fully utilised for the examination of cementitious composites to resolve the research objectives introduced.

This thesis consists of four sections that have been divided based on the order of the publications and research questions.

The Introduction chapter is followed by the chapter Methodology, which provides a review of the experimental approaches used to study the interface or bonding between fibres and the cementitious matrix and their outcomes. The choice of the experimental techniques applied in this research to study the properties of the cement paste near the fibre (Publication I, Publication II, Publication III), the fibre surface (Publication I, Publication III) and the mechanical performance of the fibre–matrix bond (Publication IV) are explained in that order. The novelty of the measuring methods and the data analysis of the results are also described.

The results and observations are discussed in the chapter Results and discussions and follow the same sequence as that of the publications in the chapter Methodology. First, the distributions of CH, C-S-H, unhydrated (UH) cement grains and pores in the cement paste near the fibre are reported through the comparison of experimental approaches. Then, the roughness and wettability of the steel fibre surface are examined, and their effects on the formation of the fibre–matrix bond are discussed. The effect of the measured fibre properties on the microstructure of the cement paste surrounding the fibre is analysed and explained based on the outcomes of the previous two sections. The mechanical response of the fibre, which is based on the bond between the cement paste and steel fibres with different surface roughness, to the gradually increasing tension cycles is measured, and the deterioration of the fibre–matrix bond prior to the peak load is discussed while taking into consideration the micro-scale properties measured for the fibre and the cement paste.

The major conclusions and future perspectives are discussed in the chapter Conclusions.
Introduction

Objective 1: Determine the relationship between the micro-scale properties of the fibre and the cement matrix surrounding it.

Research question 1.1: Which characteristics of the fibre and the cement paste surrounding it should be considered to define the strength of the fibre–matrix bond? (Pub. 1, 2)

Research question 1.2: How does the steel fibre surface influence the microstructure of the cement paste around it? (Pub. 3)

Research question 1.3: How do the changes in properties of the steel fibre and the cement paste surrounding it influence the response of the fibre–matrix bond to the tensile load? (Pub. 4)

Objective 2: Advance the micro-scale investigations of the fibre-matrix interface by applying modern experimental techniques.

Research question 2.1: What methodologies need to be applied and how should they be used to answer the research questions of Objective 1?

Outcomes

Objective 1: Surface roughness of the steel fibres affects the microstructure of ITZ, thus, the capacity and degradation of the fibre–matrix bond.

Objective 2: Three-dimensional microstructure of ITZ, mobility of water along the fibre surface and fracture of the fibre-matrix bond with gradual increase of load amplitude were defined by applying modern experimental techniques.

Figure 1.2. Flowchart of the thesis in the sequence of the research questions.
2. Methodology

2.1 Experimental techniques

SEM has been widely applied to study the ITZ around aggregates and steel bars in the cementitious matrix [10, 28–32]. In the case of the interface between fibres and cementitious matrix, SEM has been used for qualitative observations, which provided supporting information for the results measured using nanoindentation [6, 15] or mechanical testing, such as bending or pull-out tests [16, 19, 33]. The morphology of the fibre surface was examined before and after a pull-out test using SEM in [34–36] to detect the amount of the cement paste left on the fibre surface. Bentur et al. in [37] took SEM images of the fibre–matrix interface before, during and after the fibre pull-out, which were used as the basis for understanding the precipitation and deformation of the cement hydrates near the fibre. The authors observed a duplex film of $1 - 2 \, \mu m$ that consisted of CH and C-S-H phases, followed by large agglomerations of CH crystals up to a distance of $40 \, \mu m$ from the fibre surface that transitioned into a high-porosity layer.

Hwang et al. in [14] evaluated the porosity formed around steel fibres based on a backscattered electron (BSE) image analysis. Their results indicated the existence of great porosity up to a distance of $270 \, \mu m$ from the fibre surface; however, in their study, an increase in porosity could have developed during the oven-drying of the samples before the SEM investigation. Lee et al. in [5] quantified the microstructure of the ITZ in terms of the distributions of pores and UH cement grains, which were calculated based on the BSE image analysis, with a step of $10 \, \mu m$ and the distribution of the Ca/Si ratio with a step of $2 \, \mu m$ from the fibre surface. The authors reported a decrease in porosity near the steel fibres with a decrease in the w/c ratio and an addition of silica fume. They also observed a non-continuous layer of the CH and C-S-H phases at the surface of the steel fibres, followed by the precipitation of CH for the next $4 \, \mu m$. The obtained information about the chemical and microstructural properties
of the ITZ near the fibres was correlated with the results received from
the macro-scale bending and compressive tests. Pi et al. in [38] made
an attempt to improve the fibre–matrix bond by brass- and nano-SiO$_2$-
coating the steel fibres and evaluated the effect of coating by measuring
the distributions of the pores, UH cement grains and CH phases from
the fibre surface based on the analysis of BSE images. However, the
quantitative information available about the ITZ around the fibres in the
cementitious matrix is still limited for obtaining a robust understanding of
the origins of the fibre–matrix bond formation.

NANO-indentation was applied in [6, 7, 15] to study the mechanical prop-
erties of the interface between the fibres and the cement paste, such as
hardness and the elasticity modulus. Pinchin et al. in [7] measured the
hardness gradient of the interface between a steel wire with a diameter of
0.807 mm and cement paste. The authors reported a decrease in hardness
that extended up to a distance of 0.75 mm from the wire surface. Wang et
al. [6] used an indentation step of 10 or 11.4 μm and covered the cement
paste up to a distance of 80 μm from the fibre surface. The results indicated
that the hardness of the cement paste near the surface of the steel fibre
increased with a decrease in the w/c ratio and the addition of silica fume.
In another study [15], four areas with sizes of 120×80 μm$^2$ and 80×60 μm$^2$
were studied line-wise with nanoindentation using indentation steps of
5 μm from the tangent plane of the fibre surface and 10 μm spacing between
these lines for the steel and polypropylene (PP) fibres, respectively. The
results revealed a decrease in the hardness and elastic modulus with an
increase in the w/c ratio near both steel and PP fibres. The latter nanoind-
entaion results were analysed with deconvolution technique and coupled
with the X-ray diffraction results, which identified the volume fractions
of each phase and importance of C-S-H and porosity for the mechanical
properties of the fibre–matrix bond. The thickness of the ITZ near the steel
fibres with a diameter of 0.5 mm was 30 μm and that near the PP fibres
with a diameter of 30 μm was 15 μm.

Since the stress transferring along the tensile zone in FRCC elements
depends on the pull-out behaviour of each fibre during crack bridging, the
pull-out test is usually employed to study the mechanical performance of
the fibre–matrix bond. The pull-out test quantifies the displacement or slip
of the fibre as a function of the load [3]. The configuration of the specimens
and the setups used for the execution of the pull-out test vary greatly
[39]. Pulling out a single fibre from the dog-bone specimen is a common
method for measuring the response of a fibre–matrix bond. The pull-out
test is also used to evaluate the changes in the fibre–matrix bond due to
the modification of the fibre surface or matrix composition [23, 24, 36, 40].
The measured load-slip curves are also used as a basis for developing the
models used for predicting the response of the fibre–matrix bond to tensile
loads [41, 42]. The first theoretical assumptions that were based on the
Methodology

Experimental results and that described the load-slip relationship and stress distribution of the fibre–matrix bond under a tensile load were made by Naaman et al. in [17]. Based on this theory, the measured load-slip curves include three fracture stages of the fibre–matrix bond: linear-elastic, debonding and frictional slippage (Fig. 2.1). The authors assumed that, during the linear-elastic stage, the bond between the fibre and the cement paste is complete and that the displacements of the fibre and the cement paste are compatible. This implies that the slip is zero from the beginning of loading up to the formation of the first crack in the fibre–matrix bond, which is when maximum bond stress is reached. Within this stage, the slip measured with the increase in the load is assumed to be attributed to the elastic extension of the fibre–matrix bond, which is mainly defined by the elastic characteristics of the cementitious matrix bonded to the fibre. However, the linear elasticity of the first stage of the bond response was not proved experimentally, and the slip generated may also include the elastic extension of the steel fibre.

![Figure 2.1. Stages of the fibre-matrix bond response to the tensile load based on [17].](image)

The pull-out results of the fibres studied with monotonic tensile loading directly provide the values of the maximum pull-out capacity of the fibre–matrix bond and the maximum load reached during frictional slippage. The average value of the bond shear stress can be calculated by assuming the complete bond between the fibre and the cementitious matrix [36, 43]. However, this is not the most reliable interpretation of the pull-out results, as a discontinuity of the fibre–matrix bond was reported by SEM investigations in [5, 14, 37]. The micro-scale properties of the fibre–matrix bond cannot be directly extracted from the results of the pull-out test carried out with monotonic loading [44]. Therefore, the conventional pull-out test needs to be modified.

To study the detailed stress distribution along the continuous steel fibre when it is being pulled out from the cement paste, Moire interferometry was used in [45]. The authors observed high-stress concentrations in a zone
that was between 40 and 70 μm around the fibre surface. However, this technique does not provide microstructural information about the boundary zone with a sufficient resolution that can allow to draw conclusions about the deformation of the fibre–matrix interface.

2.2 Quantification of the ITZ microstructure

2.2.1 Scanning electron microscopy

SEM enables the investigation of the cementitious matrix in two dimensions [46, 47]. However, previous research that concentrates on the microstructure of the cement paste near the fibres is limited. In addition, numerous SEM images of the cement paste obtained within previous decades have lacked the resolution required to quantitatively evaluate the composition of the cement paste and, specifically, the ITZ [48].

Moreover, in a number of studies [14, 49, 50] that have investigated the fibre–matrix interface, the samples were cured in an oven prior to SEM examination. This was done to stop hydration and remove any free water from the cementitious matrix that can harm the SEM; but this might have caused irreversible damage to the cement hydrates and the structure of pores [51, 52].

The present study used an environmental SEM operating in a low-vacuum mode at a pressure of 60 Pa, which enables the investigation of samples that contain liquids. In addition, the partial collision of the incident electrons from the beam with the water molecules in the chamber forms positive ions that settle on the surface of non-conductive cement paste samples and create a conductive layer [53]. This phenomenon enables the analysis of the cementitious samples without requiring a coating of a conductive metal, such as gold or carbon. These are often used to prevent the samples from becoming charged but may affect the SEM observations [51].

Cement paste samples with the embedded steel fibres were prepared using CEM II/B-M (S-LL) 42.5N, which is a widely used cement clinker, and the cold-drawn steel fibres HE 1/60 with a diameter of 1 mm. The chemical composition of the cement clinker is reported in Publication I. To study the microstructure of the cement paste in the vicinity of the fibre exactly at 28 days, the hydration of the cement paste was stopped using the solid exchange method by soaking the samples in ethanol for several days based on the size of the studied sample.

An SEM investigation with BSE and energy dispersive x-ray (EDX) detectors requires a well-polished cross-section of the material in order to eliminate any topographical defects that can result in the incorrect charac-
terisation of the material. The difference in the hardness between the steel fibres and cement hydrates complicates the polishing process. Inaccurate polishing and cutting may result in the erosion or crashing of the ITZ layer, which will affect measuring results [54, 55]. Therefore, the grinding and polishing processes were developed based on the recommendations from the literature [5, 6, 51, 56] and numerous experimental trials. The procedure of sample preparation is explained in more detail in Publication I and Publication III. The developed grinding and polishing program is illustrated in Table 2.1. The state of the sample surface after each polishing step was checked using the optical microscope.

**Table 2.1.** Grinding and polishing parameters applied on epoxy impregnated samples.

<table>
<thead>
<tr>
<th>Step</th>
<th>Plate</th>
<th>Lubricant</th>
<th>Abrasive</th>
<th>Time, Force, Rotation</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>Diamond pad (125 μm)</td>
<td>Industol</td>
<td>Remove excess epoxy</td>
<td>Manual 150</td>
</tr>
<tr>
<td>2</td>
<td>1200 grade SiC</td>
<td>Industol</td>
<td>Remove excess epoxy</td>
<td>Manual 150</td>
</tr>
<tr>
<td>3</td>
<td>MD-Largo</td>
<td>Lamp oil</td>
<td>9 μm diamond spray</td>
<td>15 15 150</td>
</tr>
<tr>
<td>4</td>
<td>MD-Largo</td>
<td>Lamp oil</td>
<td>3 μm diamond spray</td>
<td>45 20 150</td>
</tr>
<tr>
<td>5</td>
<td>DP-Dac (cloth)</td>
<td>Lamp oil</td>
<td>1 μm diamond spray</td>
<td>60 25 150</td>
</tr>
</tbody>
</table>

**Analysis of the BSE images**

The BSE detector provides high-resolution grey-level images of the cement paste microstructure and enables the examination of the distribution of the cement paste phases. The phases of the cement paste in the BSE images contain different shades of grey that vary between 0 (black) and 255 (white) depending on the atomic number (Z) of their chemical elements. The most abundant chemical elements in the studied cement clinkers are Ca (Z = 20), Si (Z = 14), Al (Z = 13), Fe (Z = 26) and Mg (Z = 12) according to the manufacturer. When no chemical elements are detected in the scanned regions, they appear as black pixels on the BSE images and indicate pores. Apart from their grey-levels, different cement hydrates and UH cement grains can also be distinguished based on their morphology or hydration rim, as demonstrated in Fig. 2.2.

In the present study, the BSE images of the boundary zone near the steel fibres were obtained with a resolution of 0.1 μm/px, which covered the porosity on the scale of the coarse capillary pores [57]. Around 16 BSE images were taken per sample to cover the entire fibre–matrix interface (Fig. 7 in Publication III). The steel fibres that mainly contain Fe appear as the brightest phase in the BSE images. An example of the analysed BSE images is shown in Fig. 2.3(a). To analyse the distributions of the pores and the UH cement grains with the distance from the surface of steel...
fibres, a suitable segmentation method should be chosen.

The global threshold method is usually applied for the segmentation of cement paste phases from BSE images. This method uses the grey-level histogram of the image to define the thresholds or the grey-level values, which separate different phases from each other. Each peak on the histogram corresponds to a specific cement phase (Fig. 6 in Publication II). This segmentation method was first applied by Scrivener et al. in [58] to study the cement paste and in [59] to quantify the microstructure of the boundary zone around aggregates. The following studies applied the global threshold method to investigate the gradients of pores, UH grains or CH phases in the cementitious matrix near inclusions, such as rebars [30], aggregates [29, 32] and fibres [5]. However, the recognition of the thresholds between the C-S-H and pores, CH and slag or other SCM may be complicated due to the absence of the corresponding peak on the histogram. Recent studies have begun to improve previous methods or apply new methods for the segmentation of BSE images. Wong et al. in [60] introduced a method for defining the threshold of pores by using the inflection point at the cumulative grey-level histogram in cases where the peak that corresponds to the pores was not evident. Kenny et al. in [49, 61] used the mean-shift (MS) method to segregate the pores and UH
grains from the cementitious matrix near the reinforcement bars. The MS algorithm does not use the grey-level histogram for the segmentation of phases but considers the pixels of the BSE image as data points of a probability density function, where the densest regions are defined as the local maxima of the point distribution and are interpreted as the centroids of the clusters. This eliminates the problems associated with threshold detection. Another algorithm that is similar to the MS method is the k-means clustering algorithm. The k-means clustering also considers pixels as data points, which are grouped by this algorithm into a predefined number of clusters. The number of clusters is chosen based on the minimum clustering error (Fig. 2.3(b)), which entails that the update of the centroids of the clusters stops when the sum of the squared values of the distances between the data points and centroids of each cluster is minimum, as explained in [62, 63]. Both clustering algorithms were applied for the

Figure 2.3. (a) An example of BSE image of the fibre–matrix interface. (b) Clustering error as a function of a number of clusters. (c) All phases and (d) UH cement grains and pores segmented with the k-means clustering algorithm.

segmentation of cement paste and were compared in Publication III. Since the phases of cement paste can contain sub-phases or fragments of other phases, such as fragments of aluminates in the belite (Fig. 2.2(b)) [47], the results obtained with MS clustering varied from three to six clusters, resulting with a high clustering error in the case of three and four clusters.
Methodology

In the case of k-mean clustering, the pre-defined number of clusters used was five based on the clustering error calculation, which improved the quality of the segmentation (Fig. 2.3(c)). Therefore, the k-means clustering algorithm was adapted to segment pores and UH cement grains in the present research. The distributions of the UH cement grains and pores were calculated as the area fraction of each phase per band of 5 μm in width for a distance of 100 μm from the fibre surface (Fig. 2.3(d)).

EDX analysis

The EDX detector was used to examine the distributions of chemical elements and the cement hydrates, such as CH and C-S-H, with the respective chemical composition in the cement paste. The perception and definition of the cement hydrates based only on the shades of grey may be subjective and affected by the contrast and brightness settings of each microscope. In the case of CH and limestone or different clinker grains, the shades of grey may be similar or barely distinguishable from each other. Therefore, the chemical analysis of these phases can resolve the uncertainty related to phase definition. The results of the EDX analysis are represented with the atomic percentage (at. %) of each detected chemical element per scanned point of the material. Most of the phases in the cement paste can be defined based on the ratios of Ca, Si and Al. The scatter plot of the Al/Ca versus Si/Ca ratios is often used to determine the intermixing of the phases and the variation in the C-S-H composition [51]. Even the three-dimensional scatter plots with Al/Ca, Si/Ca and S/Ca ratios can be analysed [64]. In the case of slag, the mapping of Mg distribution works as a slag marker, and the chemical mapping can be overlaid with the BSE image to segment the slag grains, as was introduced in [65].

Based on the literature, a clear variation in CH precipitation has not been observed further than 45 μm from the surface of inclusion [5, 30, 37]. Therefore, in the present research, x-ray line analysis was used to determine the changes in the distributions of the CH and C-S-H phases from the surface of the steel fibres up to a distance of 60 μm with steps of 0.5 μm (Fig. 2.4). Four lines were measured in four different directions from the fibre surface for each sample. In total, five samples were studied. The C-S-H and CH phases were targeted as C-S-H is the main binding hydrate in the cementitious composites that defines the strength of the material, and the CH near the fibre surface results in a brittle failure of the fibre–matrix bond under loading according to the outcomes reported in [5]. Each measured point along the line was interpreted as a specific phase based on the scatter plot of the Al/Ca versus Si/Ca ratios, which is further discussed in the results section 3.1.1. In addition, the chemical measurements of the corresponding chemical composition of the CH and C-S-H phases are demonstrated in Publication I (Figs. 12-14).
2.2.2 Micro-computed tomography

\( \mu \text{CT} \) enables the non-destructive examination of the volumetric inner structure of the materials through the analysis of the attenuation of the x-ray waves transmitted through the studied material. When the x-ray passes through the studied object, its energy is absorbed by the atoms of said object, which entails that this x-ray is attenuated. The information about the attenuation of the transmitted x-rays captured by the detector represents the projections of the studied object that was rotated by 360°. The projections are then reconstructed into the stack of the cross-sectional images that illustrate the scanned material. The absorption of x-rays and the shade of grey of the material compounds on the reconstructed images are connected to the density of the scanned compounds. The lower the absorption of the x-rays is, the darker the compound will appear on the reconstructed image. In the case of the cementitious matrix, each cement phase has a different density or a density variation that can overlap with other phases, as demonstrated in Table 2.2. However, the density values of UH cement grains and pores are significantly larger than those of the hydrates. Therefore, \( \mu \text{CT} \) has already been used to examine the porosity network, which is connected to the strength and permeability of the composite, and the distribution of UH cement grains, which can provide information about the hydration rate of the cementitious matrix [9, 52, 66–68]. The recent study reported in [69] managed to implement nano-CT together with nano-x-ray fluorescence, which provided chemical information about \( \text{C}_3\text{S} \) composition, although this coupled approach has not been yet applied to larger volumes of cement paste or the ITZ microstructure. The utilisation of \( \mu \text{CT} \) eliminates the main limitations of the SEM investigation, which are the damage caused to the microstructure of the cement paste during sample preparation and the two-dimensional
nature of the SEM method [52].

**Table 2.2.** Densities of the most abundant phases in the cement paste.

<table>
<thead>
<tr>
<th>Phase group</th>
<th>Chemical name</th>
<th>Formula</th>
<th>Density (kg/m$^3$)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>UH grains</td>
<td>alite</td>
<td>$3\text{CaO} \cdot \text{SiO}_2 = \text{C}_3\text{S}$</td>
<td>3130–3220</td>
<td>[70, 71]</td>
</tr>
<tr>
<td></td>
<td>belite</td>
<td>$2\text{CaO} \cdot \text{SiO}_2 = \text{C}_2\text{S}$</td>
<td>3280–3310</td>
<td>[70]</td>
</tr>
<tr>
<td></td>
<td>aluminate$^1$</td>
<td>$3\text{CaO} \cdot \text{Al}_2\text{O}_3 = \text{C}_3\text{A}$</td>
<td>2560–3064</td>
<td>[70, 71]</td>
</tr>
<tr>
<td></td>
<td>ferrite</td>
<td>$4\text{CaO} \cdot \text{Al}_2\text{O}_3 \cdot \text{Fe}_2\text{O}_3 = \text{C}_4\text{AF}$</td>
<td>3480–4026</td>
<td>[70, 71]</td>
</tr>
<tr>
<td></td>
<td>slag</td>
<td>-</td>
<td>2900</td>
<td>[72]</td>
</tr>
<tr>
<td></td>
<td>limestone/calcite</td>
<td>$\text{CaCO}_3$</td>
<td>2709–2927</td>
<td>[71, 73]</td>
</tr>
<tr>
<td>Hydrates</td>
<td>portlandite</td>
<td>$\text{Ca(OH)}_2 = \text{CH}$</td>
<td>2242</td>
<td>[71]</td>
</tr>
<tr>
<td></td>
<td>calcium silicate hydrate</td>
<td>$x\text{CaO} \cdot \text{SiO}_2 \cdot y\text{H}_2\text{O}$</td>
<td>1440–2030</td>
<td>[74–76]</td>
</tr>
<tr>
<td></td>
<td>ettringite/AFt</td>
<td>$6\text{CaO} \cdot \text{Al}_2\text{O}_3 \cdot 3\text{SO}_4 \cdot 32\text{H}_2\text{O}$</td>
<td>1775–1886</td>
<td>[71]</td>
</tr>
<tr>
<td></td>
<td>AFm</td>
<td>$4\text{CaO} \cdot \text{Al}_2\text{O}_3 \cdot x\text{H}_2\text{O}$</td>
<td>1802–2170</td>
<td>[71]</td>
</tr>
<tr>
<td>Porosity</td>
<td>0</td>
<td></td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

$^1$ Aluminate was not present in cement clinker used.

$^2$ $x, y, z =$ coefficients that indicate the variable amount of corresponding compound; $X = \text{OH}^-, \text{SO}_4^{2-}, \text{CO}_3^{2-}$ anions.

However, $\mu$CT has limitations when examining heterogeneous cementitious composites, such as those related to distinguishing the micro-scale porosity, cement phases and ITZ microstructure [77, 78]. Several studies that have investigated the porosity network of the cement paste highlighted the suboptimal resolution of the received $\mu$CT reconstruction images for investigating the capillary porosity [52, 79]. A higher resolution of the $\mu$CT images can be obtained by scanning a smaller volume of material, whereas, for the scans of large material volumes the resolution should be sacrificed. The authors in [52, 79, 80] managed to obtain the $\mu$CT images of plain cement paste with a voxel resolution of $0.5 – 0.684 \mu\text{m}$. However, the size of the scanned volumes varied from only $100 – 600 \mu\text{m}^3$.

The application of $\mu$CT for examining FRCC on the meso- or macroscales has recently evolved and was reported in [81–83]. These studies covered the scanning volumes of $35 – 750000 \text{mm}^3$ with a voxel resolution of $3.32 – 60 \mu\text{m}$, which enabled the investigation of the distributions of meso- and macro-scale pores and aggregates, and fibres orientation. Lavrov et al. in [84] attempted to access the ITZ between cement paste and the wall of a glass tube in which the cement paste was cast. The authors managed to identify the ITZ thickness of $20 \mu\text{m}$ by analysing the distribution of the sizes of the UH cement grains from the wall of the glass tube. However, the ITZ around the fibres may have not been previously studied using $\mu$CT. The investigation of composite materials using $\mu$CT is also complicated by the difference in the densities of their constituents. In the case of FRCC, the density of steel fibres is around $7850 \text{kg/m}^3$, whereas the UH cement grains have a highest density of around $4026 \text{kg/m}^3$. This difference in densities results in the over-brightening of the steel fibres, which obscures
the microstructure of the ITZ, as demonstrated in Fig. 2.5(a). To obtain μCT images with a sufficiently visible ITZ microstructure in order to be able to distinguish different cement paste phases, PP fibres with a density of 900 kg/m³ were used in the present study (Fig. 2.5(b)). Due to the low density of the PP fibres, the cement paste appears brighter on the received images. Another factor that affects resolution is the source of the x-ray radiation. The energy level of the x-ray radiation recorded with laboratory sources is not capable of producing μCT images that are of sufficient size and resolution to allow the study of the ITZ microstructure. To produce x-ray radiation with high energy, a synchrotron radiation source can be used.

![Image](actual_fibre_size_overbrightening.jpg)

(a) Steel fibre. (b) PP fibre.

**Figure 2.5.** Examples of the reconstructed μCT images that represent the cross-sections of the steel and PP fibres embedded in the cement paste.

Taking into consideration the scope of this study, the microstructure of the cement paste around the PP fibres was studied with regard to the distributions of pores and UH grains. The European Synchrotron Radiation Facility, which generates monochromatic and coherent x-ray radiation, was used. This allowed the examination of a volume of 2.22 mm³ with a voxel resolution of 0.637 μm/px of three cement paste samples with the embedded PP fibre. A more precise configuration of the beamline used is reported in Publication II. In addition, phase-contrast tomography was used, which improves the contrast between the compounds of a studied material by identifying the phase shifts of the transmitted x-ray waves or changes in the x-ray propagation direction, as explained in [85, 86]. The principles of the performed μCT measurements are briefly described in Fig. 2.6 and explained in more detail in Publication II.

The cement paste phases, mainly pores and UH grains, were conventionally segmented from μCT images based on the grey-level histogram, which
is the same segmentation approach that is used for BSE images [9, 52, 80]. In the present study, three phases were segmented from the scanned cement paste: pores, UH grains and other hydrates. The thresholds for the segmentation of the mentioned phases were calculated as the average grey-level values determined based on the grey-level histogram, the cumulative histogram of grey intensities, the over-flow criterion and the measurement of the grey-levels of the corresponding phases. The segmentation methods used are explained in detail in Publication II.

2.3 Properties of fibre surface

2.3.1 Treatment of the fibre surface

The effects of the roughness of fibre surface on the maximum capacity of the fibre–matrix bond and the post-peak resistance of the fibre to sliding have already been studied in [24–26]. An increase in the roughness of the fibre surface facilitates the occurrence of slip-hardening behaviour during the fibre slippage stage [24]. However, the effect of the surface roughness of the fibres on the wetting of these fibres, the formation of the microstructure of the surrounding cement paste and the pre-peak response of the fibre–matrix bond to the load have barely been considered in previous studies. Since the significant effect of surface roughness on the wettability of different materials was highlighted in [87, 88], it is important to research the effect of the surface roughness of fibres on the properties of FRCC.
To address these questions, the surface roughness of steel fibres was modified through electrolytic polishing in a solution of ethanol and nitric acid (2:1) at $-30^\circ$ C and by sanding the surface of the fibres in the direction perpendicular to their length using sandpaper with a grit size of $60 \mu m$. Three types of surface roughness (with corresponding notations) were further considered: electrolytically polished (R1), non-processed (R2) and sanded (R3). The descriptions of the sanding and polishing processes are reported in Publication III. The obtained SEM images of the fibre surfaces are presented in Fig. 2.7.

![SEM images of steel fibres with treated surfaces obtained using secondary electron detector](image)

**Figure 2.7.** SEM images of the steel fibres with treated surfaces obtained using secondary electron detector.

### 2.3.2 Scanning electron microscopy

Considering the scope of the present research, the surface roughness of the steel fibres was first evaluated by analysing the BSE images, which enabled the estimation of the size of the cement or SCM grains that can fit between the irregularities of the fibre surface. For this purpose, BSE images with a magnification of 240x and resolution of $1 \mu m$ were used to cover the entire cross-section of the steel fibres. The fibres were segmented as a phase with the largest grey-level values. Further, its profile was extracted as an outline, which was then unfolded, and the coordinates of each pixel on the received profile were obtained and analysed (Fig. 2.8).

The evaluation of the surface roughness of the fibre cross-section can be coupled with the chemical analysis of the composition of the surrounding cement paste. The analysis of BSE images for the estimation of the surface roughness of fibres does not represent a direct measuring technique and can only evaluate the roughness of one cross-section. Therefore, the results can be affected by the resolution of the BSE images or sample preparation. However, this technique can provide an average estimation of the fibre roughness combined with information about the distribution of the hydration phases.
Methodology

(a) BSE image of fibre cross-section

(b) Unfolded profile of fibre cross-section

(c) Calculation of height and wavelength of the fibre surface roughness.

Figure 2.8. Analysis of the surface roughness of fibre cross-section using BSE images.

2.3.3 Atomic force microscopy and stylus profilometer

An atomic force microscope (AFM) was employed to directly quantify the surface roughness of polished and non-processed fibres. AFM has been previously used in [16] to measure the surface roughness of steel and PP fibres, which has been connected to the wetting properties and the pull-out resistance of these fibres and the shrinkage of the geopolymer around them. The studies reported in [24, 89] also used an AFM to quantify the surface roughness of different types of steel fibres and examine its effect on the changes in the pull-out resistance of these fibres. Since the AFM is used for measuring nano-scale irregularities on the surface, its measuring area is usually limited. Therefore, the surface area of the fibres evaluated in the aforementioned studies varied from $5 \times 5 \mu m^2$ to $40 \times 40 \mu m^2$. In the case of steel fibres with a length in centimetres, the evaluation of the surface roughness of a small area may not represent the surface roughness of an entire fibre. However, the size of the scanned area is also influenced by fibre curvature and roughness. In the present research, the area of $80 \times 80 \mu m^2$, which is the largest area that can be scanned by the device used, was measured to quantify the surface roughness of polished and non-processed fibres. The scanned areas of the steel fibres used had a curved surface, which was flattened in the analysis software to evaluate the actual surface roughness. In total, 256 lines were scanned within the studied area and were interpreted as a three-dimensional map of the fibre surface area, as illustrated in Fig. 2.9(b).

The stylus profilometer was used to directly measure the surface roughness of the sanded fibres that exceeded the measuring limits of the AFM. In contrast to the AFM, the stylus profilometer can measure only the profile of the surface roughness as a single line. The schematic illustration of the measuring positions along the fibre surface with both the AFM and the
Methodology

A basic understanding of the water movement in cement-based composites is required to produce composites that have adequate performance [90]. A fresh cement paste is a water-based suspension. During the mixing and setting of fresh FRCC, the suspension mobility is important for the formation of the hardened microstructure of the cement matrix. The wetting of the fibres is a key factor that defines the hydration of the cement clinker and the distribution of the cement hydrates around the steel fibre. Since mixing is a dynamic process, the characterisation of the wetting parameters of the fibre surface can help evaluate the change in the water movement on the fibre surface. The authors in [13] considered the effect of the orientation of steel fibres on the concrete bleeding through the flow channels, which form around the fibres during mixing. The self-weight of the cement matrix creates hydrostatic pressure around the fibre. When the fibre is inclined in vertical plane, the pressure difference at the two ends of the fibre facilitates water migration along the fibre surface (Fig. 2.10). The larger the orientation angle between the horizontal plane and the fibre is, the more water moves to the top of the cast element and leads to the bleeding of the composite. Therefore, the orientation of the fibres allows the trapping of water on the fibre surface and decreases the overall bleeding of the composite. The authors also considered the fibre surface roughness as a parameter that resists the water migration along the fibres. Around the fibres oriented horizontally, the agglomeration of water results in:

**2.3.4 Contact angle goniometry**

Figure 2.9. Analysis of the surface roughness of different types of the steel fibres. Orange square and blue line represent area measured with AFM and line measured with stylus profilometer, respectively.
in the formation of large voids beneath the fibre surface, which are also governed by the gravity-induced movements of the water [6]. The existence of large pores near the surface of the fibre decreases the strength of the fibre–matrix bond and facilitates the transportation of chemicals, such as chloride, sodium or carbonate, which degrade the microstructure of the cement paste [14].

Figure 2.10. Representation of the water movements induced by the gravity forces and affected by fibre roughness in the fresh cement paste along the surface of the vertically inclined fibre.

The wettability of the fibre surface reported in [16] was determined by defining the static contact angle between the air–water and water–solid contact lines. However, the value of the static contact angle may vary between the values of the receding and advancing contact angles [91]. The advancing contact angle \( \theta_a \) defines the ability of the water to wet the dry surface, whereas the receding contact angle \( \theta_r \) explains the ability of water to stick to the wet surface. The difference between \( \theta_r \) and \( \theta_a \) is called hysteresis, and this characterises the mobility of the water along the surface of the studied material. Given the scope of the present study, \( \theta_r \) and \( \theta_a \) were measured for the fibres with different types of surface roughness (Fig. 2.11). The advancing contact angle was measured by increasing the volume of the water droplet using the sessile method for all types of fibre surfaces, as shown in Fig. 2.11(a). In the case of the receding contact angle, the measuring method was governed by the value of surface roughness. The sessile method with a decreasing volume of the droplet was used for non-processed and sanded fibres (Fig. 2.11(b)), whereas the meniscus method was used for polished fibres (Fig. 2.11(c)). In the meniscus method, the fibre pulled up from the water drags the water up, causing the water to recede. The measuring procedures and limitations of each measuring method used are explained in Publication III.
Methodology

Fibre

water inlet
0.1 µl/s
droplet

(a)

0.1 µl/s
water outlet
droplet

(b)

Air
Fibre
direction of fibre pulling out

(c)

Water

Figure 2.11. Photos of the measuring method employed for the evaluation of the contact angles: (a) an advancing and (b) receding sessile droplet and (c) meniscus methods.

2.4 Response of the fibre–matrix bond to the uniaxial tension cycles

2.4.1 Fibre pull-out test

Most concrete structures are affected by not only a constant dead load but also live loads, which can change over time. The repeated loading causes the accumulation of damage and the degradation of the material, which is more common than the failure caused by reaching the ultimate design load. A cyclic compression has been previously applied to study the changes in the stiffness and residual deformation of FRCC and plain concrete up to failure [92, 93]. Investigations of rocks under cyclic compression have explained the degradation of brittle rocks such as sandstone or basalt [94, 95]. Since pull-out of the fibres have not been tested using tension cycles with increasing amplitudes of loading, it was applied within the scope of this research, as this can provide additional information about the degradation of the fibre–matrix bond during each cycle.

To capture micro-scale deformations in the fibre–matrix bond with each load cycle, an appropriate rate of loading needs to be selected. The results related to the sensitivity of the pull-out response of steel fibres to the rate of the loading were a bit contradictory. Nieuwoudt et al. in [96] found an insignificant effect of the loading speed, which varied between 0.015 and 150 mm/min, on the load-slip curves obtained by pulling out straight steel fibres. In contrast, Park et al. in [97] reported an increase in the maximum capacity of the fibre–matrix bond with the growth of the loading speed from 1 mm/min to 100 mm/min. The authors in [97] explained that, at low rates of loading, the debonding crack has enough time to find the weakest regions on the fibre–matrix interface, which does not occur for higher rates of loading.

Considering the scope of this study, pull-out tests for steel fibres with
different surface roughness were performed using a loading rate of 0.001 mm/min and an increase in the load by 10 N with each loading cycle. The low rate of loading used was applied to track even minor deformations, which were measured using two linear variable differential transformers (LVDTs). Cement paste cylinders with a diameter of 45 mm and height of 60 mm were prepared with the steel fibre embedded in the middle of the cylinder for 30 mm. The samples were tested after being cured in water for 28 days. The pull-out setup was designed to eliminate any undesirable compression in the fibre–matrix bond by transferring the load directly from the fibre to the surrounding matrix. The scheme of the setup is illustrated in Fig. 2.12 and described in more detail in Publication IV. The evolution of the residual slip increment and the stiffness of the bond during the application and removal of the load per cycle were the parameters studied. The measurement of the residual slip with an increase in load can help identify the existence or absence of a complete bond between the fibre and the cement paste and define the mode of bond degradation. The development of the stiffness of the bond can indicate the compaction or damage of the matrix near the fibre.

Figure 2.12. Pull-out setup.
2.4.2 Scanning electron microscopy

The pattern of the debonding crack developed along the fibre–matrix interface can extend to the surface of the fibre or to some distance from the fibre [98]. As the surface roughness of the fibre may affect the micro-scale properties of the fibre–matrix interface [99], the failure pattern can also be affected by changing the response of the fibre–matrix bond to tensile loading. The SEM examinations of the surface of the pulled-out fibres have often been employed to explain the results of the pull-out tests [23, 35, 36, 43]. In this study, SEM was used to estimate the area fraction of the cement paste that was adhered to the surface of the pulled-out fibres. This information was used for supplementary physical reasoning associated with the degradation of the fibre–matrix bond by indicating the location of the bond failure within the boundary zone.
3. Results and discussions

3.1 Microstructure of the cement paste near the fibre surface

3.1.1 Distribution of hydrated cement phases near the fibre surface

The analysis of the chemical composition considered in Publication I is demonstrated in Fig. 3.1. The Si/Ca and Al/Ca ratios of the C-S-H vary depending on the composition of the liquid solution available for the formation of hydrates at the beginning of hydration. The variation in the mentioned ratios is caused by a higher amount of Ca$^{2+}$ or Al$^{3+}$ ions in the inter-layers or the substitution of the bridging Si tetrahedra with Ca$^{2+}$ or Al$^{3+}$ ions in the C-S-H crystalline structure [100]. The variations in the Si/Ca and Al/Ca ratios are greater when slag and/or limestone are added into the concrete mix. Since the C-S-H formed around the alite grains has the most constant and densest composition, the Al/Ca and Si/Ca ratios of C-S-H were defined based on the EDX line analysis of the respective regions, which is demonstrated in Fig. 13 in Publication I. The variation in the Al/Ca and Si/Ca ratios detected in C-S-H around the alite grain is denoted by the blue box in Fig. 3.1(a), the borders of which represent the upper and lower values of the corresponding ratios. The distribution of the C-S-H with the distance from the fibre surface in Fig. 3.1(b) was based on the measured points that belonged to the values of the Al/Ca and Si/Ca ratios denoted by the blue box in Fig. 3.1(a). The phases with a Ca at. % larger than 75% were defined as CH and CaCO$_3$ and are demarcated in Fig. 3.1(a) using blue points. All other undefined points in Fig. 3.1(a) were left out from the analysis. However, apart from the UH grains of the cement clinker and slag, a large number of points left out from the analysis also represent C-S-H with a higher rate of inter-crystalline ion substitution. A possible variation in the C-S-H found in cement paste.
was reported in [101]. The C-S-H with Si/Ca and Al/Ca ratios other than those measured around alite grains also acts as a binding material in the cementitious matrix; however, such C-S-H has a larger intrinsic porosity and lower density than that found around alite grains [47, 75], and the measuring of the corresponding atomic ratios is complicated.

Figure 3.1. (a) Scatter plot of the Al/Ca vs. Si/Ca ratios of the all points measured. CH phases are demarcated using the blue dots. More or less pure C-S-H measured around alite grains is denoted by the blue dashed-line box. (b) Distributions of C-S-H and CH phases detected with the distance from the fibre surface.

The results of the CH and C-S-H distributions in Fig. 3.1(b) were interpreted as the number of detected points per measured step along the line. Since the EDX analysis is unable to detect pores, the results measured represent the chemical composition of the matrix underneath the pore, as can be observed in Fig. 2.4(b). Therefore, the C-S-H and CH phases were calculated as the sum of the detected points for every 0.5 μm step rather than the percentage of the material – the composition of which includes pores. The main variations in the distributions of the CH and C-S-H phases were detected within a distance of 20 μm from the fibre surface. The amount of the C-S-H phases at the fibre surface was greater than that of the CH phases – the highest value for which was detected at a distance of 8 μm, where the C-S-H amount was the lowest. However, the amount of the CH phases detected at the fibre surface was larger than that found at a distance of 15 μm and beyond. Even if the amount of the C-S-H detected near the fibre surface exceeded that of CH the phases, the amount of the CH phases was still high along the first 15 μm from the fibre surface. The C-S-H that was not included in the analysis, thus, also existed along a distance of 60 μm, resulting in an even higher amount of C-S-H near the fibre surface, which indicated a good bonding of the cement paste with fibre. However, the microstructure and distribution of the pores should also be addressed to estimate the density of the hydrates near the fibre surface. Based on the results presented in Fig. 3.1(a), the
presence of the AFt or AFm phases was not identified within the cement paste studied. The slag supplies additional Al\(^{3+}\) ions, and the calcite in the limestone reacts with the Al\(^{3+}\) ions to form hemi- and mono-carbonate phases [102], which lead to the refinement of the pores in the cementitious matrix. This should decrease the porosity of the cement paste near the fibres and improve the strength of the fibre–matrix bond. However, the absence of AFm phases indicated poor effect on the refinement of pores near the fibre in the present study.

3.1.2 Distributions of UH cement phases and pores around the fibre surface

The average distributions of the pores and the UH grains from the surface of PP fibres, which was measured using \(\mu\)CT, is demonstrated in Fig. 3.2. The difference in the densities of the cement clinker, slag and limestone grains is small compared to the other hydration phases (Table 2.2), which complicates the distinction between the aforementioned grains. Therefore, the cement clinker, slag and limestone grains were classified together as UH grains. The standard deviations of both the distributions of the pores and the UH grains decreased and stabilised by a distance of around 70 \(\mu\)m from the fibre surface. This indicates the boundary zone, which is even larger than the thickness of the ITZ layer detected in previous studies [15, 32, 45]. Despite the hydrophobic nature of the PP fibres that governs the formation of large pores near the fibre, the hydration of limestone and slag was assumed to reduce porosity due to the formation of calcium aluminate hydrates. However, the existence of large pores near the fibre surface, which were observed during the \(\mu\)CT scans (Fig. 2.5(b)), indicated the insufficient effect of the hydration of the limestone and slag grains on the refinement of pores within 28 days of hydration.

![Figure 3.2](image-url) Distributions of the pores and UH grains with the distance from the surface of PP fibres. The grey band demonstrates the standard deviation of the results.
Based on the results reported in this section and Section 3.1.1, CEM I 52.5N [103] was used for further investigations in Publication III and Publication IV to study the initial porosity and packing of the clinker grains near the steel fibres. CEM I 52.5N, as a reference matrix can provide information that can be used for choosing an appropriate blend in the future. The chemical composition of CEM I 52.5N is reported in Publication III.

The utilisation of \( \mu \)CT enabled the determination of the volumes of the pores that were formed in the vicinity of fibre surface (0 – 70 \( \mu m \)) and in the bulk cement paste (70 – 140 \( \mu m \)). The volume of the pores detected was also quantified, and its distribution is illustrated in Fig. 12 in Publication II. Most of the pores that were identified represented a mesh of pores that were connected with each other (Fig. 3.3). The occurrence of a pore mesh larger than 0.1 mm\(^3\) was greater within a distance of 70 \( \mu m \) from the fibre surface. The interconnected network of pores in Fig. 3.3(a) was detected in close vicinity to the PP fibre. Figs. 3.3(b) and 3.3(c) clearly indicate the interconnectivity of the pores with different volumes. The higher gradient of porosity near the fibres that can be measured using BSE images may not necessarily indicate high permeability of the FRCC, whereas the interconnectivity of the pores can be used to estimate the permeability of the composite [8] and study the movement of water through the network of pores [9].

![Figure 3.3](image)

**Figure 3.3.** Visualisation of the pores with different volumes detected using \( \mu \)CT.

Three-dimensional information about the particle size distribution (PSD) of UH grains in the cement paste for the studied period of hydration, in comparison to the initial particle size of the clinker grains, enables the examination of the changes in the volumes of grains with different sizes. It is also possible to estimate the degree of hydration when the initial volume of the cement clinker used is known. In the case of the measured results, the PSD for the used cement clinker was determined
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using laser diffraction analysis, and the PSD of UH grains after 28 days of hydration was analysed through $\mu$CT (Fig. 3.4(b)). The size of the UH grains detected with $\mu$CT was calculated as the average diameter of the measured values of the three diameters of a fitted ellipsoid, as demonstrated in Fig. 3.4(a). Based on Fig. 3.4(b), the grains with a size lesser than 4 $\mu$m were almost completely hydrated, whereas those larger than 60 $\mu$m had barely decreased. The asymmetrical change in the PSD after 28 days of hydration primarily indicated the hydration of $C_3S$ grains, as their rate of hydration is the largest among all other grains in the clinker, such as $C_2S$, slag and limestone, within this period of time [71].

![Figure 3.4.](image)

Figure 3.4. (a) Ellipsoid fitting to the UH grain detected using $\mu$CT. $D_1$, $D_2$, $D_3$ are the three diameters of the fitted ellipsoid. (b) Particle size distribution of the UH grains before the beginning of hydration and 28 days after hydration has started.

The application of $\mu$CT, which allowed access to the three-dimensional microstructure of the cement paste near the PP fibre, still yielded a resolution lower than that achieved by SEM – the main limitation of which is a two-dimensional field of investigation. Therefore, coupling the results received through the SEM and $\mu$CT investigations can reveal additional information regarding the interface between the fibre and the cementitious matrix, as has also been noted in [52]. According to Fig. 3.5 certain similarities in the distribution of cement paste phases, such as CH and pores, can be observed in both BSE and $\mu$CT images. This also verifies the correctness of the sample preparation technique used for BSE imaging.
3.2 Properties of the fibre surface

3.2.1 Definition of the fibre surface roughness

The methods used for measuring the surface roughness of the steel fibres in this research included the analysis of BSE images and measurements using an AFM and stylus profilometer. The surface profiles of fibres with different types of surface roughness that were examined using different measuring techniques are illustrated in Fig. 3.6 and quantitatively evaluated in Table 3.1.

Table 3.1. Roughness parameters of the steel fibres with the corresponding surface treatment obtained using BSE image analysis, AFM and stylus profilometer.

<table>
<thead>
<tr>
<th>Method</th>
<th>Fibre type</th>
<th>$l_{av}$ ($\mu$m)</th>
<th>$h_{av}$ ($\mu$m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>BSE image analysis</td>
<td>R2</td>
<td>31.50</td>
<td>1.85</td>
</tr>
<tr>
<td></td>
<td>R1</td>
<td>8.14</td>
<td>0.02</td>
</tr>
<tr>
<td></td>
<td>R2</td>
<td>21.50</td>
<td>0.23</td>
</tr>
<tr>
<td>AFM</td>
<td>R3</td>
<td>55.42</td>
<td>5.58</td>
</tr>
<tr>
<td>Stylus Profilometer</td>
<td>R3</td>
<td>55.42</td>
<td>5.58</td>
</tr>
</tbody>
</table>

Note: polished (R1), non-processed (R2), sanded (R3).

In the present research, the reliability of the BSE image analysis for surface roughness estimation was checked by comparing the parameters of the surface roughness of the non-processed (R2) fibres with those obtained using the AFM for the same fibre surface. Based on Figs. 3.6(a) and 3.6(c), the difference in the precision of the AFM and the analysis of the BSE images is obvious due to the resolution obtained using the methods. The analysis of BSE images is significantly dependent on their resolution. In the present work, the resolution of the BSE images used for roughness
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evaluation was around 1 μm/px, whereas the measuring step of the AFM was 0.3 μm. The processing of the BSE images yielded an average height and wavelength of the fibre profile equal to 1.85 μm and 31.5 μm, respectively. The value of the wavelength obtained with the AFM for the R2 fibres is within the same order of magnitude and equal to 21.5 μm. However, the value of the average height measured with the AFM was 0.23 μm, which is eight times lower than that estimated through the BSE image analysis. The sample preparation required for BSE imaging could also lead to an increase in the surface roughness of the steel fibre due to the cutting and polishing processes, and, thus, this technique may not represent the actual roughness profile of the steel fibre. A BSE image analysis of the surface roughness of fibres carried out images that have a similar resolution to the measuring step of direct techniques and with careful sample preparation would make it possible to simultaneously study the roughness of the fibre surface and the microstructure of the surrounding cement paste. Since the AFM and stylus profilometer represent the direct measuring techniques which resolution is in nanometres, this can provide more precise results than the BSE image analysis. Therefore, their outcomes were used to further analyse the properties of the fibre–matrix bond.

According to the values obtained for the $h_{av}$ of the polished and non-processed fibres measured using the AFM (presented in Table 3.1), the valleys of these fibres are not capable of fitting particles that are smaller than 0.02 μm and 0.23 μm, respectively. Considering that the minimum
size of cement clinker grains in ordinary or blended cement is around $1\ \mu m$, the roughness of the polished and non-processed fibres cannot improve the compaction of the cement grains near these fibres. In the case of the sanded fibres, the $h_{av}$ can fit grains that are smaller than $5.58\ \mu m$ and improves the packing of grains around the fibre. This improvement around the aggregates with a rough surface was previously reported in [104].

### 3.2.2 Wetting properties of the fibre surface

Based on the results obtained through contact angle goniometry, the dynamics of the water on the surface of fibres with different types of roughness were interpreted in terms of advancing and receding angles and their hysteresis. The hysteresis increased with an increase in fibre roughness, as demonstrated in Fig. 3.7. The existence of the irregularities on the fibre surface acts as a barrier for the water movement. The greater the roughness is, the more energy is required for the droplet to come over the irregularities of the fibre surface. Based on the measurement results, $\theta_a$ did not change with an increase in the roughness of the fibre surface. Therefore, the advancing front of the droplet possessed enough energy to pass through all the types of surface roughness considered. The roughness affected the $\theta_r$, which increased with an increase in roughness. The receding end of the droplet has the least amount of energy, which entails that it can be easily captured by surface irregularities, as also explained in [98]. As a result, the receding end of the droplet extends, as illustrated in Fig. 3.7, or creates the residual droplet that gets separated from the main droplet and gets stuck in the valleys of the fibre surface. Changes in $\theta_r$ were found to affect the hysteresis. The larger the hysteresis is, the larger the area of the fibre surface that is covered with water and the more uniform the distribution is (Fig. 3.7). The smaller the hysteresis is, the more uneven the distribution of water is; it forms drops and can easily slip from the fibre (Fig. 3.7). The precise values of the measured wetting parameters are reported in Table 3 in Publication III.

The received results indicated that the surface roughness of the fibre can affect the fluid dynamics along this fibre on the micro-scale. The fluid arrested by the fibres reported in [13] may lead to the local bleeding at the fibre surface if its roughness is very low because the liquid will gather in agglomerates, as observed in Publication III. On the contrary, high surface roughness can cause the water to be efficiently trapped on the fibre surface and minimise the local bleeding around this fibre. Regardless of the surface roughness, the w/c ratio around the fibres is usually larger than that observed in bulk cement paste [10]. However, the water is found to migrate from regions with a high w/c ratio to those with a lower w/c ratio due to capillary forces [90]. In the case where the surface roughness of the fibre is increased, the amount of liquid that can migrate through the
capillary channels is low and is distributed along the fibre surface better than when the surface roughness of the fibre is low. Therefore, large water agglomerations that are formed on the surface of polished fibres may not be completely migrated or utilised during hydration, leaving large voids that decrease the fibre–matrix bond.

3.3 Relationship between the properties of steel fibre and the surrounding cement paste

3.3.1 Distributions of UH cement phases and pores near the fibres with different surface roughness

The roughness and wetting properties of the fibre surface define the nucleation sites for cement hydrates, which can influence the microstructure of the cementitious matrix around the fibre. Therefore, the microstructure of the cement paste was investigated along the entire circumference of the fibre cross-section. The visual observation of the fibre–matrix cross-section revealed the existence of voids with an area larger than $0.02 \text{ mm}^2$ that decreased in size and number as the surface roughness of the fibre increased (Fig. 3.8). The development of these voids was governed by the agglomeration of the water along the fibre surface, which was explained in the Section 3.2.2.

The distributions of pores and the UH cement grains from the fibres with different types of surface roughness and with a step of $5 \mu \text{m}$ are illustrated in Fig. 3.9. The boundary zone can be indicated by a decrease in porosity and an increase in the UH cement grains with the distance from the fibre.
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Figure 3.8. BSE images of the cross-section of the fibres with different types of surface roughness embedded in the cement paste.
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surface, which is similar to the distributions observed with PP fibres in Section 3.1.2. An obvious decrease in the area fraction of the pores was detected with a decrease in the surface roughness of fibre (Fig. 3.9(a)). In the cases of the non-processed and the sanded fibres, the area fractions of the pores reached the values measured in the bulk cement paste (more than 2 mm from the fibre surface) at the distances of 90 μm and 45 μm from fibre surface, respectively. The area fraction of the pores and UH cement grains in the bulk cement paste was calculated based on 50 BSE images. In the case of the polished fibres, the area fraction of the pores was greater than that measured in the bulk cement paste. The effect of the roughness of the fibre surface on the distribution of UH cement grains was not detected through the reported measurements. However, the area fractions of the UH cement grains near the sanded fibres also matched the value measured in the bulk cement paste at a distance of 45 μm from fibre surface.

![Graph showing porosity and UH cement grains](image)

Figure 3.9. Average distributions of the pores and UH cement grains with the distance from the surface of the fibres with different types of roughness.

3.3.2 Mechanical performance and degradation of the fibre–matrix bond

The pull-out of steel fibres with different types of surface roughness from cement paste cylinders was performed under direct tension cycles with gradually increasing load to analyse the fibre–matrix bond degradation. Examples of the measured load-slip curves are presented in Figs. 3.10(a)–3.10(c). The obtained load-slip curves were analysed in terms of the residual slip increment ($s_{i}^{\text{res}}$) and stiffness of the bond during ascending ($k_{i}^{\text{asc}}$) and descending ($k_{i}^{\text{des}}$) parts of the load-slip curve per cycle, as schematically shown in Fig. 3.10(d). The part of the load-slip curve that represents $k_{i}^{\text{des}}$ has a concave shape, which is also typical for the response of concrete to a cyclic compression load [93]. The concave shape of the mentioned curve
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indicates the complexity of the elastic behaviour with the gradual release of the load, which complicates the exact quantification of $k_i^{\text{desi}}$. Therefore, the average value of $k_i^{\text{desi}}$ was calculated to determine the difference between it and $k_i^{\text{asci}}$ and characterise the changes in the stiffness of the bond. The mean evolution of the mentioned parameters for each type of surface roughness (six fibres were tested for each fibre type) is illustrated in Figs. 3.11(a)–3.11(c).

![Graphs of load-slip curves](image)

**Figure 3.10.** (a-c) Examples of the load-slip curves. (d) Schematic interpretation of the residual slip increment ($s_i^{\text{res}}$) and stiffness of bond during the ascending ($k_i^{\text{asci}}$) and the descending ($k_i^{\text{desi}}$) parts of the load-slip curve.

The deceleration, steady and acceleration stages of the $s_i^{\text{res}}$ development are shown in Fig. 3.11. The deceleration stage can be associated with the crushing of the cement matrix or the closure of pores that does not exceed the size of $9 \, \mu m$ according to the cumulative value of the slip by the end of the corresponding stage (Table 2 in *Publication IV*). The increase in the surface roughness of fibres prolongs the development of the deceleration stage, the transition from which to the steady stage was identified at 26%, 21%, and 16% of the maximum capacity of the fibre–matrix bond in the cases of R1, R2 and R3 fibres, respectively. At the steady stage, the growth of $s_i^{\text{res}}$ is the lowest of all stages. The steady stage can indicate the development of microcracks within the cement paste near the fibre. Since microcracks are discontinuous and can form at a distance from and at the

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Stiffness $u_{1000}$, [N/m]

0 1 2 3 4 5

Cycle number

2 6 10 14 18 22 26 30 34

(a) R1.

(b) R2.

(c) R3.

(d) Figure 3.11. (a-c) Mean development of $s_{res}^i$, $k_{asc}^i$ and $k_{des}^i$ per load cycle for each type of the surface roughness. Deceleration, steady and acceleration stages of the bond degradation are denoted by green, yellow and red colors, respectively.

(d) The area fraction of the cement paste left on the examined regions of fibre surface.

Fibre type

R1 R2 R3

Area fraction at measured locations, [%]

0 10 20 30 40 50 60 70 80 90

Despite the growth of the residual deformation of the fibre–matrix bond, the stiffness of the bond during the application and removal of the load was found to increase with each load cycle. The observed increase in $k_{asc}^i$ supported the compaction of the matrix with an increase in load, whereas
an increase in $k_{\text{des}}$ with each load cycle indicated a decrease in the elastic deformation that resulted in the degradation of the fibre–matrix bond. The values of $k_{\text{des}}$ and $k_{\text{asc}}$ did not coincide at any point of loading, which implies that, after the release of the load, the load-slip curve did not return to the starting point of the cycle. The largest difference between the $k_{\text{asc}}$ and $k_{\text{des}}$ was observed at the beginning of the loading during the closure of pores and the matrix compaction. The next increase in this difference was observed at the beginning of the acceleration stage when the debonding crack started developing with an increase in the residual slip increment. The generalisation of the discussed stages of bond degradation is illustrated in Publication IV in Fig. 11.

The pattern of the debonding crack can be complex and can pass along the fibre surface or through the cement matrix. The pattern of the debonding crack is governed by the strength of the fibre–matrix bond and can be analysed by studying the surface of the pulled out fibres. Based on Figs. 3.11(d) and 3.12, the amount of the cement paste found on the surface of the fibres grew with an increase in surface roughness. This can be due to the changes in the cement paste microstructure formed near the fibres with different types of surface roughness, which were detected in Publication III. The higher roughness of the fibre surface shifts the debonding of the fibre–matrix bond further into the matrix, where porosity is also lower as compared to the region at the fibre surface (Fig. 3.12). Therefore, greater roughness of the fibre surface improves the maximum capacity of the fibre–matrix bond.

![Figure 3.12](image-url)
4. Conclusions

The conducted experimental research examined and characterised the properties of fibres and the surrounding cementitious matrix and their effects on the mechanisms that affect the resistance and the degradation of the fibre–matrix bond. The main conclusions based on the obtained outcomes can be drawn in terms of the measured properties that can affect fibre–matrix interface, the effect of fibre roughness on the ITZ microstructure and the capacity of the fibre–matrix bond:

- The average height and wavelength of the surface roughness of the considered steel fibres indicated the size of the grains that can fit between the surface irregularities, which can affect the packing of the cement grains near the fibre surface. The distributions of the CH and C-S-H phases revealed the main changes in the cement paste composition within a distance of 20 \( \mu m \) from the surface of the steel fibre. The largest amount of CH phases was observed within 15 \( \mu m \) from the fibre surface, with the maximum precipitation at a distance of 8 \( \mu m \). However, the amount of the C-S-H phases was found to increase with the distance from the fibre surface and exceeded that of CH at all measured distances. Therefore, having a greater proportion of C-S-H than CH phases can enhance the fibre–matrix bond. The volumetric distributions of the evaluated pores and UH grains helped identify the existence of a boundary zone around PP fibres with a thickness of around 70 \( \mu m \). The volume of the pores detected in the boundary zone was larger than that in the bulk cement paste, which suggests more pronounced interconnectivity between the pores that can decrease the durability of the FRCC.

- An increase in the surface roughness of steel fibres decreased the mobility of the water along the fibre surface, facilitating its uniform distribution and effective utilisation during hydration, which coincides with the reduced formation of large voids at the fibre–matrix interface. The increase in wettability with the roughness of the fibre surface was clearly found to decrease the porosity at the boundary zone between the steel fibre
and the cementitious matrix, indicating the thinning of the boundary zone. The porosity near the sanded and non-processed fibres decreased to the values of the bulk porosity at the distances of 40 \( \mu m \) and 90 \( \mu m \) from the fibre surface, respectively, whereas the porosity around the polished fibres did not reach the values of the bulk porosity within the measured distance from the fibre surface. The connection between the surface roughness of the fibres and the changes in the distribution of the UH cement grains were not obvious based on the measured results.

- The resistance of the fibre–matrix bond that was measured under tension cycles with gradually increasing load demonstrated the development of the residual slip from the beginning of loading, indicating the absence of a complete bond between the steel fibre and the cement paste. The residual slip increment evolved in three stages – deceleration, steady development, and acceleration – which extended up to 30\%, 70\% and 100\% of the maximum capacity of the fibre–matrix bond, respectively. An increase in the surface roughness of the steel fibres led to an increase in the maximum capacity of the fibre–matrix bond and the number of cycles during the steady development and deceleration stages of the bond degradation. The stiffness of the bond continued to grow until the last loading cycles despite an increase in the residual slip. The observations of the fibre surface after the pull-out test using SEM indicate that the debonding crack complexity increased with an increase in the surface roughness of the steel fibres, which can also explain the growth of the maximum capacity of the fibre-matrix bond.

The results indicate that the effect of the surface roughness of steel fibres changes the interfacial porosity, which affects the capacity of the fibre–matrix bond. Compared to other fibre modification techniques, the coarsening of the surface of steel fibres through sanding is a cheap and easily implementable method for modifying fibres which can significantly improve the strength and durability of the FRCC.

To obtain the reported results the novel methodologies that were introduced in this research are highlighted below:

- Phase-contrast micro-computed tomography with a synchrotron x-ray radiation source was successfully applied to study the boundary zone around the PP fibres. The studied volume of 2.22 mm\(^3\) enabled the quantification of the volumetric distributions of the pores and UH grains up to a distance of 140 \( \mu m \) from the fibre surface and allowed access to the interconnectivity of the pores and the reduction of the cement grains volumes within 28 days of hydration.

- The wettability of the steel fibres with different types of surface rough-
Conclusions

Wettability was evaluated using contact angle goniometry by measuring the advancing and receding contact angles, which define the mobility of the water along the surface of these fibres. In the literature, only static contact angles that vary between advancing and receding angles have been reported.

- The k-means clustering algorithm was introduced for the segmentation of the cement paste phases, such as pores and UH cement grains, in the BSE images. In contrast to the global threshold segmentation method that is usually used for the BSE images of a cementitious matrix, the k-means clustering algorithm segments the BSE image by clustering pixels in terms of the minimum Euclidean distance between the pixels, which eliminates the issue associated with the definition of an obscure threshold boundary.

- The pull-out of the steel fibre from the cementitious matrix was performed under the tension cycles at a low speed and with a small increase in load amplitude during each cycle to study the degradation of the fibre–matrix bond, which cannot be accessed through the conventional pull-out test under monotonic loading. The development of residual slip increment and the stiffness of the bond during the application and removal of the load in each load cycle were analysed.

The outcomes demonstrate how a comprehensive analysis of the wettability and roughness properties of the fibre together with the microstructural changes in the surrounding cement paste can explain the response of the fibre–matrix bond from the beginning of loading until the maximum capacity of the bond is reached. The reported approach of examining the wettability of fibres along with the developed microstructure of the fibre–matrix bond and its performance under loading cycles can be a robust approach for the modification of FRCC in the future.
References


References


