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A Practical Valorization Approach for Mitigating Textile Fibrous Microplastics in the Environment: Collection of Textile-Processing Waste Microfibers and Direct Reuse in Green Thermal-Insulating and Mechanical-Performing Composite Construction Materials

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Abstract: Microplastic (MP) contamination is an urgent environmental issue to address. Fibrous microplastics (FMPs) are the principal MP type in the air and have already been found in human stool and lung tissues. FMPs are generated from the lifecycle of synthetic and blended textiles and are expected to increase due to fast fashion. Among textile processes, the finishing of fabrics is estimated to generate 5000 t/year of textile waste fibers in Italy, including FMPs. To limit FMPs spread, this paper suggests, for the first time, the direct collection of blended finishing textile waste microfibers and reuse in designing thermal-insulating and mechanical-performing fiber-reinforced cementitious composites (FRCs). The microfibers were thoroughly characterized (size, morphology, composition, and density), and their use in FRCs was additionally evaluated by considering water absorption and release capacity. Untreated, water-saturated, and NaOH-treated microfibers were considered in FRCs up to 4 wt%. Up to a +320% maximum bending load, +715% toughness, −80% linear shrinkage, and double-insulating power of Portland cement were observed by increasing microfiber contents. NaOH-treated and water-saturated microfibers better enhanced toughness and linear shrinkage reduction. Therefore, green and performant composite construction materials were obtained, allowing for the mitigation of more than 4 kg FMPs per ton of cement paste. This is a great result considering the FMP contamination (i.e., 2–8 kg/day fallout in Paris), and that FRCs are promising and shortly-widely used construction materials.

Keywords: fibrous microplastics; microfibers; textile; waste; construction materials; recycling; mitigation; thermal conductivity; three-point bending; fiber-reinforced cementitious composites

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1. Introduction

In the last few years, microplastic (MP) pollution has become an environmental issue of global concern. MPs are plastic debris with sizes between 0.3 µm and 5 mm [1]. They are composed of synthetic polymers such as polyethylene (PE), polystyrene (PS), polyethylene terephthalate (PET), polyester (PL), nylon (PA6), etc. MPs can be found in several shapes, such as beads, foams, films, fragments, and fibers. Like other chemical pollutants, MPs are already distributed among the four major environmental compartments: air, water, soil, and biota. Such compartments not only behave as MP reservoirs but grant the transport of MPs within them [2], allowing MPs to behave similarly to global biogeochemical cycles and spiral around the globe with different atmospheric, oceanic, cryospheric, and terrestrial residence times [3].

Several removal technologies based on physical, chemical, and biological processes have been proposed to restore the environment from MP pollution. Since the presence of

MPs was first reported in water in 2004 [4], a broad spectrum of MP removal technologies from the aqueous medium, such as adsorption, sedimentation, ultrafiltration, photocatalysis, magnetic removal, microbubbles, and activated sludges, has been investigated [5–11]. When MPs were found in the terrestrial environment, biodegradation became the primary investigated strategy for soil remediation [12].

The presence of MPs in atmospheric fallout was reported in 2015, and fibrous microplastics (FMPs) represented the primary type of MP present in the air [13,14]. According to the European Chemical Agency (ECHA), fibrous microplastics are defined as MPs with lengths between 0.3 μm and 15 mm, and a length-to-diameter ratio of greater than three [1]. FMPs released into the atmosphere during the lifetime of synthetic and blended textiles (production, use, and disposal) significantly contribute to the overall issue of MP pollution. For instance, airborne FMPs can reach terrestrial and aquatic environments through wind transport, deposition on the surface of cities or agrosystems, and runoff [13,14]. In 2017, the International Union for Conservation of Nature (IUCN) estimated that FMPs released from textiles during industrial and household laundry accounted for 35% of the annual MP emission sources into the oceans [15]. Once entered into aquatic and terrestrial environments, FMPs impact their physicochemical characteristics [16] and interact in several ways with the biota, inducing health issues such as neurotoxicity, oxidative stress, oxidative damage, and even death [2,16,17]. Additionally, the transfer of FMPs from the atmosphere to aquatic and terrestrial environments represents a probable pathway to humans [17], and FMPs have already been found in human stool [18] and human lung tissues [19]. For this reason, developing innovative strategies to decrease the quantities of FMPs released into the atmosphere is a key aspect of the overall framework for reducing MP pollution.

The emissions of textile-derived FMPs are expected to increase due to the growing demand for synthetic or blended textiles and fibers [20]. This phenomenon is mainly attributable to fast fashion, which promotes mass consumption and a linear economic model. As a result, a large amount of textile waste is generated both from the industry (special waste) and consumers (urban waste). The former includes unsold clothes and industrial process waste, whereas the latter old-fashioned or end-of-life products. In Italy, textile waste production is significant and amounts to 490,000 tons/year of special waste and 146,000 tons/year of urban waste [21].

A portion of textile waste is currently recycled. Clothes waste, both from pre- and post-consumer use, is reused as clothing in the second-hand market or donated to charity. Fabrics and scraps are sorted according to color and material and recycled to create new yarns and fabrics, thermal-insulating panels, and upholstery for home and transport furniture. However, waste fibers from processing and other non-recyclable/reusable textile wastes are currently delivered to landfills.

Among these, waste microfibers from the raising and shearing of fabrics display a high risk of diffusion into the air and soil because of their volatility. Raising and shearing are mechanical finishing processes used to create heat-insulating and aesthetic fabrics. First, fibers are pulled out from the fabric to form a heat-insulating fluff; then, the fluff is cut at a defined and adjustable height for aesthetic purposes. During this finishing process, a suspension of waste microfibers is generated in the air, which is then collected by air-filtering systems, compacted, and delivered to landfills. In Italy, 5000 tons/year of these textile-waste-processing microfibers is currently disposed to landfills. This involves discharge costs for the producer and the risk of spreading textile waste from landfills into the environment. Particularly, the synthetic ones can be considered FMPs due to their size and composition, and can worsen the FMP issue in the environment.

To solve this environmental issue, this paper proposes the reuse of blended textile waste microfibers from the raising and shearing of fabrics, addressing the need to implement new strategies against the release of microfibers, particularly FMPs, into the environment. The decision to consider blended textile waste microfibers and not just synthetics ones arise from the desire to propose a mitigation solution for FMPs that is close to reality, thus proposing a practical solution to a real issue. For the first time, we propose the

reuse of FMPs as a mechanical reinforcement for fiber-reinforced cementitious composites (FRCs), which are innovative composite construction materials. In FRCs, fibers increase the ductility of the cement matrix by reducing cracking during shrinkage and loading, thus increasing the material durability compared with solely cement. Steel, carbon, glass, and polypropylene fibers are usually employed [22,23] for these purposes; however, using textile waste microfibers would additionally allow for green FRCs with better thermal insulation properties. Furthermore, the proposed solution aims to reduce the amount of textile waste disposed to landfills and waste management costs for the textile industry. Apart from the outlined benefits, recycling this kind of microfiber allows to directly collect FMPs before reaching and spreading into the atmosphere and to control their properties (i.e., size, composition, etc.).

In this study, blended textile waste microfibers from the raising and shearing of fabrics were chemically and physically characterized. Their use in FRCs was preceded by water absorption, water release, and real density measurements to enable proper mix design (water, cement, and fiber content). Untreated, water-saturated, and mercerized textile waste microfibers were added to the cementitious mix up to 4 wt% (40 vol%). Finally, the influence of fiber surface treatment and content on the FRC was detected in terms of linear shrinkage, three-point bending test, and thermal conductivity. It was found that at least 4 kg of FMPs per ton of cement mix could be used in creating green FRCs with increased thermal-insulating properties, mechanical strength, toughness, and reduced shrinkage compared with the reference Portland cement. Considering that 3–10 t of microfibers are deposited by atmospheric fallout every year in Paris, 29% of which constituted by FMPs (i.e., 2.38–7.95 kg of FMPs/day) [2,14], the present approach significantly allows the same amount of FMPs falling in Paris per day to be recovered before reaching and polluting the environment.

2. Materials and Methods

2.1. Textile Waste Microfibers

The textile waste microfibers involved in this study were generated by a textile finishing plant located in Carpi (Modena, Italy). The company performes finishing to impart thermal insulation and aesthetic properties to the surface of different fabrics, including cotton, cotton blends, and synthetics. The process includes raising and shearing phases for heat-insulating and aesthetic purposes. The involved textile microfibers were generated during the shearing phase and collected by air-pumping and air-filtering systems (Figure 1).

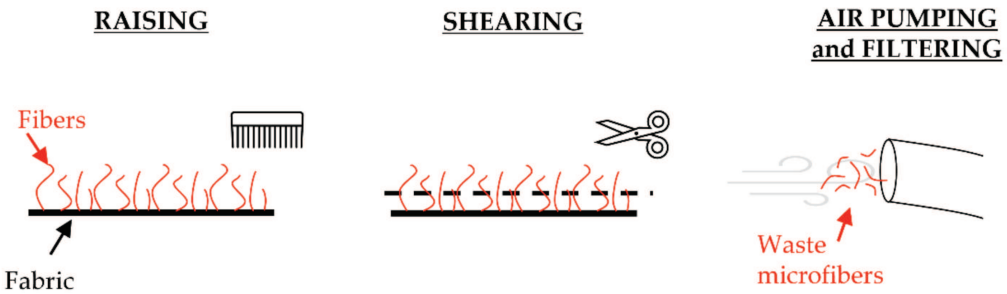

Figure 1. Generation of textile waste microfibers from the finishing of fabric for heat-insulating and aesthetic properties.

The type and coloring of fabrics change periodically according to market demand and imply a heterogeneity (in composition, diameter, and color) of the textile waste microfibers produced. In addition, the length of the waste microfibers differs due to the raising and shearing phases shown in Figure 1. Six samples were collected between the months of November and December to comprise a representative batch of blended microfibers. From the company's data on fabrics subjected to finishing processes in that period, it was found that the surveyed microfibers were composed of 61% pure cotton, 29% cotton blend (cotton and PA), and 10% synthetics (PA, PC, PL, and NY).

The different composition of textile waste microfibers (natural or synthetic) was detected by ATR-FTIR spectroscopy (FTIR VERTEX 70, Bruker Optics, Ettlingen, Germany). The spectra were acquired after 32 scans between a 600–4000 cm^{-1} transmittance range and with a 4 cm^{-1} resolution. Additionally, OPUS software (OPUS 6.5, Bruker Optics GmbH, Selb, Germany) was involved in the spectrum analysis. The data composition was then compared and confirmed with that from the manufacturer's waste datasheet.

An environmental scanning electron microscope in high vacuum mode (ESEM, ESEM-Quanta 200 Fei Company, Oxford Instruments, Abingdon, UK) was involved in distinguishing FMPs from natural fibers based on their different morphologies.

In addition, the fibers were characterized by optical microscopy mainly to assess the average size of the synthetic ones to determine whether they could be defined as FMPs. A Leica EZ4D (Leica Microsystems, Wetzlar, Germany) stereo microscope was used at different magnifications, and the average size of synthetic microfibers was computed through ImageJ software (1.53k Version), with over 30 measurements for length and 50 for diameter. The Set measurements function of ImageJ (Analyze section) was used to set the measuring reference scale based on the known reference bar length of the optical images. Then the Measurements function (Analyze section) was applied to measure the length and diameter of the synthetic fibers with respect to the set reference scale.

The real density of the textile waste microfibers was achieved through a Micromeritics Accupyc 1330 helium pycnometer by Micromeritics Instruments (Norcross, GA, USA). This value, as well as that for the water absorption and release over time by microfibers, was needed to pursue a subsequent mix design of FRCs.

The water absorption (WA) of microfibers was computed after 24 h of immersion in water at laboratory conditions (25 ± 2 °C, 60 ± 5% RH) as $[(M_{wet} - M_{dry})/M_{dry}] \times 100$, where M_{wet} is the fiber mass after 24 h of immersion and M_{dry} is the fiber mass before immersion.

The test of water release over time was performed to quantify the amount of water released by the saturated microfibers in the FRC mix. This phenomenon would promote deferred cement hydration over time and is important in determining the water/cement ratio for the mix design of FRCs. For this purpose, 10 g of microfibers was saturated in water for 24 h at laboratory conditions, then drained and placed in a glass vessel. The container was placed on a scientific balance in a climate chamber set at 25 °C and 50% RH. The weight loss of the microfibers was continuously recorded for 24 h and defined as the amount of water released over time.

2.2. Treatments of the Textile Waste Microfibers

The textile waste microfibers were considered in three different conditions within the cementitious matrix: untreated, water-saturated, and mercerized (NaOH-treated). The influence of each state was observed and derived from the characterization of FRCs.

The untreated reference condition referred to dry textile microfibers as supplied by the manufacturer.

The water-saturated condition was involved to better control the microfiber water absorption and release over time and eventual promotion of cement hydration over time.

The mercerization treatment (hereafter named NaOH treatment) was involved to possibly increase the mechanical properties of the investigated textile microfibers, and thus those of the FRCs. This treatment, generally used for cotton fiber and other cellulose fibers, confers improved physical and mechanical properties to the textile fibers. On the one hand, it results in the breakdown and fibrillation of fibers into smaller and thinner fibers, leading to an increase in aspect ratio. On the other hand, the surface becomes rougher and reactive, allowing for better adhesion and wettability of the fibers with the matrix, respectively. The polymerization degree and molecular orientation of cellulose crystals change due to the cementation of hemicellulose and lignin, which are removed during the process. These changes enhance the mechanical properties and tensile strength [24,25]. First, the microfibers were dried at 80 °C until reaching a mass change between two measurements lower than 0.1%. Then, they were soaked for 30 min in a 5% NaOH solution

of demineralized water. Subsequently, the microfibers were bathed while being stirred in demineralized water for 15 min; this process was repeated until reaching a neutral pH. In the end, the microfibers were dried again at 80 °C until obtaining a mass change between two measurements lower than 0.1%.

Water-saturated and NaOH-treated conditions improve FRC properties primarily through the optimization of natural microfiber properties. Although these treatments are not specific to synthetic microfibers, an optimization of the blended microfiber properties promotes the enhancement of all textile waste microfibers, including synthetics, and thus the mitigation of FMPs from the environment.

2.3. Cement-Based Composites Reinforced by Textile Waste Microfibers (FRCs)

CEM-I 42.5R Portland cement (Cemento Grigio 425, Knauf, Iphofen, Germany) was used to manufacture FRCs. The mix design of the FRCs considered a constant effective water-to-cement ratio (w_{eff}/c) equal to 0.42 and microfiber contents of 0 wt%, 1 wt%, 2 wt%, 3 wt%, and 4 wt%. Microfiber contents above 4 wt% were not investigated because of the workability loss of the FRCs in the fresh state. For the FRCs containing water-saturated microfibers, the amount of water absorbed and then released over time ($w_{released}$) was considered as part of the effective water (w_{eff}) intended for cement hydration. Thus, for FRC mixes with water-saturated microfibers, the amount of water added during the mixing phase (w) was computed as the difference between the effective water and the water released over time ($w = w_{eff} - w_{released}$). The cement was slowly added to water in an electronic mixer. The paste was mixed at a slow speed (220 rpm) for 1 min and at a high speed (440 rpm) for 4 min. Then, the selected content of microfibers (untreated, water-saturated, or NaOH-treated) was added to the paste and manually mixed with a trowel. The mix was cast into $50 \times 35 \times 300$ and $30 \times 300 \times 300$ mm^3 assembled wooden formworks and was vibrated for 1 min on a vibrating plate (55-C0157/B Controls Group). Then, it was covered with a plastic cloth and cured for 28 days in a climatic chamber at 25 ± 2 °C, 95 ± 5% RH. Four samples for each mix were manufactured for the linear shrinkage and three-point bending tests ($50 \times 35 \times 300$ mm^3 beams), and the thermal conductivity measurement ($30 \times 300 \times 300$ mm^3 slabs). The three-point bending test was performed by using a UTM INSTRON 5567 under displacement mode (1 mm/min), considering a 30 kN load cell and 10 cm mid-span. A heat flow meter HFM Lambda (Netzsch-Gerätebau GmbH, Selb, Germany) was used to measure the thermal conductivity of the FRC samples reinforced with untreated microfibers. For all tests on FRCs, the desired mean features were averaged over four measurements, and the standard deviation was computed.

3. Results and Discussion

3.1. Characterization of Textile Waste Microfibers

The composition of the microfibers under investigation was already known (61% cotton, 29% cotton blend, and 10% synthetics) from the composition data supplied by the producer (see Section 2.1). In addition to the 10% pure synthetic fibers, additional synthetic microfibers were present in the 29% cotton blend fraction, mainly PA.

The microfiber composition was experimentally validated through a comparison between the ATR-FTIR spectrum of the textile waste microfibers and that of the one-component textile fibers (i.e., pure cotton, nylon, polyester, etc.). For conciseness, Figure 2 reports the spectrum of the textile waste microfibers in which the characteristic peaks of cotton (green) and synthetics (red) are highlighted. The identification of the characteristic peaks is confirmed by the literature [26–28].

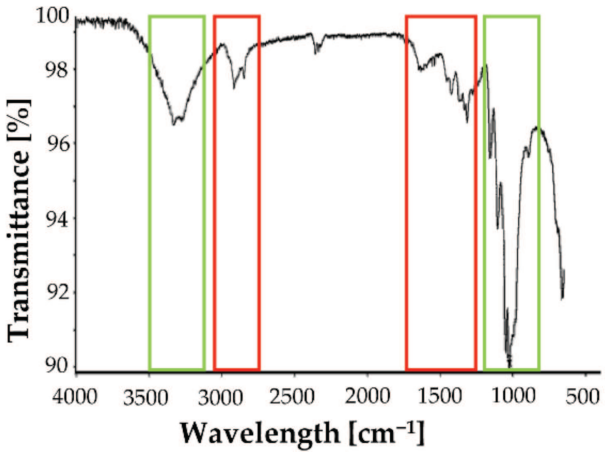

Figure 2. ATR-FTIR spectroscopy of textile waste microfibers. Characteristic peaks related to synthetic microfibers are highlighted in red, while those of cotton in green.

ESEM microscopy (Figure 3) confirmed this evidence by detecting cotton and synthetic fibers with a predominance of cotton. In particular, cotton fibers can be distinguished from synthetic ones since they are flattened and rolled up [27–29]. By contrast, synthetic fibers display a cylindrical cross-section with a rough surface and grooves (Figure 3) [27,28].

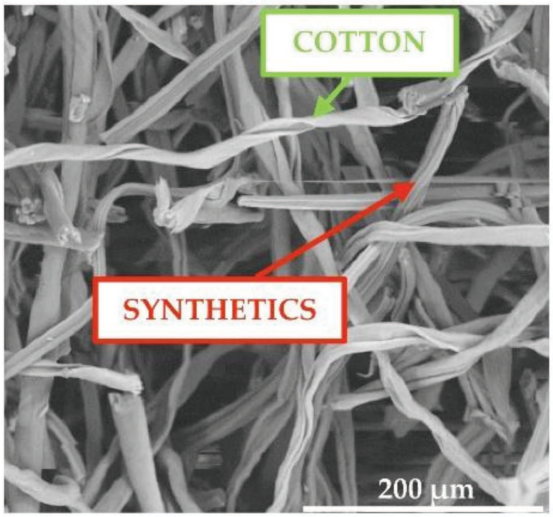

Figure 3. ESEM micrograph of textile waste microfibers at 600× magnitude.

Focusing on synthetic microfibers (at least 10% of the textile microfibers under investigation), a length of 3.65 ± 2.57 mm and a diameter of 17.28 ± 1.68 µm were determined with ImageJ software from optical images (Figure 4). Since synthetic microfibers displayed a 0.3 µm–15 mm length and a length-to-diameter ratio of greater than three, then they can be considered a source of FMPs according to the ECHA definition.

The mean real density of microfibers was evaluated at 1.15 g/cm^3. It was found that the microfibers were dry while displaying a water absorption of 340%. However, this amount of water was entirely released over time ($w_{released}$), and 95% of the water was released after 24 h. During the first few hours (from 0 to 3:45 h, Figure 5a), the water release was slower, but then it followed a faster and linear trend over time (from 3:45 to 7:30 h, Figure 5b).