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Review

# Wool-Reinforced Cement Based Composites

**Daria Józwiak´-Nied´zwiedzka [1] and Alessandro P. Fantilli [2,*]**

1 Department of Experimental Mechanics, Institute of Fundamental Technological Research, Polish Academy
of Sciences, Pawinskiego´ 5B, 02-106 Warsaw, Poland

2 Department of Structural Engineering, Construction and Soil Mechanics, Politecnico di Torino, Corso Duca Degli Abruzzi 24, 10129 Torino, Italy

**Abstract:** In this paper, an overview of the latest research activities in the field of cement-based composites incorporating sheep wool reinforcement is presented. First, the characteristics of this type of natural fibre are described. Then, the current use of sheep wool fibres in cement-based composites is discussed. The research problems regarding the properties of cement matrix composites reinforced with sheep wool are divided into four groups: thermal and acoustic properties, mechanical behavior, durability issues, and microstructure aspects. The latter two groups are analysed separately, because both durability and microstructure are of particular importance for future applications of wool reinforcement. Finally, the main directions of future researches are presented.

**Keywords:** natural fibres; sheep wool fibres; mechanical properties; durability; microstructure

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## 1. Introduction

The general purpose of fibres application in cement-based composites is to increase material toughness by improving the resistance to crack propagation. The reinforcement also increases the composite material tensile strength, especially when a large quantity of fibres is added to the cementitious matrix [1]. Various kinds of dispersed reinforcement in the form of thin fibres are used in concrete structures. Depending on the type of material, the fibres can be distinguished as metallic and non-metallic: glass, basalt, natural organic, and polymeric (like polyethylene, nylon, polyester, Kevlar, PVA-poly(vinyl alcohol).

The use of both metallic and non-metallic fibres to improve concrete behaviour in tension is not new. However, in recent years there has been growing interest in utilizing natural fibres (from plants and animals) to produce eco-friendly construction materials. The relatively high cost of industrial fibres and the aim of reducing the negative environmental impact of the construction industry make the use of natural fibres more common [2]. The availability of natural fibres is directly related to the climatic zones. This is particularly true for plant fibres, like jute [3], coir [4], sisal [5,6], bamboo [7], wood [8], palm leaf [9], coconut leaf [10] and fibres [11], cotton [12] and hemp [13], or cellulose [14]. Plant or cellulose fibres have many advantages, such as wide availability at a relatively low cost, biological renewal, recyclability, biodegradability, harmless nature, and zero carbon footprint [15]. However, the same properties can also be attributed to animal origin fibres, but with better mechanical properties, especially in the case of wool [16].

Several studies on the use of sheep's wool in the construction industry are related to the applications as an insulating material, both thermal and acoustic [17]. Indeed, sheep wool is comparable to other insulation materials, such as mineral wool and calcium silicate [18]. The results of some experimental measurements show that sheep wool is competitive in terms of thermal which made it capable to absorb moisture, prevent condensation and regulate humidity in insulation materials. Due to the high content of water and nitrogen, wool is also a naturally flame retardant [26]. Sheep's wool is also an excellent acoustic material but, according to Zach et al. [27], no additional acoustic benefits are achieved with material thicknesses greater than 170 mm.

An undoubted advantage of sheep wool is the influence with human health. Unlike fibre glass, wool can be installed without protective clothing, because it does not cause irritation to the skin, eyes or respiratory tract [20]. The research conducted by Liang and Ho [28] revealed that the toxicity of combusted insulating materials, such as rock wool and fibreglass, is significantly higher than that of organic materials. Wool can also absorb unhealthy carbons in the atmosphere, helping to provide a cleaner environment [26].

Despite all the above-mentioned benefits, the large-scale production of cement-based composites reinforced with sheep wool fibres is currently limited by the long-term durability [29]. However, in all the previous researches, including the recent review papers by Parlato and Porto [30] and Allafi et al. [31], there is no information on the durability of cement-based composites with the addition of wool fibres.

The durability issue is associated with an influence of the pH of cement matrix on the sheep wool fibres. Research conducted by Fantilli and Józwiak´-Nied´zwiedzka [29] showed direct e ect of cement alkalinity and curing conditions on durability of wool fibres in cement-based mortars. The capability of the sheep wool fibres to bridge the crack surfaces, and to guarantee the presence of a residual tensile strength in the post-cracking stage is remarkably reduced in high alkali cement and in high humidity conditions. This phenomenon is particularly highlighted by the values of the residual strength and the fracture toughness in bending of wool-reinforced cement-based mortars.

To mitigate the degradation of wool fibres in cement-based composites, two main methods have been adopted: fibre pre-treatment or cement matrix modification. Attempts have been made to modify the surface of the fibres to improve their mechanical properties and durability [30,32,33], and to reduce concrete's alkalinity by partially replacing the cement with supplementary cementitious materials [34], or use blended cement [35] and/or low-alkali cement [29].

Reducing the clinker content in the cementitious matrix through the use of supplementary cementitious materials improves the durability of natural fibres by increasing cement hydration, and reducing the alkalinity of pore solutions and Portlandite consumption [36]. The modification of cement hydration may require less e ort and involve less cost compared to pre-treatment of natural fibre. In addition, the possible influence of the modifying agent on the cement matrix should also be considered [36].

All the above-mentioned researches concerning sheep wool fibres application in the cement-based composites are reviewed and presented in this paper. Specifically, the main material properties, the applications in building materials, and the behaviour of cement-based matrix composites reinforced with sheep wool fibres are described.

## 2. Characteristics of Sheep Wool Fibres

Wool is the natural protein fibre deriving from the fleece of sheep. It has one of the most complex structure among textile fibres (Figure 1), as a single wool fibre consists of a cortex and a surrounding cuticle layer.

Each of the two components is formed of various other morphological components. The cortex contains cortical cells and the cell membrane complex [26]. It has a bilateral structure and is responsible for the mechanical behaviour. Cortex is the carrier of the characteristics of wool properties such as elasticity, ductility, and swelling force [21].

Wool fibres have a particular surface structure of overlapping scales called cuticle cells, which anchor a fibre in the sheep's skin. The surface of wool fibres (Figure 2a) is very di erent with respect to typical man-made fibres, which have a very smooth surface (Figure 2b) [2].

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More information about mechanical properties of cement-based composites reinforced with sheep wool are presented in Section 4.2.

Dénes et al. [43] stated that wool can be used as carbon fibre precursor. Preliminary research showed that wool fibres can replace the synthetic polymer in the sight of carbon fibre production. Hassan et al. [46] found that carbon fibres were able to be produced through the carbonization of untreated and crosslinked wool fibre. The carbon yield of the resulting fibres was found to be a function of the type of crosslinking agents applied to wool. In addition, due to the importance of using locally available materials for rural building renovation as well as for restoration and repair of historic and cultural heritage buildings, the use of sheep wool is strongly suggested [18]. In fact, wool reinforced composites are suitable for the renovation of traditional buildings due to the comparable composition of the mixture with the original mortars [39].

## 4. Properties of Cement-Based Composites Reinforced with Sheep Wool

### 4.1. Thermal and Acoustic Properties

Sheep wool is regarded as one of the most performative insulating natural materials due to its thermo-hygrometric and acoustic properties [22,30,43,47,48]. One of the most important factors concerning the thermal insulation is thermal conductivity of material. To be considered as an insulation material, thermal conductivity should be less than 0.065 W/mK, [43]. As this value varies between 0.033 and 0.063 W/mK in the case of wool [21,23,43], it can be considered as a good insulation material. The research conducted by Korjenic et al. [ 18] showed that sheep wool, compared with mineral wool and calcium silicate, provides comparable thermal insulation characteristics, and in some applications even reveals better performance. Comparing the properties of sheep, flax and glass wool, Tuzcu [21] found that was 0.033, 0.040, and 0.034 w/mK, whereas specific heat capacity c was 1720, 1550, and 799 J/kgK, respectively. However, the thermal conductivity varies depending on the humidity conditions: increases with the content of water in the sheep wool or with the increment of the apparent density [22]. Volf et al. [23] investigated the treated sheep wool and raw sheep wool as natural insulating materials. They revealed that both types of wool had the lowest value of volumetric heat capacity $c_p$ (0.05 and 0.06 Jm $^{3}$K $^{1}$, respectively) and the highest value of thermal conductivity (0.063 and 0.062 Wm $^{1}$K $^{1}$) compared to mineral wool ($c_p$ = 0.09 Jm $^{3}$K $^{1}$ and = 0.039 Wm $^{1}$K $^{1}$) as well as to flax, hemp, and wood fibres. They concluded that natural insulations had comparable thermal properties to common building insulation materials and could bring advantages in thermal and moisture bu ering.

Some researchers showed that, with the addition of sheep wool, density and thermal insulation improve, but, at the same time, the mechanical properties of the composite decrease [39]. Fiore et al. [24] investigated the mechanical behavior and thermal conductivity of a cement mortar with various length and di erent contents (i.e., 13%, 23%, and 46% by wt. of cement) of wool fibres. They revealed that the application of wool fibres improved the thermal insulation in the analysed cement-based composites.

Sheep wool shows good acoustical performances by absorbing and reducing noise [18]. According to Asdrubali [25] panels made from sheep wool were characterized by an absorption coe cient of about 0.84 at 2000 Hz, slightly lower than rock wool (0.91) or polyester (0.95), but significantly higher than cellulose (0.53) or hemp fibres (0.52). A sheep wool panel of 20 mm thickness had also a very low index of impact noise reduction $DL_w$ (18 dB), much smaller than glass wool (31 dB) or expanded polystyrene (30 dB), even lower than wood wool (21 dB) or cellulose (22 dB).

Wool fibres are more hygroscopic than any other fibres. As a result, when moisture content increases, the thermal conductivity coe cient does not change significantly [48].

### 4.2. Mechanical Behavior

The use of natural fibres as a reinforcement of cement-based composites can increase the toughness of concretes and mortars, and represents a sustainable option to the traditional industrial fibres as well. Indeed, such fibres can bridge the surfaces of the cracks in the post-cracking stages and reduce the environmental impact of the construction industry [32].

Porubská et al. [49] investigated the gamma radiation up to 400 kGy on the mechanical properties of sheep wool. They found that the tensile strength at failure did not change significantly while the original elongation firstly increased and, then, a monotonous reduction was observed. Grădinaru et al. [50] examined the influence of sheep wool fibres and fly ash on the compressive and tensile strength of concrete. They tested seven types of mixtures, with and without the addition of fly ash, and of two percentages (i.e., 0.35% and 0.80% in weight) of wool fibres (with a length comprised by 25 mm and 55 mm). The experimental results showed that sheep wool fibres did not improve the strength of the concrete at the studied percentages of addition and, in most of the cases, a lower strength was measured. It was observed that when sheep wool fibres are used, compressive strength reduced of 15–30%, compared to the reference concrete, although the degree of reduction depended on the fibre length and dosage. A fibre length of 55 5 mm and a dosage of 0.35% had an insignificant influence on the compressive strength, but a higher dosage or a smaller length of the fibre decreased the value of the compressive strength. Fiore et al. [24] investigated the mechanical behaviour of a cement mortar with various length and di erent contents (i.e., 13%, 23%, and 46% by wt. of cement) of wool fibres. They revealed that wool-reinforced composites showed lower compressive strength than the reference no wool cement composite, regardless of the content and length of fibres. Similar conclusions were presented by Cardinale et al. [51]. In their research, the addition of sheep wool fibres was much smaller, 2%, 5%, and 7% per dry raw materials mass. They investigated flexural and compressive strength of mortars made with CEM II/A RCK 42.5 N, crushed sand of 0.63 mm, lime and water. As a result, a reduction of flexural and compressive strength of 9.1% and 14.7% was respectively observed for mortar with 2% of wool fibres. In mortars with a higher content of wool, the decrease of strength was much greater (more than 80%).

Opposite results were obtained by Fantilli et al. [32]. They analysed the influence of sheep wool fibres on the mechanical properties of cementitious mortars. Additionally, mortars reinforced with hemp were also tested. The authors stated that the flexural strength and the ductility increased when wool is added to cementitious mortars. Similar to other natural fibres, wool improved the mechanical and ecological performances of the mortars. Pederneiras et al. came to the same conclusions [52]: the use of wool fibres in cement mortars improved the flexural strength. A higher increase in flexural strength was observed for longer fibres (30 mm) in comparison to shorter fibres (15 mm). The cement-based mortar made with CEM II/B-L 32.5 N and 20% of 30 mm long wool fibres revealed an increase of 40% and 26% in flexural and compressive strength, respectively. Alyousef et al. [45] revealed that sheep wool fibres (up to 1.5% of 70 mm length fibres) can reduce the compressive strength of concrete, but undoubtedly improve the tensile and flexural strength, and concrete ductility (with higher energy absorption capacity) as well.

Sheep wool fibres as reinforcement in lime based composite materials were investigated by Tăma¸s-Gavrea et al. [53]. They analysed a mortar containing hydrated lime, rice paste and sheep wool fibres, and stated that this composite was characterize by acceptable adhesive strength (equal to 0.125 N/mm$^2$).

Wool, kenaf, and wheat straw used as fibres, and clay used as a binder, were analysed in some studies conducted by Erkmen et al. [54]. Among the results, insulating materials containing 7% of wool fibres revealed the best result concerning compressive strength (4.9 MPa), thermal conductivity coe cient (0.061 W/mK), and water absorption (% 0.0015/h) in comparison to the other commercial products.

4.3. Durability and Microstructure

The durability of wool reinforced cement-based composites depends on the conditions of exploitation and on external actions. The bonding between fibres and the cementitious matrix is a decisive element. The latter depends on the quality and processes that appear in the fibre/matrix interface. It has been shown that in glass fibre-reinforced cement-based composites, the chemical interaction between these two constituents may be destructive for the composite integrity [1]. The usefulness of natural fibres in cement-based materials is limited by their high potential to degrade in alkaline environment. Frequently, they loss the strength when used as reinforcement of a cementitious matrix exposed to aggressive environmental conditions [29,35,36,55].

As it was expected, the addition of sheep wool fibres significantly influences the workability of fresh mixes. Cardinale et al. [51] tailored some cement-based mortars with a constant water/cement ratio (equal to 0.4) and various content of sheep wool fibres. They found deteriorated mortars due to the insertion of ever-increasing percentage of wool fibres, and the necessity of increasing the programmed quantity of water, in order to ensure the workability of the mixture. Similarly, Alyousef et al. [45] investigated the properties of fresh concrete (made with Ordinary Portland cement, natural aggregate, w/c = 0.5, and up to 6% of sheep wool fibres by weight of cement), and stated that addition of sheep wool fibres caused a huge demand of water for making the concrete workable. The reason of such low workability has to be ascribed to the high specific surface area and fineness of wool fibres. Thus, workability of concrete containing sheep wool fibres decreases with the increasing content of wool. If for reference concrete the slump value is 30 mm, for the same concrete with 2% of sheep wool fibres it decreased to 8 mm. Even the pretreatment of fibre with salty water, used to increase the surface friction, did not improve fresh concrete properties [33]. The slump value was 55 mm for reference concrete without fibres, and 22 mm for 2% of both unmodified and modified sheep wool fibers. Obviously, this negative phenomenon could be minimized by the addition of chemical admixtures.

The degradation mechanisms of natural fibre in the alkaline and mineral-rich environment, which is typical of the cement-based matrixes, was investigated by Wei and Meyer [36]. They studied the degradation mechanisms and found that, by reducing alkalinity of pore solution, metakaolin e ectively mitigates the deterioration of natural fibre. Also, the alkali degradation process of natural fibre was proposed. Cement hydration was presented to be a crucial factor in understanding fibre degradation behaviour, which is confirmed by the test results conducted by Fantilli and Józwiak´-Nied´zwiedzka [29]. They analysed the influence of the alkalinity of Portland cement type I and curing conditions on the mechanical properties and microstructure of sheep wool reinforced mortars. The results revealed that the lower the alkalinity of the cement paste, the better the resistance of wool fibres in cementitious matrix, which increased the residual stress after cracking in wool reinforced mortars. The curing of mortar beams in water at room temperature significantly accelerated the process of wool fibre degradation in matrix made with high-alkali cement ($Na_2O_{eq}$ = 1.1%), compared to those obtained with normal- and low-alkali cement ($Na_2O_{eq}$ = 0.6 and 0.4%, respectively). In Figure 5, the microstructure of the specimens made with high-alkali cement and wool fibres, after 3 and 27 days of curing in water at 20 C, are presented.

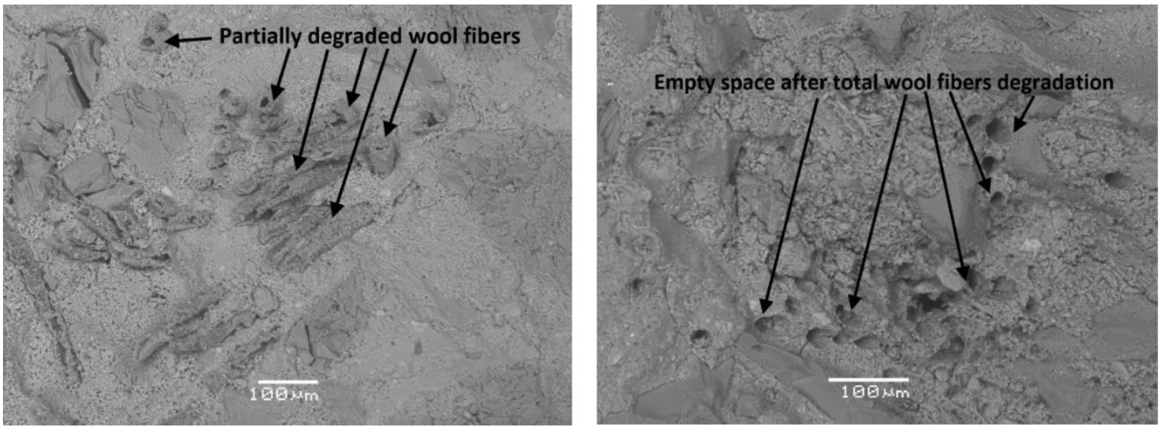
It can be observed that, with high-alkali cement, the longer the time of curing in water, the higher the degree of wool fibres degradation.

Research conducted by Fantilli et al. [35] on the compatibility between wool and polypropylene fibres and cement-based matrix (made with CEM II/B-LL 32.5 R) showed the influence of curing condition on durability of fibres. The beneficial e ect of wool was not observed when the specimens were stored in water at 20 C for 27 days (see Figure 6). Nevertheless, wool filaments were able to resist more than three days in the alkaline environment before their complete dissolution.

Thus, they can be used to contrast the e ects of plastic shrinkage, as the industrial polypropylene fibres do.

The method of pre-treatment of sheep wool fibres are not always e ective. Alyousef et al. [33] used saltwater treatment modification of sheep wool which caused an improvement of the fibre's mechanical properties and improved adhesion with cement paste. Also, atmospheric plasma was used to modify the nano-metric properties of the fibre surface [32,56]. Ceria et al. [56] analysed the influence of the atmospheric plasma jet treatment on physical and mechanical properties of wool fabrics. Their researches revealed the increment of tensile strength (up to +13%) and elongation at effectively mitigates the deterioration of natural fibre. Also, the alkali degradation process of natural fibre was proposed. Cement hydration was presented to be a crucial factor in understanding fibre degradation behaviour, which is confirmed by the test results conducted by Fantilli and Jóźwiak-Niedźwiedzka [29]. They analysed the influence of the alkalinity of Portland cement type I and curing conditionsMaterials**2020**,on13, the3590mechanical properties and microstructure of sheep wool reinforced mortars.9 Theof13 results revealed that the lower the alkalinity of the cement paste, the better the resistance of wool fibres in cementitious matrix, which increased the residual stress after cracking in wool reinforced

break (up to +19%) by increasing the intensity of the plasma treatment. Hence, Fantilli et al. [32] mortars. The curing of mortar beams in water at room temperature significantly accelerated the treated sheep wool fibres with atmospheric plasma in order to modify the nanometric properties of process of wool fibre degradation in matrix made with high-alkali cement (Na2Oeq = 1.1%), compared their surface. However, a significant e ect of treated wool fibre surface modification on the mechanical to those obtained with normal- and low- alkali cement (Na2Oeq = 0.6 and 0.4%, respectively). In Figure properties of cement-based mortars was not observed. Conversely, they found that, both the flexural 5, the microstructure of the specimens made with high- alkali cement and wool fibres, after 3 and 27 strength and the ductility increased when wool, treated or not, was added to cementitious mortars. days of curing in water at 20 °C, are presented.

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(**a**) (**b**)

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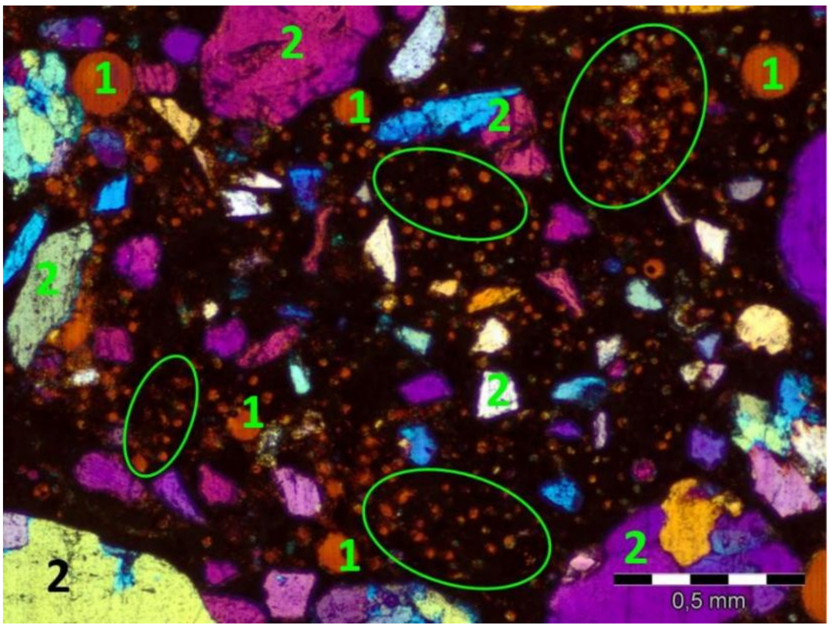
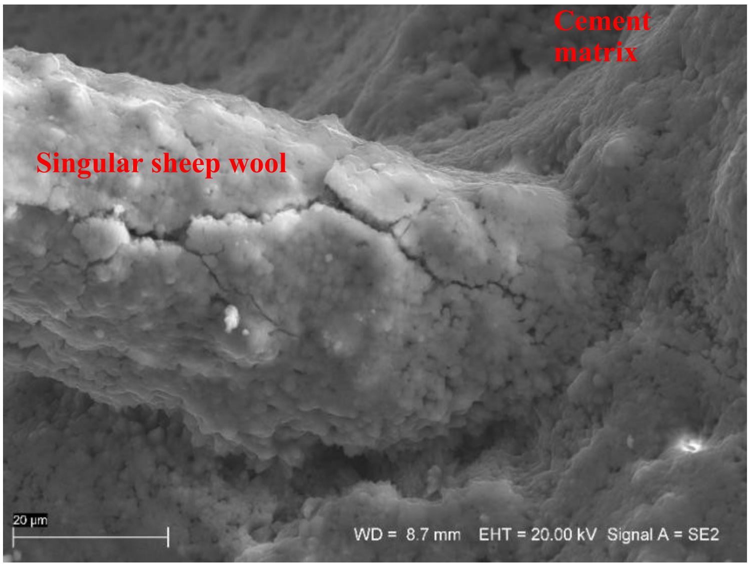


debonding and matrix micro-cracking were dominant at the interfaces. However, the evidence of a porous transition zone or massive concentration of calcium hydroxide at the interface was not found. was not found. For 250 days of curing, a high porosity was not detected in the interfacial area and For 250 days of curing, a high porosity was not detected in the interfacial area and just one EDS spot just one EDS spot indicated the presence of calcium hydroxide close to the fibres. The above indicated the presence of calcium hydroxide close to the fibres. The above conclusion is consistent conclusion is consistent with observation regarding the interface between sheep wool fibre and with observation regarding the interface between sheep wool fibre and cement matrix (see Figure 7). cement matrix (see Figure 7).



Nevertheless, future researches, aimed at improving the performance of sheep wool fibre reinforced cement-based composites, are needed. A special way to prepare the homogenous sheep wool fibres and modification of their surface will also be considered. Hybrid fibre reinforcement will be used to improve the mechanical properties and durability of the cement matrix composites. Investigations will be undertaken to increase the proportion of sheep wool fibres in cement-based composites.

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