BIO-BASED ADDITIVE MANUFACTURING USING ENZYMATICALLY RECYCLED COTTON TEXTILE WASTE

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Abstract

Apparel waste management is a global challenge that is becoming increasingly dire with the rise of fast fashion supply chain models, resulting in the projected accumulation of gigatons of textile waste in the world's landfills by 2040. Moving towards textile circularity would divert waste from landfills, reduce dependence on carbon- and resource-intensive virgin materials, and serve as an opportunity for important value retention of these highly engineered materials. However, textile recycling is challenging due to the difficulty of separating complicated apparel products and fiber blends into useful waste streams. Enzymes present a potential solution because, together with simple filtration, they are adept at separating fiber blends (unlike mechanical recycling techniques such as shredding) due to their substrate selectivity (i.e. extracting cotton fragments form a cotton/polyester blend while leaving the polyester intact). NCSU's Textile Biocatalysis Research group has developed an enzyme-mediated process to efficiently degrade the cotton components of model apparel materials into slurries of micro-scale cotton fiber fragments (CFFs) and soluble sugars under mild reaction conditions. In addition to attractive cellulose attributes such as biodegradability, thermomechanical stability, and readily modified chemical functionality, the residual CFFs are highly crystalline and have dimensions that are suitable for the development of bio-based additive manufacturing feedstocks. The focus of this research is the reassembly of these enzymatically degraded micro-scale fragments into industrially relevant, bio-based macro-scale objects (such as structural meshes and grids for apparel, footwear, and home furnishings) via 3D printing. Rather than disrupting the CFF crystallinity via harsh dissolution procedures, as occurs during the production of viscose or lyocell, the fragments will be suspended in extrudable liquid media, with minimal added compounds in alignment with green chemistry principles, and then coalesced to achieve structural integrity. Initial prototypes have shown both the feasibility of the CFFs as an additive manufacturing feedstock material, as well as their potential industrial versatility by modifying feedstock formulation and post-print processing conditions towards diverse, application-specific final object properties. The fabrication of proof-of-concept cellulosic bioink printed prototypes will serve to demonstrate a potential pathway for commercial valorization of the waste cotton fiber fragments and incentivize industrial textile waste recycling.

Introduction and Motivation

Spurred by the rise of fast fashion industrial models, reports estimate that as much as 92 million tons of textile waste is sent to landfills around the world on an annual basis [1]. Despite the potential hazards of this level of solid waste accumulation, textile recycling remains uncommon at

industrial scales – less than 1% of global fiber production can be attributed to fibers produced from recycled post-consumer textile waste [2] – because textiles are inherently complex materials for recycling (material blends, dyes, finishes, additives) [3]. Mechanical recycling strategies, while scalable, are fundamentally limited in that they are unable to separate fiber blends, and the resulting fibers must be either downcycled or blended with virgin fiber due to their decrease in quality [4]. Chemical recycling enables material-specific schemes and material separation and has been the subject of much research and development [5]. However, due to the fundamental differences in fiber chemistries, properties, and behavior under recycling conditions (especially between synthetic and natural fibers), recycling schemes are often designed to target the isolation (and preservation) of a primary fiber type. As a result, the harsh reaction conditions (elevated temperatures and/or reaction media) common in successful techniques have been reported to result in unintended damage, reduction in degree of polymerization, and crystallinity loss in non-target blend components, limiting their re-use performance and potential applications [2].

Enzyme-based recycling processes have emerged as a potential solution due to the material-specific chemical activity of these biological catalysts, as well as the environmentally benign conditions under which they operate. The Textile Biocatalysis Research group at NCSU has developed an enzyme-mediated process that can efficiently degrade the cotton fraction of model apparel fabrics into slurries of micro-scale cotton fiber fragments (CFFs) and soluble cellulose monomers and oligomers, while also separating it from non-cellulose fractions in blended-fiber fabrics [6]. Enzymatic degradation of cellulose [7], especially for the production of soluble sugars as industrial chemical precursors [8], has been previously investigated. Because complete degradation requires increased enzyme dosing and longer degradation times, exploring instead the valorization potential of the CFFs may help incentivize industrial textile waste recycling by providing an additional recycling pathway for cotton-containing fabrics that represent a significant portion of the global fiber market. In particular, the CFFs will be explored for their use in the development of bio-based additive manufacturing (AM) feedstocks, with minimal added compounds according to green chemistry principles, by reassembling the micro-scale fragments into bio-based, biodegradable macro-scale objects via 3D printing.

Review of Related Work

While poly(lactic acid) (PLA) remains the only bioderived AM feedstock material that is available at commercially relevant scales, there has been an increased interest in developing alternative bio-based AM feedstocks towards enhanced recyclability, reduced environmental hazards, and improved biocompatibility [9]. Cellulose is particularly promising for this application due to its global abundance, biodegradability, biocompatibility, thermomechanical robustness, electrically insulating properties, shear-thinning-enabled extrudability at room temperature, and readily functionalized hydroxyl groups that enable chemical modification towards application-specific properties [10]. Micro- and nano-scale cellulose particles have been widely utilized as viscosity/texture modifiers and excipients in the cosmetic, pharmaceutical, and food industries, as well as being incorporated as fillers and reinforcing agents in developing manufacturing technologies [11]. The global microcrystalline cellulose market is currently valued at approximately USD 4 billion and is forecasted to grow, though it has been noted that additional material sources will be needed to support this growth [12]. Recent processing strategies using

advanced solvent systems and nanocolloid-based suspensions have enabled the development of extrudable feedstocks with cellulose as the primary component. This work will focus on technologies using non-derivatized aqueous cellulose suspensions towards the promotion of manufacturing techniques that are conducive to environmentally responsible, circular industrial frameworks. Aqueous cellulose suspensions based on nano-scale cellulose particles have been leveraged to produce objects with highly porous microstructures [13], robust mechanical properties [14], and deformable/shape-recoverable functionality [15]. As summarized in a review of recent all-cellulose AM technologies, there are various post-printing solidification methodologies – including air-drying, freeze-drying, wet-densification, and UV-curing – which enable the production of 3D structures with a wide range of properties and enhance the industrial versatility of cellulose-based AM materials [16]. The promising development of these materials and fabrication strategies serves as inspiration for the use of these novel CFFs in developing AM materials that may have distinct properties from those previously reported using cellulose particles from conventional biomass sources and extraction methods. Demonstrating their potential applicability and versatility also highlights future valorization pathways for textile recycling.

Technology Approach

Enzymatic Degradation of Model Fabrics

Model apparel fabrics used in this study include: bleached, undyed 100% cotton knit (White Cotton, WC), 100% cotton medium-wash denim (Denim, D), 100% cotton knit dyed with Reactive Red 198 (Red Cotton, RC), and a 50/50 cotton/polyester blend knit with the polyester fraction dyed using Disperse Red 177 (Red Polyester White Cotton, RPWC). The degradation and separation procedures were adapted from those described by Egan et al. and follow the same protocol described in detail there, unless otherwise noted [6]. Enzymatic degradation was performed on 1-gram samples (precut to 1 cm x 1 cm squares) using a cellulase enzyme liquid preparation dose of 4.6% on weight of fabric. Temperature and pH were controlled at 50 °C and 5, respectively, in 100 mM sodium acetate buffer. Degradation took place in sealed containers for 19 hours in a 2mag Stirring Drybath 15–250 (2mag-USA), with samples vigorously agitated with stir bars. Residual large solids were separated from the resulting slurry using a 2 mm mesh screen and cotton fiber fragments were collected via vacuum filtration (G6 filters) and then air-dried at room temperature. Prior to further characterization and/or use in the development of additive manufacturing feedstocks, the cotton fiber fragments were dried at 105 °C until less than 1 mg of weight loss over a 50 second period was achieved (Mettler Toledo HC103 Moisture Analyzer).

Characterization of Cotton Fiber Fragments

Scanning electron microscopy (SEM) was used to observe the fabric structure of pre- and post-treatment fabrics as well as the size and morphology of the produced cotton fiber fragments (TM4000Plus Tabletop Microscope, Hitachi High-Tech Corporation). Samples were mounted on

stainless steel specimen stubs using conductive tape and then sputtered with gold using a vacuum sputter-coater (Emitech SC7620 Sputter Coater). Samples were observed using a 15 kV accelerating voltage and the backscattered electron collection setting. Fourier transform infrared spectroscopy (FTIR) was performed on the pre-degradation red polyester white cotton blend, the post-degradation "cleaned" polyester residual solids, and the post-degradation cotton fiber fragments using a Thermo Fisher Nicolet iS50 FTIR spectrometer with a diamond attenuated total reflection crystal sampling head. Each sample was scanned 32 times from 400-4000 cm^{-1} with a 4 cm^{-1} resolution.

Preparation of Additive Manufacturing Feedstocks and Prototypes

To prepare feedstocks for additive manufacturing, cotton fiber fragments were dried as described above then deionized water was added to reach a CFF-content of 20 wt%. Feedstocks were briefly mixed by hand using a spatula and then mixed for 2 minutes at 2500 rpm and 1 minute at 3000 rpm using a Flacktek 330-100 SE SpeedMixer. The mixed feedstock was then loaded into a syringe with a conical nozzle (22G for WC, D, RPWC; 18G for RC). 3D structures designed using the 3D Builder software (Microsoft) were printed using a TissueStart^{TM} bioprinter (TissueLabs) with a varying printing speed from 3-5 mm/s. Freshly printed samples were frozen at -30 °C and then freeze-dried using a FreeZone 1 Liter Benchtop Freeze Dry System (Labconco) operating under a 9 Pa vacuum pressure overnight to obtain the final structures.

Discussion

Enzymatic Degradation of Model Fabrics

The model fabrics highlighted in this study were selected for two primary reasons. First, cotton and polyester, together, represent nearly 80% of global fiber production (polyester: 59%, cotton: 19%) according to the 2025 Materials Market Report [17], thus, recycling technologies should be directed towards processing cotton, polyester, and/or blends of the two to achieve significant volume impact. Second, different fabric types were included to explore the ways in which diverse fabric structures and chemical treatments that are present in real-world textile waste materials affect recycling strategy efficacy. As shown in Figure 1, treatment of the RC fabric resulted in approximately 25% residual large solids (material that resisted enzymatic degradation) compared to the complete conversion to a slurry of small solids (micro-scale cotton fiber fragments) and glucose/other soluble cellulose oligomers (weight loss) in the case of undyed WC. Reactive dyes, like Reactive Red 198 used to dye the RC samples in this study, are relatively large molecules that are covalently bonded to the cellulose fiber and, as a result, impede the interaction between cellulase enzymes and cellulose chains necessary for complete degradation [6]. In contrast, almost no large solids remained after Denim sample degradation, highlighting the critical impact of different dyeing techniques/mechanisms on cellulase degradation activity (namely, the indigo is not covalently bonded to the cellulose polymer) and suggesting that fiber chemistry (cotton

content, dyes, etc.) play a more significant role in determining degradation outcomes than fabric structure (i.e. woven denim compared to the knit of the WC and RC samples). As expected, the sample with the largest residual large solids fraction was the RPWC blend. Remaining large solids are primarily due to the polyester fraction (50% of the original fabric, by weight) which are not affected by the substrate-specific cellulase enzymes. However, it is important to note that the measured large solids fraction was greater than the 50% associated with the polyester content signifying that there is some portion of the cotton fibers that were not completely removed by the enzymatic degradation process. This is likely due to the fact that textile fibers are intimately blended in the twisted yarns which could both inhibit enzyme access to the cotton fibers and trap degraded fiber fragments during the post-degradation separations process. While not shown here, it was previously reported that additional enzymatic treatments can achieve complete extraction of the cotton fraction and produce a completely isolated polyester fraction [6].

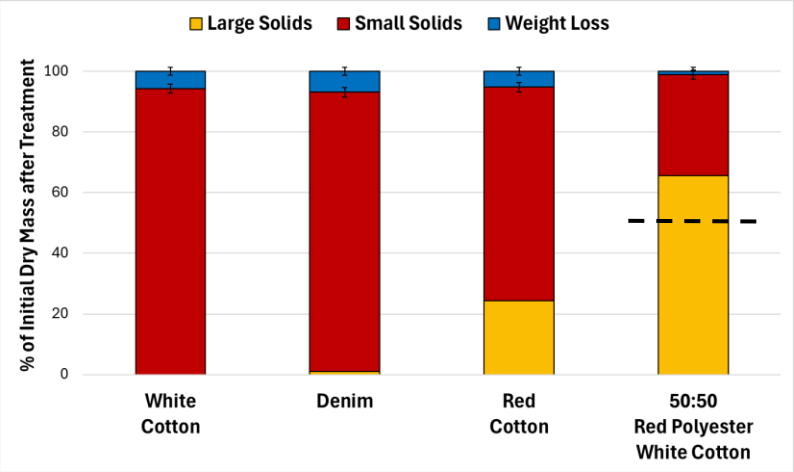

Figure 1. Enzymatic degradation of cotton-containing fabrics yields slurries of soluble fractions and cotton fiber fragments (in the case of bleached white cotton and denim), with a residual solid fraction that resists degradation due to the inhibitory effect of dye molecules (red cotton) and the presence of non-cellulosic fibers (red polyester white cotton blend).

Characterization of Degradation Products

To further understand the nature of the fractions produced by the enzymatic degradation process, the small solids and both pre- and post-treatment RPWC fabrics were inspected via SEM. These results are summarized in Figure 2 below. Prior to enzymatic degradation, the fabric is composed of a dense twisted yarn structure in which both cotton (twisted, somewhat irregularly shaped) and polyester (smooth, circular cross-section) fibers can be observed. After the enzymatic "cleaning" process, there are two distinct, separatable (a critical advantage of the enzymatic recycling process) fractions – micro-scale cotton fiber fragments (20-60 μm in length), which appear essentially identical under SEM regardless of source fabric, and an intact fabric composed almost entirely of polyester (as mentioned previously, a subsequent treatment can achieve complete cotton removal) that could be fed into polyester recycling streams as a simpler, mono-material recycling feedstock

compared to a fiber blend. As polyester recycling strategies are already the subject of significant exploration and progress [18], the technological potential of the cotton fiber fragments is the focus of this study. Namely, the size and elongated, fibrillar morphology of these CFFs both distinguish them from commercially available micro-scale cellulose particles that are most often chemically extracted from virgin woody biomass (> 100 µm in length for purchased reference material) and yet also lend themselves to use in previously demonstrated commercial and experimental applications for conventional cellulose particles. While not the only potential application, their use as a source material for novel bio-based, biodegradable additive manufacturing feedstocks is demonstrated herein.

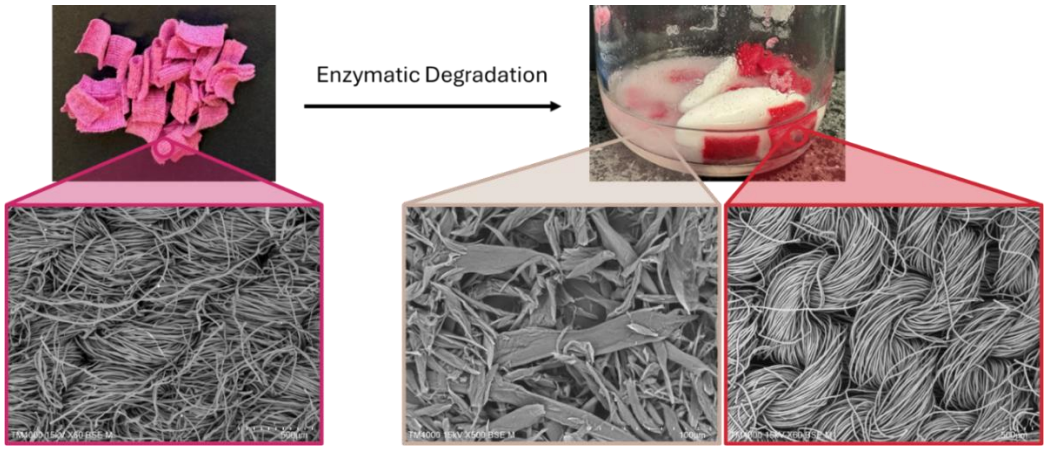

Figure 2. Scanning electron microscopy shows the enzymatic textile fiber separation of a 50/50 cotton/polyester blend (left) into micro-scale cotton fiber fragments (middle) and "cleaned", recyclable polyester that retains pre-degradation fabric structure (right).

This separation is also confirmed via FTIR as shown in Figure 3. The broad peak at 3200-3500 cm^{-1} associated with the hydroxyl groups present in cellulose [19] appears in both the untreated blend as well as the post-degradation cotton fiber fragments but is not observed in the cleaned polyester spectra. Conversely, in the spectra of the blend and post-degradation polyester samples, characteristic peaks at around 1700 cm^{-1} and 1300 cm^{-1} associated with the ester carbonyl bond stretching and ester group stretching [20], respectively, denote the presence of polyester and are not observed for the 100% cellulose cotton fiber fragments. The use of FTIR in this case does qualitatively enable the differentiation between samples that are made of predominantly cotton, predominantly polyester, and large fractions of both (pre-degradation blend). However, while cotton accounts for about 23% by weight of the large solids remaining after degradation (yellow bar – RPWC sample, Figure 1), it was not easily identifiable in the FTIR spectrum of the "cleaned" RPWC sample because it was trapped or buried within the residual polyester yarn structure. This exemplifies an important limitation of FTIR characterization of fiber blends (namely, the challenge in characterizing the presence of fibers at low to intermediate contents) which is an emerging technology of interest in the field of textile waste sorting. By extension, this demonstrates the importance of effective fiber separation strategies given that the identification of fiber blends remains a critical challenge in textile recycling implementation (to say nothing of their subsequent separation and recycling).

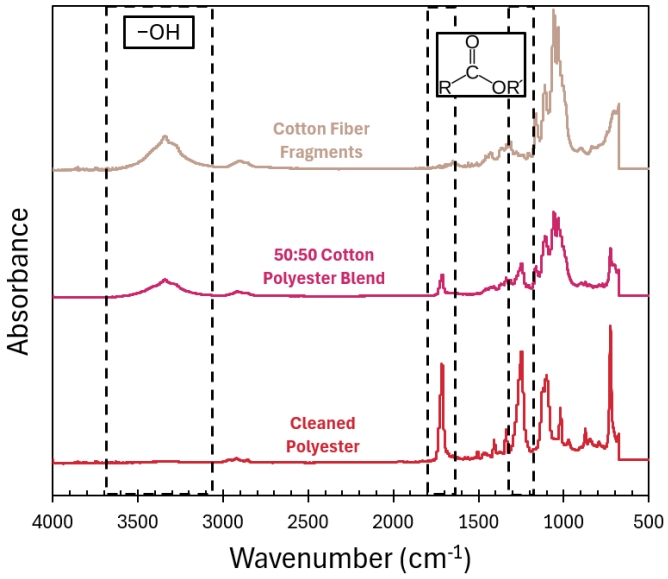

Figure 3. FTIR spectra of pre-degradation 50/50 cotton/polyester blend and post-degradation cotton fiber fragments and residual cleaned polyester confirm fiber separation.

Additive Manufacturing Using Cotton Fiber Fragments

After their isolation via the previously described separations process, the cotton fiber fragment powders were suspended in water to form extrudable feedstocks which were then used to fabricate prototypical 3D structures via non-thermal, extrusion-based additive manufacturing, as shown in Figure 4. This process was successfully demonstrated using cotton fiber fragments derived from all four model apparel fabrics, highlighting the potential for this recycling strategy to be applied to diverse, real-world textile waste materials. Notably, while the feedstocks prepared from the White Cotton, Denim, and Red Polyester White Cotton samples were consistently extrudable through a 22G conical nozzle (internal diameter = 410 µm), the Red Cotton-based feedstock faced issues with clogging and inconsistent extrusion and was thus only extruded through a 18G nozzle (internal diameter = 840 µm). Additionally, despite preparation via an identical protocol, the Red Cotton feedstock had an observably lower viscosity, resulting in poor fidelity to the digital design, as shown by its more irregular final printed shape. This result suggests the importance of feedstock formulation optimization according to the specific properties of the cotton fiber fragments – such as particle morphology, particle behavior in suspension, and hydrophilicity – which may be affected by source-fabric attributes including the presence of dyes and finishes. The prototypes shown in Figure 4 were all solidified via freeze-drying to preserve the as-printed dimensions for the purpose of demonstrating as-printed design-print shape fidelity. As a demonstration of a textile-relevant application of these materials, the WC-derived feedstock was also used to fabricate a button. Because the feedstocks have a significant initial water content (~77% be weight), the final solidification mechanism plays a critical role in determining final object properties. Preliminary studies have shown that other solidification techniques, such as air-drying at room temperature, can be used to produce densified structures with distinct microstructural and mechanical properties from the same starting feedstock. While further exploration and optimization of both feedstock

formulation and post-processing solidification mechanisms will be necessary, the ability shown here to fabricate solid objects with tunable properties serves to demonstrate the potential versatility of this recycled textile waste-based manufacturing technology.

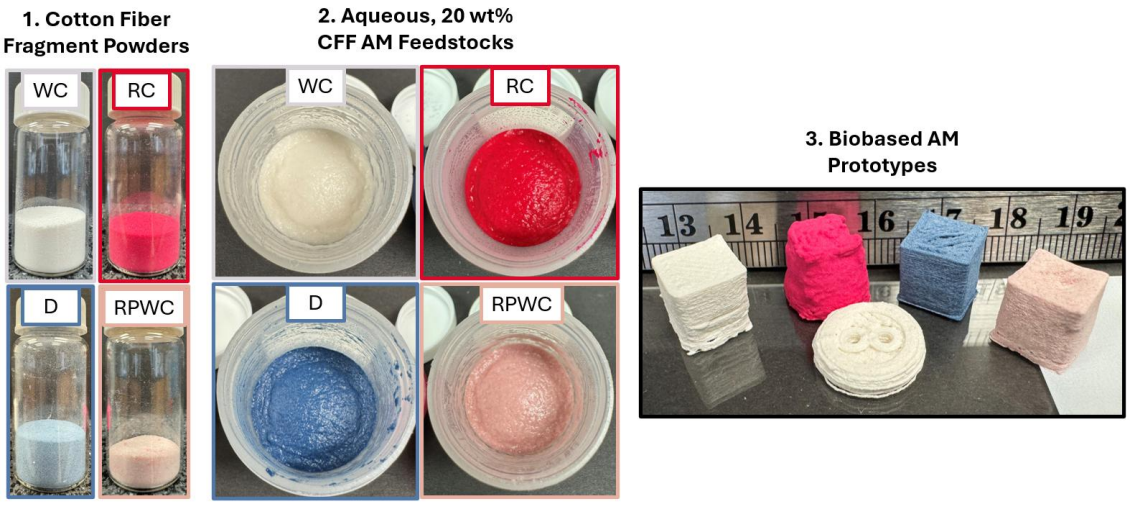

Figure 4. Cotton fiber fragment powders produced via the enzymatic degradation of model apparel fabrics (Step 1) are suspended in aqueous media to form extrudable additive manufacturing feedstocks (Step 2) which are used to produce prototypical 3D structures (Step 3).

Conclusions & Recommendations

An enzyme-mediated recycling strategy was used to convert the cotton fraction of various cotton-containing textiles to micro-scale cotton fiber fragments under mild reaction conditions. Notably, this process was able to isolate these particles from fiber blends without damaging, or otherwise affecting the recycling potential of, other fiber types. CFFs from all studied fabric sources were then utilized to formulate extrudable feedstocks that were converted to solidified 3D prototypes via additive manufacturing. Further investigation of feedstock formulation and post-print processing will seek to enhance the potential versatility of these materials. In particular, feedstocks will need to be optimized to accommodate the variations in material properties of different fabric sources towards maximizing design-object fidelity. Additionally, feedstocks will be quantitatively characterized according to relevant rheological parameters in order to establish a framework for future exploration of cellulose- and other bio-based additive manufacturing materials. Additive manufacturing feedstocks prepared using CFFs will be compared to analogous formulations using commercially available micro- and nano-cellulose particles to explore the unique properties of materials produced using this recycling pathway and potentially incentivize its future adoption at industrially relevant scales. The enzymatic recycling treatment utilized herein both unlocks an isolated synthetic fiber fraction (most commonly polyester) that, as a blend component, would have previously been more difficult to access for recyclers and generates a new potential product stream from the cotton fraction, thus potentially enhancing the recyclability of significant portions of global textile waste. By demonstrating a viable valorization pathway for this unique textile

recycling product (CFFs) – in addition to the existing market for micro-cellulose particles – using mild, environmentally benign manufacturing techniques, this work seeks to highlight ways in which sustainable manufacturing methodologies can meet critical technological needs and embed circularity and resilience in the industrial landscape.

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