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(54) Title: ENZYMATICALLY PRODUCED CELLULOSE

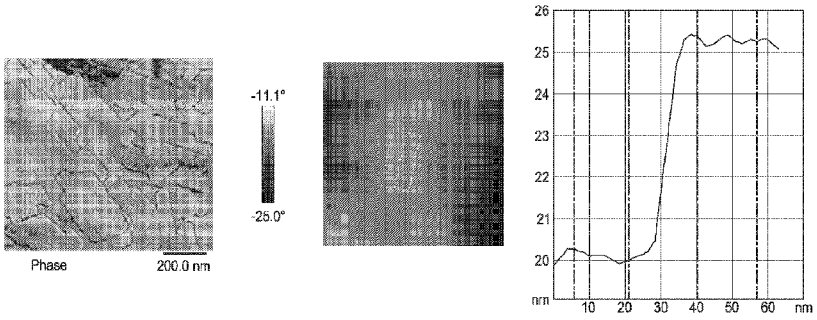

FIG. 1A

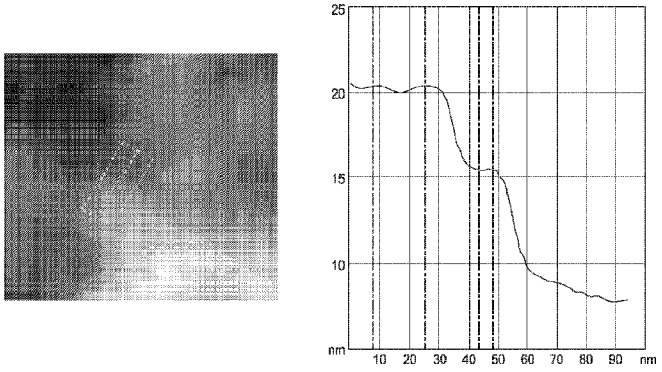

FIG. 1B

(57) Abstract: Compositions are disclosed herein comprising cellulose that has (i) a weight-average degree of polymerization (DP_w) of about 10 to about 1000, (ii) a cellulose II crystal structure, and that is (iii) insoluble in an aqueous composition. Further disclosed are cellodextrin phosphorylase enzymes that synthesize this cellulose material. Methods of using cellulose for viscosity modification or film/coating applications are also disclosed.

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TITLE

ENZYMATICALLY PRODUCED CELLULOSE

This application claims the benefit of International Application Nos. PCT/CN2014/094594 (filed December 23, 2014) and PCT/CN2014/094593 (filed December 23, 2014), both of which are incorporated herein by reference in their entireties.

FIELD OF INVENTION

The present disclosure is in the field of polysaccharides. More specifically, the disclosure pertains to low molecular weight insoluble cellulose and enzymatic reactions for its synthesis. The disclosure also regards using cellulose in various applications such as viscosity modification and film production.

REFERENCE TO SEQUENCE LISTING SUBMITTED ELECTRONICALLY

The official copy of the sequence listing is submitted electronically via EFS-Web as an ASCII formatted sequence listing with a file named CL6399WOPCT2_SequenceListing_ST25 created on December 9, 2015, and having a size of 39.4 kilobytes and is filed concurrently with the specification. The sequence listing contained in this ASCII-formatted document is part of the specification and is herein incorporated by reference in its entirety.

BACKGROUND

Driven by a desire to find new structural polysaccharides using enzymatic syntheses or genetic engineering of microorganisms, researchers have discovered polysaccharides that are biodegradable and can be made economically from renewably sourced feedstocks. One such polysaccharide is cellulose, a glucan polymer characterized by having beta-1,4-glycosidic linkages.

Microcrystalline cellulose (MCC) is a white, odorless, tasteless, relatively free flowing, crystalline powder that is virtually free from organic and inorganic contaminants. It is a purified, partially depolymerized cellulose obtained by subjecting alpha cellulose obtained as a pulp from fibrous plant material (e.g., wood) to hydrolytic degradation, typically with mineral acid. MCC is a highly crystalline particulate cellulose consisting primarily of crystalline aggregates obtained by removing amorphous (fibrous cellulose) regions of a cellulosic material. MCC is used in a variety of applications including foods, pharmaceuticals and cosmetics. Despite MCC's various applications, preparation of

this cellulose type is laborious and expensive. Also, activation of MCC requires high shear.

Development of new forms of cellulose is desirable given the potential utility thereof in various applications. The development of novel enzymatic processes may be a useful means for producing new types of cellulose material.

SUMMARY OF INVENTION

In one embodiment, the present disclosure concerns a composition comprising cellulose, wherein the cellulose:

(i) has a weight-average degree of polymerization (DP_w) of about 10 to about 1000,

(ii) has a cellulose II crystal structure, and

(iii) is insoluble in an aqueous composition.

In another embodiment, the DP_w of the cellulose is about 10 to about 100.

In another embodiment, the cellulose is a product of a cellodextrin phosphorylase enzyme comprising an amino acid sequence that is at least 90% identical to SEQ ID NO:2 or SEQ ID NO:6, wherein the substrates for the enzyme comprise cellodextrin and glucose-1-phosphate. In another embodiment, the cellulose produced by the enzyme has not been subjected to a mercerization or derivatization process.

In another embodiment, the composition is a film or coating. The film or coating has a uniform thickness of at least about 4 nm in another embodiment. In another embodiment, the film or coating exhibits low permeability to, or is impermeable to, an aqueous composition, lipophilic composition, or gaseous composition. The film or coating is on paper in another embodiment.

In another embodiment, the composition is an aqueous composition, optionally having a viscosity of at least about 100 cPs. The aqueous composition is a colloidal dispersion in another embodiment. The concentration of the cellulose in the aqueous composition is less than about 10 wt% in another embodiment. In another embodiment, the composition is a food product, personal care product, pharmaceutical product, household product, or industrial product.

In another embodiment, the cellulose is soluble in a solvent comprising DMSO and/or DMAc.

In another embodiment, the present disclosure concerns a method for increasing the viscosity of an aqueous composition. This method comprises contacting cellulose

with an aqueous composition, wherein the cellulose is insoluble in the aqueous composition and has (i) a weight-average degree of polymerization (DP_w) of about 10 to about 1000, and (ii) a cellulose II crystal structure. The contacting step results in increasing the viscosity of the aqueous composition, in comparison to the viscosity of the aqueous composition before the contacting step. In certain embodiments of this method, the shear thinning behavior of the aqueous composition is increased by the cellulose compared to the shear thinning behavior of the aqueous composition as it existed before the contacting step.

In another embodiment, the present disclosure concerns a method of treating a material. This method comprises: (a) contacting a material with an aqueous composition comprising cellulose, wherein the cellulose is insoluble in the aqueous composition and has (i) a weight-average degree of polymerization (DP_w) of about 10 to about 1000, and (ii) a cellulose II crystal structure; and (b) drying the aqueous composition, wherein the drying step leaves a deposit of the cellulose on the surface of the material.

BRIEF DESCRIPTION OF THE DRAWINGS AND SEQUENCES

FIG. 1A: Atomic force microscopy (AFM) was used to analyze a thin film made from drying a colloidal dispersion of insoluble cellulose synthesized by an *R. champanellensis* cellodextrin phosphorylase enzyme. The thickness of the sheet structure is about 5 nm. Refer to Example 4.

FIG. 1B: AFM was used to analyze a thin film made from drying a colloidal dispersion of insoluble cellulose synthesized by a *V. ruber* cellodextrin phosphorylase enzyme. The thickness of the sheet structure is about 4.8 nm. Refer to Example 4.

FIG 2: Viscosity versus shear rate, as measured for colloidal dispersions of insoluble cellulose material synthesized by *R. champanellensis* cellodextrin phosphorylase (blue diamonds, sample 1, 2.5 wt% in water) or *V. ruber* cellodextrin phosphorylase (red squares, sample 2, 1.7 wt% in water). Refer to Example 4.

FIG 3: Viscosity of various commercially available water-soluble polysaccharides (carboxymethyl cellulose [CMC] and scleroglucan) in water compared to the viscosity of colloidal dispersions of insoluble cellulose material synthesized by *R. champanellensis* cellodextrin phosphorylase (2.5 wt% in water) or *V. ruber* cellodextrin phosphorylase (1.7 wt% in water). CMC of DP_w 3200 and 2000 were from CP Kelco, and CMC of DP_w

50, 360 and 1200 were FINNFIX brand CMC from CP Kelco. Scleroglucan was from Cargill (ACTIGUM). Viscosity measurements are reported at 10 1/s shear rate.

Table 1. Summary of Nucleic Acid and Protein SEQ ID Numbers

Description	Nucleic acid SEQ ID NO.	Protein SEQ ID NO.
"VruCdp1", *Vibrio ruber* DSM14379 cellodextrin phosphorylase.	1 (2415 bases)	2 (805 aa)
"VruCdp1", *Vibrio ruber* DSM14379 cellodextrin phosphorylase. Nucleotide sequence codon-optimized for expression in *E. coli*. Amino acid sequence contains additional C-terminal residues (L-E-6xHis).	3 (2442 bases)	4 (813 aa)
"RchCdp1", *Ruminococcus champanellensis* 18P13 cellodextrin phosphorylase. GENBANK Accession No. WP_015559149 (amino acid sequence).	5 (2397 bases)	6 (798 aa)
"RchCdp1", *Ruminococcus champanellensis* 18P13 cellodextrin phosphorylase. Nucleotide sequence codon-optimized for expression in *E. coli*. Amino acid sequence contains additional C-terminal residues (L-E-6xHis).	7 (2421 bases)	8 (806 aa)

DETAILED DESCRIPTION

The disclosures of all cited patent and non-patent literature are incorporated herein by reference in their entirety.

Unless otherwise disclosed, the terms "a" and "an" as used herein are intended to encompass one or more (i.e., at least one) of a referenced feature.

Where present, all ranges are inclusive and combinable, except as otherwise noted. For example, when a range of "1 to 5" is recited, the recited range should be construed as including ranges "1 to 4", "1 to 3", "1-2", "1-2 & 4-5", "1-3 & 5", and the like.

The terms "cellodextrin phosphorylase", "cellodextrin phosphorylase enzyme" and the like are used interchangeably herein. A cellodextrin phosphorylase is of the Enzyme Commission (EC) entry 2.4.1.49 and belongs to glycosyl hydrolase family 94 (GH94) according to the CAZy (Carbohydrate-Active EnZymes) database. A cellodextrin phosphorylase can reversibly catalyze synthesis of cellulose and free phosphate (products) from alpha-D-glucose-1-phosphate and cellodextrin (substrates). Such a reaction can also be written as: glucose-1-phosphate + $(1,4\text{-beta-D-glucosyl})_{n-1}$ $\longrightarrow$ $(1,4\text{-beta-glucosyl})_n$ + phosphate, where "$(1,4\text{-beta-D-glucosyl})_{n-1}$" refers to

cellodextrin and "(1,4-beta-glucosyl)$_n$" refers to cellulose. A cellodextrin phosphorylase in certain aspects herein can synthesize low molecular weight cellulose (e.g., DP_w of 10-30) that is insoluble in aqueous compositions. A cellodextrin phosphorylase in certain aspects herein comprises an amino acid sequence that is at least 90% identical to SEQ ID NO:2 or 6.

The term "cellulose" refers to a glucan polysaccharide having a linear chain of beta-1,4-linked D-glucose monomeric units. Cellulose can optionally be represented as (1,4-beta-D-glucosyl)$_n$, where n can be the same value as a DP_w value of a low molecular weight cellulose as disclosed herein (e.g., 10 to 30). The term "glucan" herein refers to a polysaccharide of D-glucose monomers that are linked by glucosidic linkages, which are a type of glycosidic linkage.

The terms "cellulose II structure", "cellulose II crystal structure", "cellulose II" and the like are used interchangeably herein. Cellulose II structure has been described by Kolpak and Blackwell (*Macromolecules* 9:273-278) and Kroon-Batenburg and Kroon (*Glycoconjugate J.* 14:677-690), for example, both of which are incorporated herein by reference. The dominant hydrogen bonds characterizing cellulose II structure are O2-H---O6, O6-H---O6 and O2-H---O2, whereas cellulose I has O2-H---O6 as a dominant hydrogen bond. The structure of cellulose II comprises chain folding and is difficult to unravel. Cellulose II comprises anti-parallel chains, whereas in contrast, cellulose I chains are parallel.

The terms "glycosidic linkage", "glycosidic bond" and the like are used interchangeably herein and refer to the covalent bond that joins a carbohydrate molecule to another carbohydrate molecule. The terms "glucosidic linkage", "glucosidic bond" and the like are used interchangeably herein and refer to a glycosidic linkage between two glucose molecules in a glucan. The term "beta-1,4-glucosidic linkage" as used herein refers to the covalent bond that joins glucose molecules to each other through carbons 1 and 4 on adjacent glucose monomers in a glucan.

The glycosidic linkage profile of cellulose herein can be determined using any method known in the art. For example, a linkage profile can be determined using methods that use nuclear magnetic resonance (NMR) spectroscopy (e.g., ^{13}C NMR or ^{1}H NMR). These and other methods that can be used are disclosed in Food Carbohydrates: Chemistry, Physical Properties, and Applications (S. W. Cui, Ed.,

Chapter 3, S. W. Cui, Structural Analysis of Polysaccharides, Taylor & Francis Group LLC, Boca Raton, FL, 2005), which is incorporated herein by reference.

The "molecular weight" of a saccharide polymer herein, such as cellulose, can be represented as number-average molecular weight (M_n) or as weight-average molecular weight (M_w), the units of which are in Daltons or grams/mole. Alternatively, molecular weight can be represented as DP_w (weight average degree of polymerization) or DP_n (number average degree of polymerization). Various means are known in the art for calculating these molecular weight measurements such as with high-pressure liquid chromatography (HPLC), size exclusion chromatography (SEC), or gel permeation chromatography (GPC).

The term "cellodextrin" as used herein refers to one or more glucose polymers having a length of two or more beta-1,4-linked glucose monomers. Cellodextrin is typically produced via (enzymatic) hydrolysis of cellulose. "Cellobiose" is a type of cellodextrin that comprises two beta-1,4-linked glucose monomers (i.e., cellobiose is a type of disaccharide).

"Glucose-1-phosphate" (G1P) as used herein refers to a glucose molecule with a phosphate group on the 1-carbon. G1P herein can be alpha-D-glucose-1-phosphate.

The terms "enzymatic reaction", "cellodextrin phosphorylase reaction" and the like are used interchangeably herein and, except as otherwise noted, refer to a reaction that is performed by a cellodextrin phosphorylase enzyme. An enzymatic reaction generally refers to a solution comprising at least one active cellodextrin phosphorylase enzyme in a solution comprising water, glucose-1-phosphate, and cellodextrin (e.g., cellobiose), and optionally other components. It is in a cellodextrin phosphorylase reaction where the step of contacting water, glucose-1-phosphate, cellodextrin and a cellodextrin phosphorylase enzyme is performed. The term "under suitable reaction conditions" and the like refer to reaction conditions that support conversion of substrate to low molecular weight, insoluble cellulose via cellodextrin phosphorylase enzyme activity. A cellodextrin phosphorylase reaction herein is not naturally occurring. It would be understood that, as a cellodextrin phosphorylase reaction produces insoluble cellulose, such cellulose is present out of solution.

A "control" enzymatic reaction as used herein can refer to a reaction using a cellodextrin phosphorylase not comprising an amino acid sequence that is at least 90% identical to SEQ ID NO:2 or 6, for example. All the other features (e.g., substrate

concentration, temperature, pH, time) of a control reaction solution can be the same as the reaction to which it is being compared.

A "second reaction" as used herein refers to a reaction that is in addition to a cellodextrin phosphorylase reaction ("first reaction"), and which provides G1P substrate for the first reaction.

"Inorganic phosphate", which can be denoted as P_i refers to a free phosphate ion in solution, and is distinguished from phosphates bound in various phosphate esters.

A "G1P-producing enzyme" can refer to an enzyme that catalyzes synthesis of products in which at least one product is a G1P. Examples of G1P-producing enzymes include starch phosphorylase, sucrose phosphorylase, and cellodextrin phosphorylase (when catalyzing above reaction in reverse direction, i.e., cellulose hydrolysis).

"Starch phosphorylase" as used herein is of the EC entry 2.4.1.1 and can catalyze conversion of starch and inorganic phosphate to glucose-1-phosphate. Such a reaction can also be written as: $(1,4\text{-alpha-D-glucosyl})_n$ + phosphate ⟶ $(1,4\text{-alpha-D-glucosyl})_{n-1}$ + alpha-D-glucose-1-phosphate, where "$(1,4\text{-alpha-D-glucosyl})_n$" refers to starch.

A "starch debranching enzyme" as used herein refers to an enzyme that can catalyze hydrolysis of 1,6-alpha-D-glucosidic linkages, which are at branch points in starch. Examples of starch debranching enzymes herein include pullulanase and isoamylase. A "pullulanase" as used herein is of the EC entry 3.2.1.41. An "isoamylase" as used herein is of the EC entry 3.2.1.68.

The term "sucrose" herein refers to a non-reducing disaccharide composed of an alpha-D-glucose molecule and a beta-D-fructose molecule linked by an alpha-1,2-glycosidic bond. Sucrose is known commonly as table sugar.

"Sucrose phosphorylase" as used herein is of the EC entry 2.4.1.7 and can catalyze conversion of sucrose and phosphate to fructose and G1P. Such a reaction can also be written as: sucrose + phosphate ⟶ fructose + alpha-D-glucose-1-phosphate.

"Cellulosic biomass", "cellulose-comprising biomass" and the like are used interchangeably herein and refer to material comprising the structural portion of plants (e.g., wood, stems) that cannot directly be used for food ingredients or as fermentation substrates.

"Endoglucanase" and "beta-1,4-endoglucanase" are used interchangeably herein and refer to an enzyme that can cleave internal bonds within cellulose chains, making shorter cellulose chains. Such shorter chains are suitable substrates for cellodextrin phosphorylase when catalyzing the above reaction in reverse direction (i.e., cellulose hydrolysis).

The terms "percent by volume", "volume percent", "vol %", "v/v %" and the like are used interchangeably herein. The percent by volume of a solute in a solution can be determined using the formula: [(volume of solute)/(volume of solution)] x 100%.

The terms "percent by weight", "weight percentage (wt%)", "weight-weight percentage (% w/w)" and the like are used interchangeably herein. Percent by weight refers to the percentage of a material on a mass basis as it is comprised in a composition, mixture, or solution.

The term "increased" as used herein can refer to a quantity or activity that is at least about 1%, 2%, 3%, 4%, 5%, 6%, 7%, 8%, 9%, 10%, 11%, 12%, 13%, 14%, 15%, 16%, 17%, 18%, 19%, 20%, 50%, 100%, or 200% more than the quantity or activity for which the increased quantity or activity is being compared. The terms "increased", "elevated", "enhanced", "greater than", "improved" and the like are used interchangeably herein.

The terms "polynucleotide", "polynucleotide sequence", "nucleic acid sequence" and the like are used interchangeably herein. These terms encompass nucleotide sequences and the like. A polynucleotide may be a polymer of DNA or RNA that is single- or double-stranded, that optionally contains synthetic, non-natural or altered nucleotide bases. A polynucleotide may be comprised of one or more segments of cDNA, genomic DNA, synthetic DNA, or mixtures thereof.

The term "gene" as used herein refers to a DNA polynucleotide sequence that expresses an RNA (RNA is transcribed from the DNA polynucleotide sequence) from a coding region, which RNA can be a messenger RNA (encoding a protein) or a non-protein-coding RNA. A gene may refer to the coding region alone, or may include regulatory sequences upstream and/or downstream to the coding region (e.g., promoters, 5'-untranslated regions, 3'-transcription terminator regions). A coding region encoding a protein can alternatively be referred to herein as an "open reading frame" (ORF). A gene that is "native" or "endogenous" refers to a gene as found in nature with its own regulatory sequences; such a gene is located in its natural location in the

genome of a host cell. A "chimeric" gene refers to any gene that is not a native gene, comprising regulatory and coding sequences that are not found together in nature (i.e., the regulatory and coding regions are heterologous with each other). Accordingly, a chimeric gene may comprise regulatory sequences and coding sequences that are derived from different sources, or regulatory sequences and coding sequences derived from the same source, but arranged in a manner different than that found in nature. A "foreign" or "heterologous" gene refers to a gene that is introduced into the host organism by gene transfer. Foreign/heterologous genes can comprise native genes inserted into a non-native organism, native genes introduced into a new location within the native host, or chimeric genes. The polynucleotide sequences in certain embodiments disclosed herein are heterologous. A "transgene" is a gene that has been introduced into the genome by a gene delivery procedure (e.g., transformation). A "codon-optimized" open reading frame has its frequency of codon usage designed to mimic the frequency of preferred codon usage of the host cell.

A "non-native" amino acid sequence or polynucleotide sequence comprised in a cell or organism herein does not occur in a native (natural) counterpart of such cell or organism.

"Regulatory sequences" as used herein refer to nucleotide sequences located upstream of a gene's transcription start site (e.g., promoter), 5' untranslated regions, introns, and 3' non-coding regions, and which may influence the transcription, processing or stability, and/or translation of an RNA transcribed from the gene. Regulatory sequences herein may include promoters, enhancers, silencers, 5' untranslated leader sequences, introns, polyadenylation recognition sequences, RNA processing sites, effector binding sites, stem-loop structures, and other elements involved in regulation of gene expression. One or more regulatory elements herein (e.g., promoter) may be heterologous to a coding region herein.

The term "operably linked" as used herein refers to the association of two or more nucleic acid sequences such that that the function of one is affected by the other. For example, a promoter is operably linked with a coding sequence when it is capable of affecting the expression of that coding sequence. That is, the coding sequence is under the transcriptional control of the promoter. A coding sequence can be operably linked to one (e.g., promoter) or more (e.g., promoter and terminator) regulatory sequences, for example.

The term "recombinant" when used herein to characterize a DNA sequence such as a plasmid, vector, or construct refers to an artificial combination of two otherwise separated segments of sequence, e.g., by chemical synthesis and/or by manipulation of isolated segments of nucleic acids by genetic engineering techniques. Methods for preparing recombinant constructs/vectors herein can follow standard recombinant DNA and molecular cloning techniques as described by J. Sambrook and D. Russell (Molecular Cloning: A Laboratory Manual, 3rd Edition, Cold Spring Harbor Laboratory Press, Cold Spring Harbor, NY, 2001); T.J. Silhavy et al. (Experiments with Gene Fusions, Cold Spring Harbor Laboratory Press, Cold Spring Harbor, NY, 1984); and F.M. Ausubel et al. (Short Protocols in Molecular Biology, 5th Ed. Current Protocols, John Wiley and Sons, Inc., NY, 2002), for example.

The term "transformation" as used herein refers to the transfer of a nucleic acid molecule into a host organism or host cell by any method. A nucleic acid molecule that has been transformed into an organism/cell may be one that replicates autonomously in the organism/cell, or that integrates into the genome of the organism/cell, or that exists transiently in the cell without replicating or integrating. Non-limiting examples of nucleic acid molecules suitable for transformation are disclosed herein, such as plasmids and linear DNA molecules. Host organisms/cells herein containing a transforming nucleic acid sequence can be referred to as "transgenic", "recombinant", "transformed", engineered, as a "transformant", and/or as being "modified for exogenous gene expression", for example.

The terms "sequence identity" or "identity" as used herein with respect to polynucleotide or polypeptide sequences refer to the nucleic acid bases or amino acid residues in two sequences that are the same when aligned for maximum correspondence over a specified comparison window. Thus, "percentage of sequence identity" or "percent identity" refers to the value determined by comparing two optimally aligned sequences over a comparison window, wherein the portion of the polynucleotide or polypeptide sequence in the comparison window may comprise additions or deletions (i.e., gaps) as compared to the reference sequence (which does not comprise additions or deletions) for optimal alignment of the two sequences. The percentage is calculated by determining the number of positions at which the identical nucleic acid base or amino acid residue occurs in both sequences to yield the number of matched positions, dividing the number of matched positions by the total number of positions in the window

of comparison and multiplying the results by 100 to yield the percentage of sequence identity. It would be understood that, when calculating sequence identity between a DNA sequence and an RNA sequence, T residues of the DNA sequence align with, and can be considered "identical" with, U residues of the RNA sequence. For purposes of determining "percent complementarity" of first and second polynucleotides, one can obtain this by determining (i) the percent identity between the first polynucleotide and the complement sequence of the second polynucleotide (or vice versa), for example, and/or (ii) the percentage of bases between the first and second polynucleotides that would create canonical Watson and Crick base pairs.

The Basic Local Alignment Search Tool (BLAST) algorithm, which is available online at the National Center for Biotechnology Information (NCBI) website, may be used, for example, to measure percent identity between or among two or more of the polynucleotide sequences (BLASTN algorithm) or polypeptide sequences (BLASTP algorithm) disclosed herein. Alternatively, percent identity between sequences may be performed using a Clustal algorithm (e.g., ClustalW, ClustalV, or Clustal-Omega). For multiple alignments using a Clustal method of alignment, the default values may correspond to GAP PENALTY=10 and GAP LENGTH PENALTY=10. Default parameters for pairwise alignments and calculation of percent identity of protein sequences using a Clustal method may be KTUPLE=1, GAP PENALTY=3, WINDOW=5 and DIAGONALS SAVED=5. For nucleic acids, these parameters may be KTUPLE=2, GAP PENALTY=5, WINDOW=4 and DIAGONALS SAVED=4. Alternatively still, percent identity between sequences may be performed using an EMBOSS algorithm (e.g., needle) with parameters such as GAP OPEN=10, GAP EXTEND=0.5, END GAP PENALTY=false, END GAP OPEN=10, END GAP EXTEND=0.5 using a BLOSUM matrix (e.g., BLOSUM62).

Various polypeptide amino acid sequences and polynucleotide sequences are disclosed herein as features of certain embodiments. Variants of these sequences that are at least about 70-85%, 85-90%, or 90%-95% identical to the sequences disclosed herein can be used. Alternatively, a variant amino acid sequence or polynucleotide sequence can have at least 70%, 71%, 72%, 73%, 74%, 75%, 76%, 77%, 78%, 79%, 80%, 81%, 82%, 83%, 84%, 85%, 86%, 87%, 88%, 89%, 90%, 91%, 92%, 93%, 94%, 95%, 96%, 97%, 98% or 99% identity with a sequence disclosed herein. The variant amino acid sequence or polynucleotide sequence may have the same function/activity

of the disclosed sequence, or at least about 80%, 81%, 82%, 83%, 84%, 85%, 86%, 87%, 88%, 89%, 90%, 91%, 92%, 93%, 94%, 95%, 96%, 97%, 98%, or 99% of the function/activity of the disclosed sequence. Any polypeptide amino acid sequence disclosed herein not beginning with a methionine can typically further comprise at least a start-methionine at the N-terminus of the amino acid sequence. Any polypeptide amino acid sequence disclosed herein beginning with a methionine can optionally be considered without this methionine residue (i.e., a polypeptide sequence can be referred to in reference to the position-2 residue to the C-terminal residue of the sequence).

The term "isolated" as used herein refers to any cellular component (e.g. polynucleotide, polypeptide), or cellulose material that has been completely or partially purified. In some instances, the isolated polynucleotide, polypeptide, or cellulose material is part of a greater composition, buffer system, or reagent mix. For example, the isolated polynucleotide or polypeptide molecule can be comprised within a cell or organism in a heterologous manner. Such a cell or organism containing heterologous components and/or one or more genetic deletions does not occur in nature. Another example is an isolated cellodextrin phosphorylase enzyme or reaction. Cellulose compositions herein and the enzymes and reactions used to produce these compositions are synthetic/man-made, and/or exhibit properties not believed to naturally occur.

An "aqueous composition" herein has a liquid component that comprises at least about 10 wt% water, for example. Examples of aqueous compositions include mixtures, solutions, dispersions (e.g., colloidal dispersions), suspensions and emulsions, for example. An aqueous composition in certain embodiments can comprise an insoluble cellulose as disclosed herein, in which case the aqueous composition can optionally be characterized as a solid-in-liquid composition, given the cellulose insolubility.

As used herein, the term "colloidal dispersion" refers to a heterogeneous system having a dispersed phase and a dispersion medium, i.e., microscopically dispersed insoluble particles are suspended throughout another substance (e.g., an aqueous composition such as water or aqueous solution). An example of a colloidal dispersion herein is a hydrocolloid. All, or a portion of, the particles of a colloidal dispersion such as a hydrocolloid can comprise cellulose of the present disclosure. The terms

"dispersant" and "dispersion agent" are used interchangeably herein to refer to a material that promotes the formation and/or stabilization of a dispersion.

The terms "hydrocolloid" and "hydrogel" are used interchangeably herein. A hydrocolloid refers to a colloid system in which water or an aqueous solution is the dispersion medium.

The term "aqueous solution" herein refers to a solution in which the solvent comprises water. An aqueous solution can serve as a dispersant in certain aspects herein. Cellulose in certain embodiments can be dispersed or mixed within an aqueous solution.

The term "viscosity" as used herein refers to the measure of the extent to which a fluid or an aqueous composition such as a hydrocolloid resists a force tending to cause it to flow. Various units of viscosity that can be used herein include centipoise (cPs) and Pascal-second (Pa·s). A centipoise is one one-hundredth of a poise; one poise is equal to 0.100 $kg \cdot m^{-1} \cdot s^{-1}$., or 1 mPa·s. Thus, the terms "viscosity modifier", "viscosity-modifying agent" and the like as used herein refer to anything that can alter/modify the viscosity of a fluid or aqueous composition.

The term "shear thinning behavior" as used herein refers to a decrease in the viscosity of an aqueous composition as shear rate increases. "Shear rate" herein refers to the rate at which a progressive shearing deformation is applied to an aqueous composition. A shearing deformation can be applied rotationally, for example.

The term "contacting" as used herein with respect to methods of increasing the viscosity of an aqueous composition refers to any action that results in bringing together an aqueous composition with cellulose as presently disclosed. Contacting can be performed by any means known in the art, such as mixing, shaking, or homogenization, for example.

"DMSO" as used herein refers to dimethyl sulfoxide, which has the formula $(CH_3)_2SO$.

"DMAc" as used herein refers to *N,N*-dimethylacetamide, which has the formula $CH_3CON(CH_3)_2$.

The terms "mercerization", "mercerization process" and the like are used interchangeably herein to refer to a process in which cellulose material is treated under caustic alkali conditions, typically comprising sodium hydroxide. Cellulose as disclosed in certain embodiments herein has not been mercerized.

The terms “derivatization”, “derivatization process” and the like are used interchangeably herein to refer to a process in which cellulose material is treated under conditions leading to the substitution of one or more hydrogens of cellulose -OH groups with a different moiety/functional group (e.g., carboxymethyl group). Cellulose as disclosed in certain embodiments herein has not been derivatized.

The term “film” as used herein refers to a thin, visually continuous material. A film can be comprised as a thin layer or coating on a material, or can be alone (e.g., not attached to a material surface). A “coating” as used herein refers to a thin layer covering a surface of a material.

The term “uniform thickness” as used to characterize a film or coating herein can refer to a contiguous area that (i) is at least 20% of the total film/coating area, and (ii) has a standard deviation of thickness of less than about 50 nm, for example.

A film or coating herein can be characterized as being of “low permeability” to a particular substance if the film/coating permeability to the substance is below a threshold value commonly assigned in the art of interest. To illustrate, the threshold value for styrene permeability in the SMC (super-multicoated) release film field is $200x10^{-9}$ g $cm/cm^2/h$, such as measured using the method described in *American Institute of Chemical Engineer, 53rd National Meeting, Preprint No.32d* (Bixler and Michaels, 1964). A film or coating can be characterized as being “impermeable” to a particular substance if it does not permit passage of the substance over an extended period of time (e.g., one or more days).

The terms “fabric”, “textile”, “cloth” and the like are used interchangeably herein to refer to a woven material having a network of natural and/or artificial fibers. Such fibers can be thread or yarn, for example.

A “fabric care composition” herein is any composition suitable for treating fabric in some manner. Examples of such a composition include laundry detergents and fabric softeners.

The terms “heavy duty detergent”, “all-purpose detergent” and the like are used interchangeably herein to refer to a detergent useful for regular washing of white and/or colored textiles at any temperature. The terms “low duty detergent” or “fine fabric detergent” are used interchangeably herein to refer to a detergent useful for the care of delicate fabrics such as viscose, wool, silk, microfiber or other fabric requiring special

care. "Special care" can include conditions of using excess water, low agitation, and/or no bleach, for example.

A "detergent composition" herein typically comprises at least one surfactant (detergent compound) and/or at least one builder. A "surfactant" herein refers to a substance that tends to reduce the surface tension of a liquid in which the substance is dissolved. A surfactant may act as a detergent, wetting agent, emulsifier, foaming agent, and/or dispersant, for example.

The terms "anti-redeposition agent", "anti-soil redeposition agent", "anti-greying agent" and the like herein refer to agents that help keep soils from redepositing onto clothing in laundry wash water after these soils have been removed, therefore preventing greying/discoloration of laundry. Anti-redeposition agents can function by helping keep soil dispersed in wash water and/or by blocking attachment of soil onto fabric surfaces.

An "oral care composition" herein is any composition suitable for treating an soft or hard surface in the oral cavity such as dental (teeth) and/or gum surfaces.

The term "adsorption" herein refers to the adhesion of a compound to the surface of a material.

Development of new forms of cellulose is desirable given the potential utility thereof in various applications. The development of novel enzymatic processes may be a useful means for producing new types of cellulose material.

Embodiments of the present disclosure concern an enzymatic reaction comprising at least water, glucose-1-phosphate, cellodextrin, and a cellodextrin phosphorylase enzyme comprising an amino acid sequence that is at least 90% identical to SEQ ID NO:2 or SEQ ID NO:6, wherein the cellodextrin phosphorylase enzyme synthesizes cellulose. Significantly, such an enzymatic reaction is able to produce a low molecular weight, insoluble cellulose that has enhanced features under both dry and aqueous conditions, rendering such cellulose as having broad applicability.

An enzyme with cellodextrin phosphorylase activity suitable for use in an enzymatic reaction as presently disclosed can comprise an amino acid sequence that is at least 90% identical to SEQ ID NO:2 or SEQ ID NO:6. In some embodiments, such an enzyme can comprise, or consist of, an amino acid sequence that is 100% identical to, or at least 90%, 91%, 92%, 93%, 94%, 95%, 96%, 97%, 98%, or 99% identical to,

SEQ ID NO:2 or SEQ ID NO:6. Non-limiting examples of a cellodextrin phosphorylase enzyme comprising SEQ ID NO:2 include cellodextrin phosphorylase enzymes comprising, or consisting of, an amino acid sequence that is 100% identical to, or at least 90%, 91%, 92%, 93%, 94%, 95%, 96%, 97%, 98%, or 99% identical to, SEQ ID NO:4. Non-limiting examples of a cellodextrin phosphorylase enzyme comprising SEQ ID NO:6 include cellodextrin phosphorylase enzymes comprising, or consisting of, an amino acid sequence that is 100% identical to, or at least 90%, 91%, 92%, 93%, 94%, 95%, 96%, 97%, 98%, or 99% identical to, SEQ ID NO:8. A variant cellodextrin phosphorylase enzyme (e.g., between 90-99% amino acid identity with SEQ ID NO:2, 4, 6, or 8 reference sequence) should have some of (e.g., at least 30%, 40%, 50%, 60%, 70%, 80%, or 90% of), or all of, the enzymatic activity (refer to above definitions) of the corresponding non-variant reference sequence.

A polynucleotide sequence encoding SEQ ID NO:2 or SEQ ID NO:4 can optionally comprise a nucleotide sequence that is 100% identical to, or at least 70%, 71%, 72%, 73%, 74%, 75%, 76%, 77%, 78%, 79%, 80%, 81%, 82%, 83%, 84%, 85%, 86%, 87%, 88%, 89%, 90%, 91%, 92%, 93%, 94%, 95%, 96%, 97%, 98%, or 99% identical to, SEQ ID NO:1 or 3, respectively. A polynucleotide sequence encoding SEQ ID NO:6 or SEQ ID NO:8 can optionally comprise a nucleotide sequence that is 100% identical to, or at least 70%, 71%, 72%, 73%, 74%, 75%, 76%, 77%, 78%, 79%, 80%, 81%, 82%, 83%, 84%, 85%, 86%, 87%, 88%, 89%, 90%, 91%, 92%, 93%, 94%, 95%, 96%, 97%, 98%, or 99% identical to, SEQ ID NO:5 or 7, respectively.

Given that certain amino acids share similar structural and/or charge features with each other (i.e., conserved), one or more amino acids of a cellodextrin phosphorylase sequence herein (and/or other types of polypeptides herein) can be substituted with a conserved amino acid residue ("conservative amino acid substitution") as follows:

1. The following small aliphatic, nonpolar or slightly polar residues can substitute for each other: Ala (A), Ser (S), Thr (T), Pro (P), Gly (G);
2. The following polar, negatively charged residues and their amides can substitute for each other: Asp (D), Asn (N), Glu (E), Gln (Q);
3. The following polar, positively charged residues can substitute for each other: His (H), Arg (R), Lys (K);

4. The following aliphatic, nonpolar residues can substitute for each other: Ala (A), Leu (L), Ile (I), Val (V), Cys (C), Met (M); and
5. The following large aromatic residues can substitute for each other: Phe (F), Tyr (Y), Trp (W).

An enzyme with cellodextrin phosphorylase activity herein can be obtained from any microbial source, for example, such as a bacteria or fungus (e.g., yeast). Examples of suitable bacteria include *Vibrio* species and *Ruminococcus* species. Examples of suitable *Vibrio* species include *V. ruber*, *V. cholerae*, *V. adaptatus*, *V. alginolyticus*, *V. mimicus, V. parahaemolyticus*, *V. proteolyticus*, *and V. vulnificus*. Examples of suitable *Ruminococcus* species include *R. champanellensis*, *R. albus*, *R. bromii*, *R. flavefaciens*, *R. gnavus*, *R. lactaris*, *R. obeum*, and *R. torques*.

Examples of enzymes with cellodextrin phosphorylase activity herein can be any of the amino acid sequences disclosed herein and that further include 1-300 (or any integer there between [e.g., 10, 15, 20, 25, 30, 35, 40, 45, or 50]) residues on the N-terminus and/or C-terminus. Such additional residues may be a heterologous sequence such as an epitope tag (at either N- or C-terminus) (e.g., His tag such as a hexa histidine) or a heterologous signal peptide (at N-terminus), for example. In those embodiments in which a heterologous amino acid sequence is incorporated at the N-terminus, such a heterologous sequence can be adjacent to the original start-methionine of the cellodextrin phosphorylase, or can replace the original start methionine, for example. In the latter embodiment, a new start-methionine can be employed at the N-terminus of the added heterologous sequence.

An enzyme with cellodextrin phosphorylase activity as presently disclosed typically lacks an N-terminal signal peptide. However, an expression system for producing a cellodextrin phosphorylase enzyme can optionally employ an enzyme-encoding polynucleotide that further comprises sequence encoding an N-terminal signal peptide to direct extra-cellular secretion. The signal peptide in such embodiments is cleaved from the enzyme during the secretion process. Since it is believed that the cellodextrin phosphorylase enzymes disclosed herein (e.g., SEQ ID NO:2 and 6) are not associated with a signal peptide as natively expressed, any added signal peptide may be considered as heterologous to the enzyme. An example of a signal peptide useful herein is one from a bacterial (e.g., a *Bacillus* species such as *B. subtilis*) or fungal species. An example of a bacterial signal peptide is an aprE signal peptide, such as

one from *Bacillus* (e.g., *B. subtilis*, see Vogtentanz et al., *Protein Expr. Purif.* 55:40-52, which is incorporated herein by reference).

A cellodextrin phosphorylase enzyme in some embodiments does not occur in nature; for example, an enzyme herein is not believed to be one that is naturally secreted (i.e., mature form) from a microbe (from which the cellodextrin phosphorylase enzyme herein could possibly have been derived).

A cellodextrin phosphorylase enzyme herein can be prepared by fermentation of an appropriately engineered microbial strain, for example. Recombinant enzyme production by fermentation is well known in the art using microbial strains such as *E. coli*, *Bacillus* strains (e.g., *B. subtilis*), *Ralstonia eutropha*, *Pseudomonas fluorescens*, *Saccharomyces cerevisiae*, *Pichia pastoris*, *Hansenula polymorpha*, and species of *Aspergillus* (e.g., *A. awamori*) and *Trichoderma* (e.g., *T. reesei*) (e.g., see Adrio and Demain, *Biomolecules* 4:117-139, which is incorporated herein by reference).

A cellodextrin phosphorylase enzyme of the present disclosure may be used in any purification state (e.g., pure or non-pure). For example, a cellodextrin phosphorylase enzyme may be purified and/or isolated prior to its use. Examples of cellodextrin phosphorylase enzymes that are non-pure include those in the form of a cell lysate. A cell lysate or extract may be prepared from a bacteria (e.g., *E. coli*) used to heterologously express the enzyme. For example, the bacteria may be subjected to disruption using a French pressure cell. In alternative embodiments, bacteria may be homogenized with a homogenizer (e.g., APV, Rannie, Gaulin). A cellodextrin phosphorylase enzyme is typically soluble in these types of preparations. A bacterial cell lysate, extract, or homogenate herein may be used at about 0.15-0.3% (v/v) in an enzymatic reaction herein, if desired. In other embodiments, an enzyme with cellodextrin phosphorylase activity can be isolated after its expression. For example, the enzyme can be isolated using a binding/washing or binding/washing/elution approach (e.g., binding enzyme to a column of other fixed surface, followed by washing and optionally eluting enzyme off column or other fixed surface). An enzyme isolation approach can comprise binding a heterologous amino acid sequence-tagged cellodextrin phosphorylase enzyme in certain embodiments, wherein such binding is via the heterologous amino acid sequence tag (e.g., His tag). A cellodextrin phosphorylase enzyme can be isolated from a cell lysate or any other composition (e.g., medium into

which enzyme is optionally secreted), for example. In certain aspects, a cellodextrin phosphorylase preparation can lack glucose-1-phosphatase activity. A cellodextrin phosphorylase enzyme in some aspects can be immobilized (e.g., to a matrix) or expressed on cell surfaces. A cellodextrin phosphorylase enzyme can optionally be modified with polyethylene glycol (PEG), for instance.

Cellodextrin phosphorylase enzyme of the present disclosure can synthesize low molecular weight cellulose that is insoluble in aqueous compositions. For example, a cellodextrin phosphorylase as employed in an enzymatic reaction herein can produce low molecular weight, insoluble cellulose.

Cellulose produced by a cellodextrin phosphorylase enzyme in certain embodiments can have a DP_w or DP_n of about 10-1000. For example, DP_w or DP_n of cellulose herein can be about 10-500, 10-250, 10-100, 10-75, 10-50, 10-45, 10-40, 10-35, 10-30, 10-25, 15-50, 15-45, 15-40, 15-35, 15-30, or 15-25. DP_w or DP_n of cellulose in some aspects can be about, at least about, or less than about, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19, 20, 21, 22, 23, 24, 25, 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, 40, 41, 42, 43, 44, 45, 46, 47, 48, 49, or 50.

Cellulose produced by a cellodextrin phosphorylase enzyme in some aspects can have an M_w of about 1700-170000, 1700-86000, 1700-43000, 1700-17000, 1700-13000, 1700-8500, 1700-6800, 1700-5100, 2550-5100, or 2550-4250. M_w can be about, at least about, or less than about, 1700, 1900, 2100, 2300, 2500, 2700, 2900, 3100, 3300, 3500, 3700, 3900, 4100, 4300, 4500, 4700, 4900, or 5100 in some aspects.

About 100% of the glycosidic linkages of cellulose produced by a cellodextrin phosphorylase enzyme herein are beta-1,4 linkages, for example. Cellulose in other aspects can have a glycosidic linkage profile of at least about 90%, 91%, 92%, 93%, 94%, 95%, 96%, 97%, 98%, or 99% beta-1,4 linkages. Accordingly, cellulose enzymatically produced herein can have, for example, less than 10%, 9%, 8%, 7%, 6%, 5%, 4%, 3%, 2%, or 1% of glycosidic linkages that are other than beta-1,4.

The backbone of a cellulose synthesized by cellodextrin phosphorylase enzyme herein can be linear/unbranched. Alternatively, there can be branches in the cellulose. Thus, in certain embodiments, cellulose can have no branch points or less than about 5%, 4%, 3%, 2%, or 1% branch points as a percent of the glycosidic linkages in the polymer.

Cellulose produced by a cellodextrin phosphorylase enzyme in some aspects herein can have a cellulose II crystal structure. For example, cellulose herein can comprise about 100% cellulose, by weight, that is of a cellulose II crystal structure. As other examples, cellulose can comprise at least about 80%, 81%, 82%, 83%, 84%, 85%, 86%, 87%, 88%, 89%, 90%, 91%, 92%, 93%, 94%, 95%, 96%, 97%, 98%, or 99% cellulose, by weight, that is of a cellulose II crystal structure. Cellulose in some aspects can comprise less than about 20%, 19%, 18%, 17%, 16%, 15%, 14%, 13%, 12%, 11%, 10%, 9%, 8%, 7%, 6%, 5%, 4%, 3%, 2%, or 1% cellulose material, by weight, that is of a cellulose I, III, and/or IV crystal structure. Cellulose II crystal structure has been described by Kolpak and Blackwell (*Macromolecules* 9:273-278) and Kroon-Batenburg and Kroon (*Glycoconjugate J.* 14:677-690), for example, both of which are incorporated herein by reference. The dominant hydrogen bonds characterizing a cellulose II structure are O2-H---O6, O6-H---O6 and O2-H---O2, whereas cellulose I has O2-H---O6 as a dominant hydrogen bond. The structure of cellulose II comprises chain folding and is difficult to unravel.

Cellulose is produced by a cellodextrin phosphorylase enzyme of the present disclosure directly as cellulose II. In contrast to cellulose as presently disclosed, cellulose produced in nature (e.g., in plants) typically is of a cellulose I structure and generally requires mercerization and/or other chemical treatments (e.g., derivatization followed by un-derivatization, formation of regenerated cellulose) to convert it into cellulose II. Cellulose in certain embodiments herein is in the cellulose II crystal state under both aqueous and dry conditions.

Cellulose as produced herein is insoluble in aqueous solvents such as water. However, it can be soluble in solvents comprising dimethyl sulfoxide (DMSO) and/or *N,N*-dimethylacetamide (DMAc). Examples of such solvents include DMSO or DMAc alone or further comprising lithium chloride (LiCl) (e.g., DMSO/LiCl and DMAc/LiCl). A DMSO/LiCl solvent or DMSO/LiCl solvent herein can comprise about 0.5, 1, 2, 3, 4, 5, 6, 7, 8, 9, or 10 wt% LiCl, for example, or can be LiCl-saturated. The concentration of cellulose herein can be at about 0.1-30 wt%, 0.1-20 wt%, 0.1-10 wt%, or 0.1-5 wt%, for example, or can be at about, or at least about, 0.1, 0.3, 0.5, 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 15, 20, 25, or 30 wt% in a non-aqueous solvent such as one comprising DMSO and/or DMAc. DMSO- and DMAc-comprising solvents herein do not further comprise an acid in certain aspects. Cellulose herein can be dissolved in any of the foregoing DMSO-

and DMAc-based solvents at a relatively low temperature, such as at 15-30 °C, 20-30 °C, or 20-25 °C (e.g., room temperature), for example. In preferred embodiments, heat does not need to be applied to dissolve the cellulose.

Enzymatic reactions of the present disclosure comprise cellodextrin. Examples of cellodextrin suitable for use in an enzymatic reaction herein include cellobiose (DP2), cellotriose (DP3), cellotetraose (DP4), cellopentaose (DP5), and cellohexaose (DP6). Cellobiose is used as a cellodextrin in certain aspects. Other examples of cellodextrin suitable herein include glucose polymers of 7 or more beta-1,4-linked glucose monomers resulting from the breakdown (e.g., enzymatic breakdown) of cellulose. One or more (e.g., a mixture of 2, 3, 4 or more) of the above types of cellodextrin can be employed in some embodiments.

The temperature of an enzymatic reaction herein comprising a cellodextrin phosphorylase enzyme can be controlled, if desired. In certain embodiments, the temperature is between about 5 °C to about 50 °C. The temperature in certain other embodiments is between about 20 °C to about 40 °C. In still other embodiments, the temperature may be about 20, 21, 22, 23, 24, 25, 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, or 40 °C. The temperature of an enzymatic reaction can be maintained using various means known in the art. For example, the temperature can be maintained by placing the vessel containing the reaction in an air or water bath incubator set at the desired temperature.

The pH of an enzymatic reaction in certain embodiments herein can be between about 5.0 to about 9.0. Alternatively, the pH can be about 5.0, 5.5, 6.0, 6.5, 7.0, 7.5, 8.0, 8.5, or 9.0. The pH can be adjusted or controlled by the addition or incorporation of a suitable buffer, including but not limited to: phosphate, tris, citrate, or a combination thereof. Buffer concentration in the enzymatic reaction can be from 0 mM to about 100 mM, or about 10, 25, 50, or 75 mM, for example.

The initial concentration of glucose-1-phosphate (G1P) in the presently disclosed cellodextrin phosphorylase reaction can be about, or at least about, 1 to 100 mM, for example. Other G1P initial concentrations can be, for example, about, or at least about, 1, 5, 10, 20, 30, 40, 50, 60, 70, 80, 90, or 100 mM, or about 10-50 mM. The initial concentration of cellodextrin (e.g., cellobiose) in the presently disclosed cellodextrin

phosphorylase reaction can be about 1 to 50 mM, for example. Other cellodextrin initial concentrations can be, for example, about 1, 5, 10, 15, 20, 25, 30, 35, 40, 45, or 50 mM, or about 5-10 mM. "Initial concentration" of a substrate such as G1P or cellodextrin refers to the substrate concentration in an enzymatic reaction just after all the reaction components have been added (at least water, G1P, cellodextrin, cellodextrin phosphorylase enzyme).

The activity of a cellodextrin phosphorylase enzyme herein can be about 1 to 30 units per mg of enzyme protein in some embodiments. Enzyme activity can be about 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19, 20, 21, 22, 23, 24, 25, 26, 27, 28, 29, 30, 10-20, or 15-20 units per mg of enzyme protein, for example. Cellodextrin phosphorylase enzyme activity can be determined using any method known in the art. A unit of cellodextrin phosphorylase activity can refer to, for example, the amount of enzyme that releases 1 micro-mol of inorganic phosphorus (released from cellobiose) per minute under the following conditions: ~10 mM G1P, ~5 mM cellobiose, ~25 mM Tris-HCl buffer, ~pH 7.0, held at ~37 °C, optionally for ~10 minutes. Inorganic phosphate release from cellobiose can be gauged using a reagent or kit designed to detect free phosphate (e.g., PiBlue™ Phosphate Assay Kit, BioAssay Systems, Hayward, CA).

The amount of a cellodextrin phosphorylase enzyme comprised in an enzymatic reaction in some aspects can be about 0.1-2.0 or 0.5-1.0 units/mL. For example, at least about 0.2, 0.4, 0.6, 0.7, 0.8, 0.9, 1.0, 1.2, 1.4, 1.6, 1.8, or 2.0 units/mL of enzyme can be employed in a reaction.

Embodiments of the present disclosure also concern a method for producing cellulose, comprising:

a) contacting at least water, glucose-1-phosphate (G1P), cellodextrin, and a cellodextrin phosphorylase enzyme comprising an amino acid sequence that is at least 90% identical to SEQ ID NO:2 or SEQ ID NO:6, wherein insoluble cellulose is produced; and

b) optionally, isolating the cellulose produced in step (a).

The contacting step in a method herein of producing cellulose can optionally be characterized as providing an enzymatic reaction comprising water, glucose-1-phosphate, cellodextrin, and a cellodextrin phosphorylase enzyme of the present

disclosure. The contacting step in a cellulose production method herein can be performed in any number of ways. For example, the desired amount of G1P and/or cellodextrin (e.g., cellobiose) can first be dissolved in water (optionally, other components may also be added at this stage of preparation, such as buffer components), followed by addition of one or more cellodextrin phosphorylase enzymes. The reaction may be kept still, or agitated via stirring or orbital shaking, for example. The reaction can be, and typically is, cell-free.

The enzymatic reaction of a cellulose production method can be contained within any vessel suitable for applying one or more of the reaction conditions disclosed herein. For example, a stainless steel, plastic, or glass vessel or container of a size suitable to contain a particular reaction can be employed. Such a vessel can optionally be equipped with a stirring device.

Completion of an enzymatic reaction of a cellulose production method in certain embodiments can be determined visually (e.g., no more accumulation of insoluble cellulose) and/or by measuring the amount of substrate (G1P and/or cellodextrin) left in the reaction (e.g., no more decrease in substrate levels over time). Typically, a reaction of the disclosed method can take about 12, 18, 24, 30, 36, 48, 60, 72, 84, or 96 hours to complete, for example. Reaction time may depend, for example, on certain parameters such as the amount of substrate and/or cellodextrin phosphorylase enzyme employed.

Insoluble cellulose produced in the disclosed method may optionally be isolated. For example, insoluble cellulose may be separated by centrifugation or filtration. In doing so, the cellulose is separated from the reaction solution, which can comprise water, residual substrate(s) and reaction byproducts.

Insoluble cellulose produced in a contacting step of a cellulose production method herein can have any of the features disclosed herein. For example, any of the features of water-insolubility, DP_w (e.g., DP_w of 10-30) and/or M_w, glycosidic linkage profile, backbone structure (e.g., linearity), cellulose II structural content, and/or solubility in certain non-aqueous compositions as disclosed elsewhere herein can characterize cellulose produced in step (a).

Insoluble cellulose produced in a contacting step of a cellulose production method in some aspects can have a cellulose II crystal structure (i.e., the cellulose is enzymatically synthesized directly as cellulose II). In contrast to cellulose as presently

disclosed, cellulose produced in nature (e.g., in plants) typically is of a cellulose I structure and generally requires mercerization and/or other chemical treatments (e.g., derivatization followed by un-derivatization, formation of regenerated cellulose) to convert it into cellulose II. Cellulose in certain embodiments herein is in the cellulose II crystal state under both aqueous and dry conditions.

Any features disclosed herein characterizing enzymatic reaction embodiments can be employed in performing a contacting step of a cellulose production method. For example, any of the features of cellodextrin phosphorylase enzyme amino acid sequence and source, substrate levels, temperature, pH and buffer levels, and/or enzyme activity/amount as disclosed elsewhere herein can characterize a reaction performed in the contacting step.

The contacting step of a cellulose production method in some aspects can comprise cellobiose as a cellodextrin. Other examples of cellodextrin suitable for use in an enzymatic reaction herein include cellotriose, cellotetraose, cellopentaose, and cellohexaose. Still other examples of cellodextrin suitable herein include glucose polymers of 7 or more beta-1,4-linked glucose monomers resulting from the breakdown (e.g., enzymatic breakdown) of cellulose. One or more (e.g., a mixture of 2, 3, 4 or more) of the above types of cellodextrin can be employed in some embodiments.

Glucose-1-phosphate (G1P) provided in a contacting step of a cellulose production method can be providing directly via addition of isolated G1P (e.g., G1P obtained from a commercial source), for example. Alternatively, G1P can be provided in the contacting step by providing at least a second reaction, wherein the products of the second reaction comprise G1P (i.e., the second reaction produces G1P as a product). A "second reaction" refers to a reaction that is in addition to the cellodextrin phosphorylase reaction performed in the contacting step (can optionally be denoted as a "first reaction"), and which provides G1P substrate for the cellodextrin phosphorylase reaction. A second reaction can optionally be characterized as employing a "G1P-producing enzyme" such as a starch phosphorylase, sucrose phosphorylase, or cellodextrin phosphorylase (when catalyzing cellulose hydrolysis).

A second reaction for providing G1P in some aspects can be provided in the same vessel in which a cellodextrin phosphorylase enzymatic reaction is performed.

Alternatively, a second reaction can be performed outside of (separate from) the vessel in which a cellodextrin phosphorylase enzymatic reaction is performed. A second reaction can be performed before and/or continuously with a cellodextrin phosphorylase enzymatic reaction of a cellulose production method.

A second reaction in some embodiments can comprise contacting water, inorganic phosphate, starch, a starch phosphorylase, and optionally a starch debranching enzyme such as a pullulanase and/or an isoamylase. This type of second reaction can optionally be characterized as a starch phosphorylase reaction. Starch phosphorylases (EC 2.4.1.1) suitable for use herein include those disclosed in U.S. Patent Appl. Publ. No. 2002/0133849 and Tiwari and Kumar (*Biotechnol. Mol. Biol. Rev.* 7:69-83), for example, which are incorporated herein by reference. A starch phosphorylase in some aspects can be a plant, microbial (e.g., bacterial), or fungal (e.g., yeast) starch phosphorylase. Pullulanases (EC 3.2.1.41) suitable for use herein include those disclosed in U.S. Patent Nos. 8354101, 7906306, 7449320, and 7399623, for example, which are incorporated herein by reference. A pullulanase in some aspects can be a plant, microbial (e.g., bacterial), or fungal (e.g., yeast) pullulanase. Isoamylases (EC 3.2.1.68) suitable for use herein include those disclosed in U.S. Patent Nos. 5352602, 5811277, 7615365 and 8735105, for example, which are incorporated herein by reference. An isoamylase in some aspects can be a plant, microbial (e.g., bacterial), or fungal (e.g., yeast) isoamylase.

A second reaction in some embodiments can comprise contacting water, inorganic phosphate, sucrose, and a sucrose phosphorylase enzyme. This type of second reaction can optionally be characterized as a sucrose phosphorylase reaction. Sucrose phosphorylases (EC 2.4.1.7) suitable for use herein include those disclosed in U.S. Patent Nos. 5716837, 7229801 and 7968309, for example, which are incorporated herein by reference. A sucrose phosphorylase in some aspects can be a plant, microbial (e.g., bacterial), or fungal (e.g., yeast) sucrose phosphorylase.

A second reaction in some embodiments can comprise contacting water, inorganic phosphate, cellulosic biomass (cellulose-comprising biomass such as lignocellulosic biomass), an endoglucanase, a cellodextrin phosphorylase, and optionally, a lytic polysaccharide monooxygenase and/or a cellobiohydrolase. Endoglucanases (e.g., cellulase, beta-1,4-glucanase) suitable for use herein include those disclosed in U.S. Patent Nos. 4435307, 5776757 and 7604974, for example,

which are incorporated herein by reference. An endoglucanase (e.g., cellulase) in some aspects can be a plant, microbial (e.g., bacterial), or fungal (e.g., yeast) endoglucanase. A cellodextrin phosphorylase suitable for use herein can be any cellodextrin phosphorylase as presently disclosed, or as disclosed in U.S. Patent No. 8889379, or U.S. Patent Appl. Publ. Nos. 2014/0087435, 2014/0057323, and 2013/0059340, for example, which are incorporated herein by reference. This type of second reaction (i.e., endoglucanase + cellodextrin phosphorylase) can typically be performed separately from a cellodextrin phosphorylase enzymatic reaction of a cellulose production method herein. Lytic polysaccharide monooxygenases suitable for use herein include those disclosed in Isaksen et al. (*J. Biol. Chem.* 289:2632-2642) and Eibinger et al. (*J. Biol. Chem.,* Oct 31, 2014, pii: jbc.M114.602227. [Epub ahead of print]), for example, which are incorporated herein by reference.

Embodiments of the present disclosure further concern a composition comprising an enzyme comprising an amino acid sequence that is at least 90% identical to SEQ ID NO:2, wherein the enzyme has cellodextrin phosphorylase activity. Significantly, such an enzyme is able to produce a low molecular weight, insoluble cellulose that has enhanced features under both dry and aqueous conditions, rendering such cellulose as having broad applicability. A non-limiting example of a composition comprising a cellodextrin phosphorylase enzyme having an amino acid sequence that is at least 90% identical to SEQ ID NO:2 is an enzymatic reaction, such as one also comprising at least water, glucose-1-phosphate, and one or more cellodextrins.

An enzyme herein with cellodextrin phosphorylase activity can comprise an amino acid sequence that is at least 90% identical to SEQ ID NO:2. In other embodiments, such an enzyme can comprise, or consist of, an amino acid sequence that is 100% identical to, or at least 90%, 91%, 92%, 93%, 94%, 95%, 96%, 97%, 98%, or 99% identical to, SEQ ID NO:2. Non-limiting examples of a cellodextrin phosphorylase enzyme comprising SEQ ID NO:2 include cellodextrin phosphorylase enzymes comprising, or consisting of, an amino acid sequence that is 100% identical to, or at least 90%, 91%, 92%, 93%, 94%, 95%, 96%, 97%, 98%, or 99% identical to, SEQ ID NO:4. A variant cellodextrin phosphorylase enzyme (e.g., between 90-99% amino acid identity with SEQ ID NO:2 or 4 reference sequence) should have some of (e.g., at

least 30%, 40%, 50%, 60%, 70%, 80%, or 90% of), or all of, the enzymatic activity (refer to above definitions) of the corresponding non-variant reference sequence.

An enzyme with cellodextrin phosphorylase activity of the present disclosure can, optionally, synthesize cellulose in a reaction comprising water, glucose-1-phosphate, and cellodextrin. Cellulose produced in such a reaction can be insoluble (water-insoluble) and have a weight-average degree of polymerization (DP_w) of about 10 to about 30.

Certain aspects herein concern a polynucleotide sequence comprising a nucleotide sequence encoding a cellodextrin phosphorylase comprising an amino acid sequence that is at least 90% identical to SEQ ID NO:2. Any such amino acid sequence as disclosed herein, for example, can be encoded by the nucleotide sequence. The nucleotide sequence may optionally be in operable linkage with a promoter sequence (e.g., heterologous promoter). Some embodiments include, for example, a polynucleotide (e.g., vector or construct) comprising at least one open reading frame encoding a cellodextrin phosphorylase comprising an amino acid sequence that is at least 90% identical to SEQ ID NO:2. Such a coding region can optionally be operably linked to a promoter sequence (e.g., heterologous promoter) suitable for expression in a cell (e.g., bacteria cell; eukaryotic cell such as a yeast, insect, or mammalian cell) or in an *in vitro* protein expression system, for example. Examples of a vector or construct include circular (e.g., plasmid) and non-circular (e.g., linear DNA such as an amplified DNA sequence) polynucleotide molecules.

Certain embodiments herein concern a method of producing a cellodextrin phosphorylase comprising an amino acid sequence that is at least 90% identical to SEQ ID NO:2. This method can comprise the steps of: providing a polynucleotide sequence having a nucleotide sequence encoding a cellodextrin phosphorylase comprising an amino acid sequence that is at least 90% identical to SEQ ID NO:2 (e.g., any such amino acid sequence as disclosed herein), and expressing the cellodextrin phosphorylase from the polynucleotide sequence, thereby producing the cellodextrin phosphorylase. The expression step in such a method can optionally be performed in a cell (e.g., bacteria cell such as *E. coli*; eukaryotic cell such as a yeast [e.g., *S. cerevisiae*], insect, or mammalian cell). Alternatively, expression of can be performed in an *in vitro* protein expression system (e.g., cell-free protein expression systems such as those employing rabbit reticulocyte lysate or wheat germ extract). Also, cellodextrin

phosphorylase produced in the expression step can optionally be isolated. Such isolation can be performed in a manner that produces a composition having any of the features disclosed herein (e.g., purity, pH, buffer, and/or salt level), for example.

Embodiments of the present disclosure further concern a composition comprising cellulose, wherein the cellulose:

(i) has a weight-average degree of polymerization (DP_w) of about 10 to about 1000,

(ii) has a cellulose II crystal structure, and

(iii) is insoluble in an aqueous composition.

Significantly, such low molecular weight, insoluble cellulose has broad utility, owing to its having enhanced features under both dry and aqueous conditions as further disclosed herein.

Cellulose of a composition as presently disclosed is of low molecular weight cellulose and water-insoluble.

Cellulose in certain embodiments can have a DP_w or DP_n of about 10-1000. For example, DP_w or DP_n of cellulose herein can be about 10-500, 10-250, 10-100, 10-75, 10-50, 10-45, 10-40, 10-35, 10-30, 10-25, 15-50, 15-45, 15-40, 15-35, 15-30, or 15-25. DP_w or DP_n of cellulose in some aspects can be about, or at least about, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19, 20, 21, 22, 23, 24, 25, 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, 40, 41, 42, 43, 44, 45, 46, 47, 48, 49, or 50.

In some aspects herein, cellulose can have an M_w of about 1700-170000, 1700-86000, 1700-43000, 1700-17000, 1700-13000, 1700-8500, 1700-6800, 1700-5100, 2550-5100, or 2550-4250. M_w can be about, or at least about, 1700, 1900, 2100, 2300, 2500, 2700, 2900, 3100, 3300, 3500, 3700, 3900, 4100, 4300, 4500, 4700, 4900, or 5100 in some examples.

About 100% of the glycosidic linkages of cellulose as presently disclosed are beta-1,4 linkages, for example. Cellulose in other aspects can have a glycosidic linkage profile of at least about 90%, 91%, 92%, 93%, 94%, 95%, 96%, 97%, 98%, or 99% beta-1,4 linkages. Accordingly, cellulose herein can have, for example, less than 10%, 9%, 8%, 7%, 6%, 5%, 4%, 3%, 2%, or 1% of glycosidic linkages that are other than beta-1,4.

The backbone of cellulose disclosed herein can be linear/unbranched. Alternatively, there can be branches in the cellulose. Thus, in certain embodiments, cellulose can have no branch points or less than about 5%, 4%, 3%, 2%, or 1% branch points as a percent of the glycosidic linkages in the polymer.

Cellulose as disclosed herein can have a cellulose II crystal structure. For example, cellulose herein can comprise about 100% cellulose, by weight, that is of a cellulose II crystal structure. As other examples, cellulose can comprise at least about 80%, 81%, 82%, 83%, 84%, 85%, 86%, 87%, 88%, 89%, 90%, 91%, 92%, 93%, 94%, 95%, 96%, 97%, 98%, or 99% cellulose, by weight, that is of a cellulose II crystal structure. Cellulose in some aspects can comprise less than about 20%, 19%, 18%, 17%, 16%, 15%, 14%, 13%, 12%, 11%, 10%, 9%, 8%, 7%, 6%, 5%, 4%, 3%, 2%, or 1% cellulose material, by weight, that is of a cellulose I, III, and/or IV crystal structure. Cellulose II crystal structure has been described by Kolpak and Blackwell (*Macromolecules* 9:273-278) and Kroon-Batenburg and Kroon (*Glycoconjugate J.* 14:677-690), for example, both of which are incorporated herein by reference. The dominant hydrogen bonds characterizing a cellulose II structure are O2-H---O6, O6-H---O6 and O2-H---O2, whereas cellulose I has O2-H---O6 as a dominant hydrogen bond. The structure of cellulose II comprises chain folding and is difficult to unravel.

Cellulose herein can be characterized as being isolated, for example. Compositions comprising cellulose as presently disclosed are not believed to occur in nature.

Cellulose as disclosed herein can optionally be characterized as having a flake or flake-like shape at nanometer scale. Flake or flake-like shapes formed by the cellulose have nano-size dimensions; such shapes can appear as flat, thin pieces of material when using appropriate microscopic techniques such as disclosed in the present Examples. In other aspects, cellulose herein is not, nor has been, derivatized. Thus, cellulose as disclosed herein does not comprise added functional groups such as ether groups (e.g., carboxymethyl groups) or ester groups (e.g., acetate groups).

Cellulose of a composition as presently disclosed herein can be a product of a cellodextrin phosphorylase enzyme comprising, or consisting of, an amino acid sequence that is at least 90% identical to SEQ ID NO:2 or SEQ ID NO:6. In other embodiments, cellulose can be a product of a cellodextrin phosphorylase enzyme that

comprises, or consists of, an amino acid sequence that is 100% identical to, or at least 90%, 91%, 92%, 93%, 94%, 95%, 96%, 97%, 98%, or 99% identical to, SEQ ID NO:2 or SEQ ID NO:6. Non-limiting examples of a cellodextrin phosphorylase enzyme comprising SEQ ID NO:2 include cellodextrin phosphorylase enzymes comprising, or consisting of, an amino acid sequence that is 100% identical to, or at least 90%, 91%, 92%, 93%, 94%, 95%, 96%, 97%, 98%, or 99% identical to, SEQ ID NO:4. Non-limiting examples of a cellodextrin phosphorylase enzyme comprising SEQ ID NO:6 include cellodextrin phosphorylase enzymes comprising, or consisting of, an amino acid sequence that is 100% identical to, or at least 90%, 91%, 92%, 93%, 94%, 95%, 96%, 97%, 98%, or 99% identical to, SEQ ID NO:8. A variant cellodextrin phosphorylase enzyme (e.g., between 90-99% amino acid identity with SEQ ID NO:2, 4, 6, or 8 reference sequence) should have some of (e.g., at least 30%, 40%, 50%, 60%, 70%, 80%, or 90% of), or all of, the enzymatic activity (refer to above definitions) of the corresponding non-variant reference sequence. Production of cellulose using a cellodextrin phosphorylase enzyme can be accomplished with an enzymatic reaction as disclosed herein, for example.

Cellulose as produced by a cellodextrin phosphorylase enzyme of the present disclosure can have a cellulose II crystal structure; such cellulose has not been subjected to a mercerization or derivatization process. Cellulose herein as it exists immediately or shortly after (e.g., less than about .5, 1, 5, 10, 15, 30, 60, 90, or 120 minutes) its enzymatic synthesis by a cellodextrin phosphorylase enzyme can comprise cellulose in the cellulose II crystal state. In contrast to cellulose as presently disclosed, cellulose produced in nature (e.g., in plants) typically is of a cellulose I structure and generally requires mercerization and/or other chemical treatments (e.g., derivatization followed by un-derivatization, formation of regenerated cellulose) to convert it into cellulose II. Cellulose in certain embodiments herein comprises cellulose in the cellulose II crystal state under both aqueous and dry conditions.

Cellulose of a composition as presently disclosed is insoluble in aqueous solvents such as water. In contrast, it can be soluble in certain non-aqueous solvents such as those comprising dimethyl sulfoxide (DMSO) and/or *N,N*-dimethylacetamide (DMAc). Examples of such solvents include DMSO or DMAc alone or further comprising lithium chloride (LiCl) (e.g., DMSO/LiCl and DMAc/LiCl). A DMSO/LiCl

solvent or DMSO/LiCl solvent herein can comprise about 0.5, 1, 2, 3, 4, 5, 6, 7, 8, 9, or 10 wt% LiCl, for example, or can be LiCl-saturated. The concentration of cellulose herein can be at about 0.1-30 wt%, 0.1-20 wt%, 0.1-10 wt%, or 0.1-5 wt%, for example, or can be at about, or at least about, 0.1, 0.3, 0.5, 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 15, 20, 25, or 30 wt% in a non-aqueous solvent such as one comprising DMSO and/or DMAc. DMSO- and DMAc-comprising solvents herein do not further comprise an acid in certain aspects. Cellulose herein can be dissolved in any of the foregoing DMSO- and DMAc-based solvents at a relatively low temperature, such as at 15-30 °C, 20-30 °C, or 20-25 °C (e.g., room temperature), for example. In preferred embodiments, heat does not need to be applied to dissolve the cellulose.

A composition comprising a cellulose herein can be non-aqueous (e.g., a dry composition). Examples of such embodiments include films/coatings, powders, granules, microcapsules, flakes, or any other form of particulate matter. Other examples include larger compositions such as pellets, bars, kernels, beads, tablets, sticks, or other agglomerates. A non-aqueous or dry composition herein typically has less than 3, 2, 1, 0.5, or 0.1 wt% water comprised therein. The amount of cellulose herein in a non-aqueous or dry composition can be about, or at least about, 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19, 20, 21, 22, 23, 24, 25, 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, 40, 41, 42, 43, 44, 45, 46, 47, 48, 49, 50, 51, 52, 53, 55, 56, 57, 58, 59, 60, 61, 62, 63, 64, 65, 66, 67, 68, 69, 70, 71, 72, 73, 74, 75, 76, 77, 78, 79, 80, 81, 82, 83, 84, 85, 86, 87, 88, 89, 90, 91, 92, 93, 94, 95, 96, 97, 98, 99, 99.5, or 99.9 wt%, for example. A non-aqueous composition herein can be in the form of a household product, personal care product, pharmaceutical product, industrial product, or food product, for example.

In certain embodiments of the present disclosure, a composition comprising cellulose can be an aqueous composition having a viscosity of about, at least about, 100 cPs. An aqueous composition herein can have a viscosity of about, or at least about, 100, 250, 500, 750, 1000, 2000, 3000, 4000, 5000, 6000, 7000, 8000, 9000, 10000, 11000, 12000, 13000, 14000, 15000, 16000, 17000, 18000, 19000, 20000, 25000, 30000, 35000, 40000, 45000, or 50000 cPs (or any integer between 100 and

50000 cPs), for example. Examples of aqueous compositions herein include colloidal dispersions.

Viscosity can be measured with an aqueous composition herein at any temperature between about 3 °C to about 110 °C (or any integer between 3 and 110 °C), for example. Alternatively, viscosity can be measured at a temperature between about 4 °C to 30 °C, or about 20 °C to 25 °C, for instance. Viscosity can be measured at atmospheric pressure (about 760 torr) or any other higher or lower pressure.

The viscosity of an aqueous composition disclosed herein can be measured using a viscometer or rheometer, or using any other means known in the art. It would be understood by those skilled in the art that a viscometer or rheometer can be used to measure the viscosity of aqueous compositions herein that exhibit shear thinning behavior (i.e., having viscosities that vary with flow conditions). The viscosity of such embodiments can be measured at a rotational shear rate of about 0.1 to 1000 rpm (revolutions per minute), for example. In some embodiments, viscosity can be measured at a rotational shear rate of about 10, 60, 150, 250, or 600 rpm.

The pH of an aqueous composition disclosed herein can be between about 2.0 to about 12.0, for example. Alternatively, pH can be about 2.0, 3.0, 4.0, 5.0, 6.0, 7.0, 8.0, 9.0, 10.0, 11.0, 12.0; or between 5.0 to about 12.0; or between about 4.0 and 8.0; or between about 5.0 and 8.0, for example.

An aqueous composition herein can comprise a solvent having at least about 10 or 20 wt% water. In other embodiments, a solvent comprises at least about 30, 40, 50, 60, 70, 80, 90, or 100 wt% water (or any integer value between 10 and 100 wt%), for example.

Cellulose of the present disclosure can be present as insoluble material in an aqueous composition at a wt% of about, or at least about, 0.01, 0.05, 0.1, 0.2, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8, 0.9, 1.0, 1.2, 1.4, 1.6, 1.8, 2.0, 2.5, 3.0, 3.5, 4.0, 4.5, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19, 20, 21, 22, 23, 24, 25, 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, 40, 41, 42, 43, 44, 45, 46, 47, 48, 49, 50, 51, 52, 53, 55, 56, 57, 58, 59, 60, 61, 62, 63, 64, 65, 66, 67, 68, 69, 70, 71, 72, 73, 74, 75, 76, 77, 78, 79, 80, 81, 82, 83, 84, 85, 86, 87, 88, 89, or 90 wt%, for example. Example 4 below demonstrates that cellulose in certain aspects provides high viscosity to aqueous compositions at relatively low concentrations of the cellulose. Thus, certain

embodiments of the present disclosure are drawn to aqueous compositions with less than about 30, 29, 28, 27, 26, 25, 24, 23, 22, 21, 20, 19, 18, 17, 16, 15, 14, 13, 12, 11, 10, 9, 8, 7, 6, 5, 4, 3, 2, 1, or 0.5 wt% cellulose herein.

An aqueous composition herein can comprise other components in addition to the disclosed cellulose. For example, an aqueous composition can comprise one or more salts such as a sodium salt (e.g., NaCl, Na_2SO_4). Other non-limiting examples of salts include those having (i) an aluminum, ammonium, barium, calcium, chromium (II or III), copper (I or II), iron (II or III), hydrogen, lead (II), lithium, magnesium, manganese (II or III), mercury (I or II), potassium, silver, sodium strontium, tin (II or IV), or zinc cation, and (ii) an acetate, borate, bromate, bromide, carbonate, chlorate, chloride, chlorite, chromate, cyanamide, cyanide, dichromate, dihydrogen phosphate, ferricyanide, ferrocyanide, fluoride, hydrogen carbonate, hydrogen phosphate, hydrogen sulfate, hydrogen sulfide, hydrogen sulfite, hydride, hydroxide, hypochlorite, iodate, iodide, nitrate, nitride, nitrite, oxalate, oxide, perchlorate, permanganate, peroxide, phosphate, phosphide, phosphite, silicate, stannate, stannite, sulfate, sulfide, sulfite, tartrate, or thiocyanate anion. Thus, any salt having a cation from (i) above and an anion from (ii) above can be in an aqueous composition, for example. A salt can be present in an aqueous composition herein at a wt% of about (or at least about) .01 to about 10.00 (or any hundredth increment between .01 and 10.00), for example.

An aqueous composition comprising cellulose herein can be a colloidal dispersion, for example. The average size/diameter of cellulose particles in a colloidal dispersion herein typically ranges from between about 1 nm to 200000 nm (200 micrometers). Average particle size can be about 1-100 nm, 1-1000 nm, 1-10000 nm, 1-100000 nm, 1-200000 nm, 10-100 nm, 10-1000 nm, 10-10000 nm, 10-100000 nm, 10-200000 nm, 100-1000 nm, 100-10000 nm, 100-100000 nm, 100-200000 nm, 1000-10000 nm, 1000-100000, 1000-200000 nm, 10000-100000 nm, or 10000-200000 nm in some examples.

Aqueous compositions in certain embodiments have shear thinning behavior. Shear thinning behavior is observed as a decrease in viscosity of an aqueous composition as shear rate increases. Modification of the shear thinning behavior of an aqueous composition can be due to the admixture of cellulose herein to the aqueous composition. Thus, one or more cellulose materials of the present disclosure can be

added to an aqueous composition to modify its rheological profile (i.e., the flow properties of an aqueous liquid, solution, or mixture are modified). Also, one or more cellulose materials herein can be added to an aqueous composition to modify its viscosity.

The rheological properties of aqueous compositions herein can be observed by measuring viscosity over an increasing rotational shear rate (e.g., from about 0.1 rpm to about 1000 rpm). For example, shear thinning behavior of an aqueous composition disclosed herein can be observed as a decrease in viscosity (cPs) by at least about 5%, 10%, 15%, 20%, 25%, 30%, 35%, 40%, 45%, 50%, 55%, 60%, 65%, 70%, 75%, 80%, 85%, 90%, or 95% (or any integer between 5% and 95%) as the rotational shear rate increases from about 10 rpm to 60 rpm, 10 rpm to 150 rpm, 10 rpm to 250 rpm, 60 rpm to 150 rpm, 60 rpm to 250 rpm, or 150 rpm to 250 rpm.

A composition comprising cellulose herein, such as an aqueous composition or non-aqueous composition, may optionally contain one or more active enzymes. Non-limiting examples of suitable enzymes include proteases, peroxidases, lipolytic enzymes (e.g., metallolipolytic enzymes), xylanases, lipases, phospholipases, esterases (e.g., arylesterase, polyesterase), perhydrolases, cutinases, pectinases, pectate lyases, mannanases, keratinases, reductases, oxidases (e.g., choline oxidase), phenoloxidases, lipoxygenases, ligninases, pullulanases, tannases, pentosanases, malanases, arabinosidases, hyaluronidases, chondroitinases, laccases, metalloproteinases, amadoriases, glucoamylases, arabinofuranosidases, phytases, isomerases, transferases and amylases. If an enzyme(s) is included, it may be comprised in a composition herein at about 0.0001-0.1 wt% (e.g., 0.01-0.03 wt%) active enzyme (e.g., calculated as pure enzyme protein), for example.

An aqueous composition in certain aspects herein can be in the form of, and/or comprised in, a food product, personal care product, pharmaceutical product, household product, or industrial product, such as any of those products described below. Cellulose of the present disclosure can be used as thickening agents in each of these products, for example. Such a thickening agent may be used in conjunction with one or more other types of thickening agents if desired, such as those disclosed in U.S. Patent No. 8541041, the disclosure of which is incorporated herein by reference in its entirety.

Cellulose compounds disclosed herein are believed to be useful for providing one or more of the following physical properties to a personal care product, pharmaceutical product, household product, industrial product, or food product: thickening, freeze/thaw stability, lubricity, moisture retention and release, texture, consistency, shape retention, emulsification, binding, suspension, dispersion, gelation, reduced mineral hardness, for example. Examples of a concentration or amount of a dextran in a product can be any of the weight percentages provided herein, for example.

Personal care products herein are not particularly limited and include, for example, skin care compositions, cosmetic compositions, antifungal compositions, and antibacterial compositions. Personal care products herein may be in the form of, for example, lotions, creams, pastes, balms, ointments, pomades, gels, liquids, combinations of these and the like. The personal care products disclosed herein can include at least one active ingredient, if desired. An active ingredient is generally recognized as an ingredient that causes an intended pharmacological effect.

In certain embodiments, a skin care product can be applied to skin for addressing skin damage related to a lack of moisture. A skin care product may also be used to address the visual appearance of skin (e.g., reduce the appearance of flaky, cracked, and/or red skin) and/or the tactile feel of the skin (e.g., reduce roughness and/or dryness of the skin while improved the softness and subtleness of the skin). A skin care product typically may include at least one active ingredient for the treatment or prevention of skin ailments, providing a cosmetic effect, or for providing a moisturizing benefit to skin, such as zinc oxide, petrolatum, white petrolatum, mineral oil, cod liver oil, lanolin, dimethicone, hard fat, vitamin A, allantoin, calamine, kaolin, glycerin, or colloidal oatmeal, and combinations of these. A skin care product may include one or more natural moisturizing factors such as ceramides, hyaluronic acid, glycerin, squalane, amino acids, cholesterol, fatty acids, triglycerides, phospholipids, glycosphingolipids, urea, linoleic acid, glycosaminoglycans, mucopolysaccharide, sodium lactate, or sodium pyrrolidone carboxylate, for example. Other ingredients that may be included in a skin care product include, without limitation, glycerides, apricot kernel oil, canola oil, squalane, squalene, coconut oil, corn oil, jojoba oil, jojoba wax, lecithin, olive oil, safflower oil, sesame oil, shea butter, soybean oil, sweet almond oil, sunflower oil, tea tree oil, shea butter, palm oil, cholesterol, cholesterol esters, wax esters, fatty acids, and orange oil.

A personal care product herein can also be in the form of makeup, lipstick, mascara, rouge, foundation, blush, eyeliner, lip liner, lip gloss, other cosmetics, sunscreen, sun block, nail polish, nail conditioner, bath gel, shower gel, body wash, face wash, lip balm, skin conditioner, cold cream, moisturizer, body spray, soap, body scrub, exfoliant, astringent, scruffing lotion, depilatory, permanent waving solution, antidandruff formulation, antiperspirant composition, deodorant, shaving product, pre-shaving product, after-shaving product, cleanser, skin gel, rinse, dentifrice composition, toothpaste, or mouthwash, for example.

A personal care product in some aspects can be a hair care product. Examples of hair care products herein include shampoo, hair conditioner (leave-in or rinse-out), cream rinse, hair dye, hair coloring product, hair shine product, hair serum, hair anti-frizz product, hair split-end repair product, mousse, hair spray, and styling gel. A hair care product can be in the form of a liquid, paste, gel, solid, or powder in some embodiments. A hair care product as presently disclosed typically comprises one or more of the following ingredients, which are generally used to formulate hair care products: anionic surfactants such as polyoxyethylenelauryl ether sodium sulfate; cationic surfactants such as stearyltrimethylammonium chloride and/or distearyltrimethylammonium chloride; nonionic surfactants such as glyceryl monostearate, sorbitan monopalmitate and/or polyoxyethylenecetyl ether; wetting agents such as propylene glycol, 1,3-butylene glycol, glycerin, sorbitol, pyroglutamic acid salts, amino acids and/or trimethylglycine; hydrocarbons such as liquid paraffins, petrolatum, solid paraffins, squalane and/or olefin oligomers; higher alcohols such as stearyl alcohol and/or cetyl alcohol; superfatting agents; antidandruff agents; disinfectants; anti-inflammatory agents; crude drugs; water-soluble polymers such as methyl cellulose, hydroxycellulose and/or partially deacetylated chitin (in addition to one or more dextrans as disclosed herein); antiseptics such as paraben; ultra-violet light absorbers; pearling agents; pH adjustors; perfumes; and pigments.

A pharmaceutical product herein can be in the form of an emulsion, liquid, elixir, gel, suspension, solution, cream, or ointment, for example. Also, a pharmaceutical product herein can be in the form of any of the personal care products disclosed herein, such as an antibacterial or antifungal composition. A pharmaceutical product can further comprise one or more pharmaceutically acceptable carriers, diluents, and/or pharmaceutically acceptable salts. A cellulose material disclosed herein can also be

used in capsules, encapsulants, tablet coatings, and as an excipients for medicaments and drugs.

Non-limiting examples of food products herein include vegetable, meat, and soy patties; reformed seafood; reformed cheese sticks; cream soups; gravies and sauces; salad dressing; mayonnaise; onion rings; jams, jellies, and syrups; pie filling; potato products such as French fries and extruded fries; batters for fried foods, pancakes/waffles and cakes; pet foods; confectioneries (candy); beverages; frozen desserts; ice cream; cultured dairy products such as cottage cheese, yogurt, cheeses, and sour creams; cake icing and glazes; whipped topping; leavened and unleavened baked goods; and the like.

In certain embodiments, cellulose herein can be comprised in a foodstuff or any other ingestible material (e.g., enteral pharmaceutical preparation) in an amount that provides the desired degree of thickening and/or dispersion. For example, the concentration or amount of cellulose in a product can be about 0.1-3 wt%, 0.1-4 wt%, 0.1-5 wt%, or 0.1-10 wt%.

A household and/or industrial product herein can be in the form of drywall tape-joint compounds; mortars; grouts; cement plasters; spray plasters; cement stucco; adhesives; pastes; wall/ceiling texturizers; binders and processing aids for tape casting, extrusion forming, injection molding and ceramics; spray adherents and suspending/dispersing aids for pesticides, herbicides, and fertilizers; fabric care products such as fabric softeners and laundry detergents; hard surface cleaners; air fresheners; polymer emulsions; gels such as water-based gels; surfactant solutions; paints such as water-based paints; protective coatings; adhesives; sealants and caulks; inks such as water-based ink; metal-working fluids; emulsion-based metal cleaning fluids used in electroplating, phosphatizing, galvanizing and/or general metal cleaning operations; or hydraulic fluids (e.g., those used for downhole operations such as fracking and oil recovery), for example.

A cellulose material herein can be comprised in a personal care product, pharmaceutical product, household product, or industrial product in an amount that provides a desired degree of thickening and/or dispersion, for example. Examples of a concentration or amount of a cellulose in a product herein can be any of the cellulose weight percentages provided in the present disclosure, for example.

A food product comprising cellulose as disclosed herein can be in the form of a confectionery, for example. A confectionary herein can contain one or more sugars (e.g., sucrose, fructose, dextrose) for sweetening, or otherwise be sugar-free.

Examples of confectioneries herein include boiled sugars (hard boiled candies [i.e., hard candy]), dragees, jelly candies, gums, licorice, chews, caramels, toffee, fudge, chewing gums, bubble gums, nougat, chewy pastes, halawa, tablets, lozenges, icing, frosting, pudding, and gels (e.g., fruit gels, gelatin dessert). Other examples of confectioneries include aerated confectioneries such as marshmallows, and baked confectioneries.

A confectionery herein can optionally be prepared with chocolate, in any form (e.g., bars, candies, bonbons, truffles, lentils). A confectionary can be coated with chocolate, sugar-coated, candied, glazed, and/or film-coated, for example. Film-coating processes typically comprise applying to the surface of a confectionery a film-forming liquid composition which becomes, after drying, a protective film. This film-coating serves, for example, to protect the active principles contained in the confectionery; to protect the confectionery itself from moisture, shocks, and/or friability; and/or to confer the confectionery attractive visual properties (e.g., shine, uniform color, smooth surface). Such a film can comprise cellulose as disclosed herein.

In certain embodiments, a confectionery can be filled with a filling that is liquid, pasty, solid, or powdered. Cellulose herein can be comprised in such a filling, in which case cellulose is optionally also included in the confectionery component being filled.

A confectionery herein is optionally sugar-free, comprising no sugar and typically instead having one or more artificial and/or non-sugar sweeteners (optionally non-caloric) (e.g., aspartame, saccharin, STEVIA, SUCRALOSE). A sugar-free confectionery in certain embodiments can comprise one or more polyols (e.g., erythritol, glycerol, lactitol, mannitol, maltitol, xylitol), soluble fibers, and/or proteins in place of sugar.

A food product comprising cellulose as disclosed herein can be in the form of a pet food, for example. A pet food herein can be a food for a domesticated animal such as a dog or cat (or any other companion animal), for example. A pet food in certain embodiments provides to a domestic animal one or more of the following: necessary dietary requirements, treats (e.g., dog biscuits), food supplements. Examples of pet

food include dry pet food (e.g., kernels, kibbles), semi-moist compositions, wet pet food (e.g., canned pet food), or any combination thereof. Wet pet food typically has a moisture content over 65%. Semi-moist pet food typically has a moisture content of 20-65% and can include humectants such as propylene glycol, potassium sorbate, and ingredients that prevent microbial growth (bacteria and mold). Dry pet food typically has a moisture content less than 20% and its processing usually includes extruding, drying and/or baking. A pet food can optionally be in the form of a gravy, yogurt, powder, suspension, chew, or treat (e.g., biscuits); all these compositions can also be used as pet food supplements, if desired. Pet treats can be semi-moist chewable treats; dry treats; chewable bones; baked, extruded or stamped treats; or confection treats, for example. Examples of pet food compositions/formulations in which cellulose herein can be added include those disclosed in U.S. Patent Appl. Publ. Nos. 2013/0280352 and 2010/0159103, and U.S. Patent No. 6977084, which are all incorporated herein by reference.

Compositions comprising cellulose as disclosed herein can be in the form of a fabric care composition. A fabric care composition herein can be used for hand wash, machine wash and/or other purposes such as soaking and/or pretreatment of fabrics, for example. A fabric care composition may take the form of, for example, a laundry detergent; fabric conditioner; any wash-, rinse-, or dryer-added product; unit dose or spray. Fabric care compositions in a liquid form may be in the form of an aqueous composition as disclosed herein. In other aspects, a fabric care composition can be in a dry form such as a granular detergent or dryer-added fabric softener sheet. Other non-limiting examples of fabric care compositions herein include: granular or powder-form all-purpose or heavy-duty washing agents; liquid, gel or paste-form all-purpose or heavy-duty washing agents; liquid or dry fine-fabric (e.g., delicates) detergents; cleaning auxiliaries such as bleach additives, “stain-stick”, or pre-treatments; substrate-laden products such as dry and wetted wipes, pads, or sponges; sprays and mists.

A detergent composition herein may be in any useful form, e.g., as powders, granules, pastes, bars, unit dose, or liquid. A liquid detergent may be aqueous, typically containing up to about 70 wt% of water and 0 wt% to about 30 wt% of organic solvent. It may also be in the form of a compact gel type containing only about 30 wt% water.

A detergent composition herein typically comprises one or more surfactants, wherein the surfactant is selected from nonionic surfactants, anionic surfactants, cationic surfactants, ampholytic surfactants, zwitterionic surfactants, semi-polar nonionic surfactants and mixtures thereof. In some embodiments, the surfactant is present at a level of from about 0.1% to about 60%, while in alternative embodiments the level is from about 1% to about 50%, while in still further embodiments the level is from about 5% to about 40%, by weight of the detergent composition. A detergent will usually contain 0 wt% to about 50 wt% of an anionic surfactant such as linear alkylbenzenesulfonate (LAS), alpha-olefinsulfonate (AOS), alkyl sulfate (fatty alcohol sulfate) (AS), alcohol ethoxysulfate (AEOS or AES), secondary alkanesulfonates (SAS), alpha-sulfo fatty acid methyl esters, alkyl- or alkenylsuccinic acid, or soap. In addition, a detergent composition may optionally contain 0 wt% to about 40 wt% of a nonionic surfactant such as alcohol ethoxylate (AEO or AE), carboxylated alcohol ethoxylates, nonylphenol ethoxylate, alkylpolyglycoside, alkyldimethylamineoxide, ethoxylated fatty acid monoethanolamide, fatty acid monoethanolamide, or polyhydroxy alkyl fatty acid amide (as described for example in WO92/06154, which is incorporated herein by reference).

A detergent composition herein typically comprises one or more detergent builders or builder systems. In some embodiments incorporating at least one builder, the cleaning compositions comprise at least about 1%, from about 3% to about 60%, or even from about 5% to about 40%, builder by weight of the composition. Builders include, but are not limited to, alkali metal, ammonium and alkanolammonium salts of polyphosphates, alkali metal silicates, alkaline earth and alkali metal carbonates, aluminosilicates, polycarboxylate compounds, ether hydroxypolycarboxylates, copolymers of maleic anhydride with ethylene or vinyl methyl ether, 1, 3, 5-trihydroxy benzene-2, 4, 6-trisulphonic acid, and carboxymethyloxysuccinic acid, various alkali metal, ammonium and substituted ammonium salts of polyacetic acids such as ethylenediamine tetraacetic acid and nitrilotriacetic acid, as well as polycarboxylates such as mellitic acid, succinic acid, citric acid, oxydisuccinic acid, polymaleic acid, benzene 1,3,5-tricarboxylic acid, carboxymethyloxysuccinic acid, and soluble salts thereof. Indeed, it is contemplated that any suitable builder will find use in various embodiments of the present disclosure. Examples of a detergent builder or complexing agent include zeolite, diphosphate, triphosphate, phosphonate, citrate, nitrilotriacetic

acid (NTA), ethylenediaminetetraacetic acid (EDTA), diethylenetriaminepentaacetic acid (DTMPA), alkyl- or alkenylsuccinic acid, soluble silicates or layered silicates (e.g., SKS-6 from Hoechst).

In some embodiments, builders form water-soluble hardness ion complexes (e.g., sequestering builders), such as citrates and polyphosphates (e.g., sodium tripolyphosphate and sodium tripolyphospate hexahydrate, potassium tripolyphosphate, and mixed sodium and potassium tripolyphosphate, etc.). It is contemplated that any suitable builder will find use in the present disclosure, including those known in the art (See, e.g., EP2100949).

In some embodiments, suitable builders can include phosphate builders and non-phosphate builders. In some embodiments, a builder is a phosphate builder. In some embodiments, the builder is a non-phosphate builder. If present, a builder can be used at a level of from 0.1% to 80%, or from 5% to 60%, or from 10% to 50%, by weight of the composition. In some embodiments, the product comprises a mixture of phosphate and non-phosphate builders. Suitable phosphate builders include mono-phosphates, di-phosphates, tri-polyphosphates or oligomeric-polyphosphates, including the alkali metal salts of these compounds, including the sodium salts. In some embodiments, a builder can be sodium tripolyphosphate (STPP). Additionally, the composition can comprise carbonate and/or citrate, preferably citrate that helps to achieve a neutral pH composition. Other suitable non-phosphate builders include homopolymers and copolymers of polycarboxylic acids and their partially or completely neutralized salts, monomeric polycarboxylic acids and hydroxycarboxylic acids and their salts. In some embodiments, salts of the above mentioned compounds include ammonium and/or alkali metal salts, i.e., lithium, sodium, and potassium salts, including sodium salts. Suitable polycarboxylic acids include acyclic, alicyclic, hetero-cyclic and aromatic carboxylic acids, wherein in some embodiments, they can contain at least two carboxyl groups which are in each case separated from one another by, in some instances, no more than two carbon atoms.

A detergent composition herein can comprise at least one chelating agent. Suitable chelating agents include, but are not limited to copper, iron and/or manganese chelating agents and mixtures thereof. In embodiments in which at least one chelating agent is used, the composition comprises from about 0.1% to about 15%, or even from about 3.0% to about 10%, chelating agent by weight of the composition.

A detergent composition herein can comprise at least one deposition aid. Suitable deposition aids include, but are not limited to, polyethylene glycol, polypropylene glycol, polycarboxylate, soil release polymers such as polytelephthalic acid, clays such as kaolinite, montmorillonite, atapulgite, illite, bentonite, halloysite, and mixtures thereof.

A detergent composition herein can comprise one or more dye transfer inhibiting agents. Suitable polymeric dye transfer inhibiting agents include, but are not limited to, polyvinylpyrrolidone polymers, polyamine N-oxide polymers, copolymers of N-vinylpyrrolidone and N-vinylimidazole, polyvinyloxazolidones and polyvinylimidazoles or mixtures thereof. Additional dye transfer inhibiting agents include manganese phthalocyanine, peroxidases, polyvinylpyrrolidone polymers, polyamine N-oxide polymers, copolymers of N-vinylpyrrolidone and N-vinylimidazole, polyvinyloxazolidones and polyvinylimidazoles and/or mixtures thereof; chelating agents examples of which include ethylene-diamine-tetraacetic acid (EDTA); diethylene triamine penta methylene phosphonic acid (DTPMP); hydroxy-ethane diphosphonic acid (HEDP); ethylenediamine N,N'-disuccinic acid (EDDS); methyl glycine diacetic acid (MGDA); diethylene triamine penta acetic acid (DTPA); propylene diamine tetracetic acid (PDT A); 2-hydroxypyridine-N-oxide (HPNO); or methyl glycine diacetic acid (MGDA); glutamic acid N,N-diacetic acid (N,N-dicarboxymethyl glutamic acid tetrasodium salt (GLDA); nitrilotriacetic acid (NTA); 4,5-dihydroxy-m-benzenedisulfonic acid; citric acid and any salts thereof; N-hydroxyethyl ethylenediaminetri-acetic acid (HEDTA), triethylenetetraaminehexaacetic acid (TTHA), N-hydroxyethyliminodiacetic acid (HEIDA), dihydroxyethylglycine (DHEG), ethylenediaminetetrapropionic acid (EDTP) and derivatives thereof, which can be used alone or in combination with any of the above. In embodiments in which at least one dye transfer inhibiting agent is used, a composition herein may comprise from about 0.0001% to about 10%, from about 0.01% to about 5%, or even from about 0.1% to about 3%, by weight of the composition.

A detergent composition herein can comprise silicates. In some of these embodiments, sodium silicates (e.g., sodium disilicate, sodium metasilicate, and/or crystalline phyllosilicates) find use. In some embodiments, silicates are present at a level of from about 1% to about 20% by weight of the composition. In some embodiments, silicates are present at a level of from about 5% to about 15% by weight of the composition.

A detergent composition herein can comprise dispersants. Suitable water-soluble organic materials include, but are not limited to the homo- or co-polymeric acids or their salts, in which the polycarboxylic acid comprises at least two carboxyl radicals separated from each other by not more than two carbon atoms.

A detergent composition herein may additionally comprise one or more enzymes. Examples of enzymes include proteases, peroxidases, lipolytic enzymes (e.g., metallolipolytic enzymes), xylanases, lipases, phospholipases, esterases (e.g., arylesterase, polyesterase), perhydrolases, cutinases, pectinases, pectate lyases, mannanases, keratinases, reductases, oxidases (e.g., choline oxidase, phenoloxidase), phenoloxidases, lipoxygenases, ligninases, pullulanases, tannases, pentosanases, malanases, arabinosidases, hyaluronidases, chondroitinases, laccases, metalloproteinases, amadoriases, glucoamylases, alpha-amylases, beta-amylases, galactosidases, galactanases, catalases, carageenases, hyaluronidases, keratinases, lactases, ligninases, peroxidases, phosphatases, polygalacturonases, rhamnogalactouronases, tannases, transglutaminases, xyloglucanases, xylosidases, metalloproteases, arabinofuranosidases, phytases, isomerases, transferases and/or amylases in any combination.

In some embodiments of the present disclosure, a detergent composition can comprise one or more enzymes, each at a level from about 0.00001% to about 10% by weight of the composition and the balance of cleaning adjunct materials by weight of composition. In some other embodiments of the present disclosure, a detergent composition can also comprise each enzyme at a level of about 0.0001% to about 10%, about 0.001% to about 5%, about 0.001% to about 2%, or about 0.005% to about 0.5%, by weight of the composition.

Suitable proteases include those of animal, vegetable or microbial origin. In some embodiments, microbial proteases are used. In some embodiments, chemically or genetically modified mutants are included. In some embodiments, the protease is a serine protease, preferably an alkaline microbial protease or a trypsin-like protease. Examples of alkaline proteases include subtilisins, especially those derived from Bacillus (e.g., subtilisin, lentus, amyloliquefaciens, subtilisin Carlsberg, subtilisin 309, subtilisin 147 and subtilisin 168). Additional examples include those mutant proteases described in U.S. Pat. Nos. RE34606, 5955340, 5700676, 6312936 and 6482628, all of which are incorporated herein by reference. Additional protease examples include, but

are not limited to, trypsin (e.g., of porcine or bovine origin), and the Fusarium protease described in WO89/06270. In some embodiments, commercially available protease enzymes include, but are not limited to, MAXATASE®, MAXACAL™, MAXAPEM™, OPTICLEAN®, OPTIMASE®, PROPERASE®, PURAFECT®, PURAFECT® OXP, PURAMAX™, EXCELLASE™, PREFERENZ™ proteases (e.g. P100, P110, P280), EFFECTENZ™ proteases (e.g. P1000, P1050, P2000), EXCELLENZ™ proteases (e.g. P1000), ULTIMASE®, and PURAFAST™ (Genencor); ALCALASE®, SAVINASE®, PRIMASE®, DURAZYM™, POLARZYME®, OVOZYME®, KANNASE®, LIQUANASE®, NEUTRASE®, RELASE® and ESPERASE® (Novozymes); BLAP™ and BLAP™ variants (Henkel Kommanditgesellschaft auf Aktien, Duesseldorf, Germany), and KAP (B. alkalophilus subtilisin; Kao Corp., Tokyo, Japan). Various proteases are described in WO95/23221, WO92/21760, WO09/149200, WO09/149144, WO09/149145, WO11/072099, WO10/056640, WO10/056653, WO11/140364, WO12/151534, U.S. Pat. Publ. No. 2008/0090747, and U.S. Pat. Nos. 5801039, 5340735, 5500364, 5855625, RE34606, 5955340, 5700676, 6312936, 6482628, 8530219, and various other patents. In some further embodiments, neutral metalloproteases find use in the present disclosure, including but not limited to, the neutral metalloproteases described in WO1999014341, WO1999033960, WO1999014342, WO1999034003, WO2007044993, WO2009058303 and WO2009058661, all of which are incorporated herein by reference. Exemplary metalloproteases include nprE, the recombinant form of neutral metalloprotease expressed in Bacillus subtilis (See e.g., WO07/044993), and PMN, the purified neutral metalloprotease from Bacillus amyloliquefaciens.

Suitable mannanases include, but are not limited to, those of bacterial or fungal origin. Chemically or genetically modified mutants are included in some embodiments. Various mannanases are known which find use in the present disclosure (See, e.g., U.S. Pat. Nos. 6566114, 6602842, and 6440991, all of which are incorporated herein by reference). Commercially available mannanases that find use in the present disclosure include, but are not limited to MANNASTAR®, PURABRITE™, and MANNAWAY®.

Suitable lipases include those of bacterial or fungal origin. Chemically modified, proteolytically modified, or protein engineered mutants are included. Examples of useful lipases include those from the genera *Humicola* (e.g., *H. lanuginosa*, EP258068 and EP305216; *H. insolens*, WO96/13580), *Pseudomonas* (e.g., *P. alcaligenes* or *P. pseudoalcaligenes*, EP218272; *P. cepacia*, EP331376; *P. stutzeri*, GB1372034; *P.*

fluorescens and *Pseudomonas* sp. strain SD 705, WO95/06720 and WO96/27002; *P. wisconsinensis*, WO96/12012); and *Bacillus* (e.g., *B. subtilis*, Dartois et al., *Biochemica et Biophysica Acta* 1131:253-360; *B. stearothermophilus*, JP64/744992; *B. pumilus*, WO91/16422). Furthermore, a number of cloned lipases find use in some embodiments of the present disclosure, including but not limited to, Penicillium camembertii lipase (See, Yamaguchi et al., Gene 103:61-67 [1991]), Geotricum candidum lipase (See, Schimada et al., J. Biochem., 106:383-388 [1989]), and various Rhizopus lipases such as R. delemar lipase (See, Hass et al., Gene 109:117-113 [1991]), a R. niveus lipase (Kugimiya et al., Biosci. Biotech. Biochem. 56:716-719 [1992]) and R. oryzae lipase. Additional lipases useful herein include, for example, those disclosed in WO92/05249, WO94/01541, WO95/35381, WO96/00292, WO95/30744, WO94/25578, WO95/14783, WO95/22615, WO97/04079, WO97/07202, EP407225 and EP260105. Other types of lipase polypeptide enzymes such as cutinases also find use in some embodiments of the present disclosure, including but not limited to, cutinase derived from Pseudomonas mendocina (See, WO88/09367), and cutinase derived from Fusarium solani pisi (See, WO90/09446). Examples of certain commercially available lipase enzymes useful herein include M1 LIPASE™, LUMA FAST™, and LIPOMAX™ (Genencor); LIPEX®, LIPOLASE® and LIPOLASE® ULTRA (Novozymes); and LIPASE P™ "Amano" (Amano Pharmaceutical Co. Ltd., Japan).

Suitable polyesterases include, for example, those disclosed in WO01/34899, WO01/14629 and U.S. Patent No. 6933140.

A detergent composition herein can also comprise 2,6-beta-D-fructan hydrolase, which is effective for removal/cleaning of certain biofilms present on household and/or industrial textiles/laundry.

Suitable amylases include, but are not limited to those of bacterial or fungal origin. Chemically or genetically modified mutants are included in some embodiments. Amylases that find use in the present disclosure include, but are not limited to, alpha-amylases obtained from B. licheniformis (See e.g., GB1296839). Additional suitable amylases include those disclosed in W09510603, WO9526397, WO9623874, WO9623873, WO9741213, WO9919467, WO0060060, WO0029560, WO9923211, WO9946399, WO0060058, WO0060059, WO9942567, WO0114532, WO02092797, WO0166712, WO0188107, WO0196537, WO0210355, WO9402597, WO0231124, WO9943793, WO9943794, WO2004113551, WO2005001064, WO2005003311,

WO0164852, WO2006063594, WO2006066594, WO2006066596, WO2006012899, WO2008092919, WO2008000825, WO2005018336, WO2005066338, WO2009140504, WO2005019443, WO2010091221, WO2010088447, WO0134784, WO2006012902, WO2006031554, WO2006136161, WO2008101894, WO2010059413, WO2011098531, WO2011080352, WO2011080353, WO2011080354, WO2011082425, WO2011082429, WO2011076123, WO2011087836, WO2011076897, WO94183314, WO9535382, WO9909183, WO9826078, WO9902702, WO9743424, WO9929876, WO9100353, WO9605295, WO9630481, WO9710342, WO2008088493, WO2009149419, WO2009061381, WO2009100102, WO2010104675, WO2010117511, and WO2010115021, all of which are incorporated herein by reference.

Suitable amylases include, for example, commercially available amylases such as STAINZYME®, STAINZYME PLUS®, NATALASE®, DURAMYL®, TERMAMYL®, TERMAMYL ULTRA®, FUNGAMYL® and BAN™ (Novo Nordisk A/S and Novozymes A/S); RAPIDASE®, POWERASE®, PURASTAR® and PREFERENZ™ (DuPont Industrial Biosciences).

Suitable peroxidases/oxidases contemplated for use in the compositions include those of plant, bacterial or fungal origin. Chemically modified or protein engineered mutants are included. Examples of peroxidases useful herein include those from the genus *Coprinus* (e.g., *C. cinereus*, WO93/24618, WO95/10602, and WO98/15257), as well as those referenced in WO2005056782, WO2007106293, WO2008063400, WO2008106214, and WO2008106215. Commercially available peroxidases useful herein include, for example, GUARDZYME™ (Novo Nordisk A/S and Novozymes A/S).

In some embodiments, peroxidases are used in combination with hydrogen peroxide or a source thereof (e.g., a percarbonate, perborate or persulfate). In some alternative embodiments, oxidases are used in combination with oxygen. Both types of enzymes are used for "solution bleaching" (i.e., to prevent transfer of a textile dye from a dyed fabric to another fabric when the fabrics are washed together in a wash liquor), preferably together with an enhancing agent (See e.g., WO94/12621 and WO95/01426). Suitable peroxidases/oxidases include, but are not limited to, those of plant, bacterial or fungal origin. Chemically or genetically modified mutants are included in some embodiments.

Enzymes that may be comprised in a detergent composition herein may be stabilized using conventional stabilizing agents, e.g., a polyol such as propylene glycol

or glycerol; a sugar or sugar alcohol; lactic acid; boric acid or a boric acid derivative (e.g., an aromatic borate ester).

A detergent composition herein may contain about 1 wt% to about 65 wt% of a detergent builder or complexing agent such as zeolite, diphosphate, triphosphate, phosphonate, citrate, nitrilotriacetic acid (NTA), ethylenediaminetetraacetic acid (EDTA), diethylenetriaminepentaacetic acid (DTMPA), alkyl- or alkenylsuccinic acid, soluble silicates or layered silicates (e.g., SKS-6 from Hoechst). A detergent may also be unbuilt, i.e., essentially free of detergent builder.

A detergent composition in certain embodiments may comprise one or more other types of polymers in addition to a cellulose as disclosed herein. Examples of other types of polymers useful herein include carboxymethyl cellulose (CMC), poly(vinylpyrrolidone) (PVP), polyethylene glycol (PEG), poly(vinyl alcohol) (PVA), polycarboxylates such as polyacrylates, maleic/acrylic acid copolymers and lauryl methacrylate/acrylic acid copolymers.

A detergent composition herein may contain a bleaching system. For example, a bleaching system can comprise an H_2O_2 source such as perborate or percarbonate, which may be combined with a peracid-forming bleach activator such as tetraacetylethylenediamine (TAED) or nonanoyloxybenzenesulfonate (NOBS). Alternatively, a bleaching system may comprise peroxyacids (e.g., amide, imide, or sulfone type peroxyacids). Alternatively still, a bleaching system can be an enzymatic bleaching system comprising perhydrolase, for example, such as the system described in WO2005/056783.

A detergent composition herein may also contain conventional detergent ingredients such as fabric conditioners, clays, foam boosters, suds suppressors, anti-corrosion agents, soil-suspending agents, anti-soil redeposition agents, dyes, bactericides, tarnish inhibiters, optical brighteners, or perfumes. The pH of a detergent composition herein (measured in aqueous solution at use concentration) is usually neutral or alkaline (e.g., pH of about 7.0 to about 11.0).

Particular forms of detergent compositions that can be adapted for purposes disclosed herein are disclosed in, for example, US20090209445A1, US20100081598A1, US7001878B2, EP1504994B1, WO2001085888A2, WO2003089562A1, WO2009098659A1, WO2009098660A1, WO2009112992A1, WO2009124160A1, WO2009152031A1, WO2010059483A1, WO2010088112A1,

WO2010090915A1, WO2010135238A1, WO2011094687A1, WO2011094690A1, WO2011127102A1, WO2011163428A1, WO2008000567A1, WO2006045391A1, WO2006007911A1, WO2012027404A1, EP1740690B1, WO2012059336A1, US6730646B1, WO2008087426A1, WO2010116139A1, and WO2012104613A1, all of which are incorporated herein by reference.

Laundry detergent compositions herein can optionally be heavy duty (all purpose) laundry detergent compositions. Exemplary heavy duty laundry detergent compositions comprise a detersive surfactant (10%-40% wt/wt), including an anionic detersive surfactant (selected from a group of linear or branched or random chain, substituted or unsubstituted alkyl sulphates, alkyl sulphonates, alkyl alkoxylated sulphate, alkyl phosphates, alkyl phosphonates, alkyl carboxylates, and/or mixtures thereof), and optionally non-ionic surfactant (selected from a group of linear or branched or random chain, substituted or unsubstituted alkyl alkoxylated alcohol, e.g., C8-C18 alkyl ethoxylated alcohols and/or C6-C12 alkyl phenol alkoxylates), where the weight ratio of anionic detersive surfactant (with a hydrophilic index (HIc) of from 6.0 to 9) to non-ionic detersive surfactant is greater than 1:1. Suitable detersive surfactants also include cationic detersive surfactants (selected from a group of alkyl pyridinium compounds, alkyl quaternary ammonium compounds, alkyl quaternary phosphonium compounds, alkyl ternary sulphonium compounds, and/or mixtures thereof); zwitterionic and/or amphoteric detersive surfactants (selected from a group of alkanolamine sulpho-betaines); ampholytic surfactants; semi-polar non-ionic surfactants and mixtures thereof.

A detergent herein such as a heavy duty laundry detergent composition may optionally include, a surfactancy boosting polymer consisting of amphiphilic alkoxylated grease cleaning polymers (selected from a group of alkoxylated polymers having branched hydrophilic and hydrophobic properties, such as alkoxylated polyalkylenimines in the range of 0.05 wt% - 10 wt%) and/or random graft polymers (typically comprising of hydrophilic backbone comprising monomers selected from the group consisting of: unsaturated C1-C6 carboxylic acids, ethers, alcohols, aldehydes, ketones, esters, sugar units, alkoxy units, maleic anhydride, saturated polyalcohols such as glycerol, and mixtures thereof; and hydrophobic side chain(s) selected from the group consisting of: C4-C25 alkyl group, polypropylene, polybutylene, vinyl ester of a

saturated C1-C6 mono-carboxylic acid, C1-C6 alkyl ester of acrylic or methacrylic acid, and mixtures thereof.

A detergent herein such as a heavy duty laundry detergent composition may optionally include additional polymers such as soil release polymers (include anionically end-capped polyesters, for example SRP1, polymers comprising at least one monomer unit selected from saccharide, dicarboxylic acid, polyol and combinations thereof, in random or block configuration, ethylene terephthalate-based polymers and co-polymers thereof in random or block configuration, for example REPEL-O-TEX SF, SF-2 AND SRP6, TEXCARE SRA100, SRA300, SRN100, SRN170, SRN240, SRN300 AND SRN325, MARLOQUEST SL), anti-redeposition agent(s) (0.1 wt% to 10 wt%), include carboxylate polymers, such as polymers comprising at least one monomer selected from acrylic acid, maleic acid (or maleic anhydride), fumaric acid, itaconic acid, aconitic acid, mesaconic acid, citraconic acid, methylenemalonic acid, and any mixture thereof, vinylpyrrolidone homopolymer, and/or polyethylene glycol, molecular weight in the range of from 500 to 100,000 Da); and polymeric carboxylate (such as maleate/acrylate random copolymer or polyacrylate homopolymer).

A detergent herein such as a heavy duty laundry detergent composition may optionally further include saturated or unsaturated fatty acids, preferably saturated or unsaturated C12-C24 fatty acids (0 wt% to 10 wt%); deposition aids in addition to a cellulose compound disclosed herein (examples for which include polysaccharides, poly diallyl dimethyl ammonium halides (DADMAC), and co-polymers of DAD MAC with vinyl pyrrolidone, acrylamides, imidazoles, imidazolinium halides, and mixtures thereof, in random or block configuration, cationic guar gum, cationic starch, cationic polyacylamides, and mixtures thereof.

A detergent herein such as a heavy duty laundry detergent composition may optionally further include dye transfer inhibiting agents, examples of which include manganese phthalocyanine, peroxidases, polyvinylpyrrolidone polymers, polyamine N-oxide polymers, copolymers of N-vinylpyrrolidone and N-vinylimidazole, polyvinyloxazolidones and polyvinylimidazoles and/or mixtures thereof; chelating agents, examples of which include ethylene-diamine-tetraacetic acid (EDTA), diethylene triamine penta methylene phosphonic acid (DTPMP), hydroxy-ethane diphosphonic acid (HEDP), ethylenediamine N,N'-disuccinic acid (EDDS), methyl glycine diacetic acid (MGDA), diethylene triamine penta acetic acid (DTPA), propylene diamine tetracetic

acid (PDTA), 2-hydroxypyridine-N-oxide (HPNO), or methyl glycine diacetic acid (MGDA), glutamic acid N,N-diacetic acid (N,N-dicarboxymethyl glutamic acid tetrasodium salt (GLDA), nitrilotriacetic acid (NTA), 4,5-dihydroxy-m-benzenedisulfonic acid, citric acid and any salts thereof, N-hydroxyethylethylenediaminetriacetic acid (HEDTA), triethylenetetraaminehexaacetic acid (TTHA), N-hydroxyethyliminodiacetic acid (HEIDA), dihydroxyethylglycine (DHEG), ethylenediaminetetrapropionic acid (EDTP), and derivatives thereof.

A detergent herein such as a heavy duty laundry detergent composition may optionally include silicone or fatty-acid based suds suppressors; hueing dyes, calcium and magnesium cations, visual signaling ingredients, anti-foam (0.001 wt% to about 4.0 wt%), and/or a structurant/thickener (0.01 wt% to 5 wt%) selected from the group consisting of diglycerides and triglycerides, ethylene glycol distearate, microcrystalline cellulose, microfiber cellulose, biopolymers, xanthan gum, gellan gum, and mixtures thereof). Such structurant/thickener would be, in certain aspects, in addition to the one or more cellulose materials disclosed herein comprised in the detergent. A structurant can also be referred to as a structural agent.

A detergent herein can be in the form of a heavy duty dry/solid laundry detergent composition, for example. Such a detergent may include: (i) a detersive surfactant, such as any anionic detersive surfactant disclosed herein, any non-ionic detersive surfactant disclosed herein, any cationic detersive surfactant disclosed herein, any zwitterionic and/or amphoteric detersive surfactant disclosed herein, any ampholytic surfactant, any semi-polar non-ionic surfactant, and mixtures thereof; (ii) a builder, such as any phosphate-free builder (e.g., zeolite builders in the range of 0 wt% to less than 10 wt%), any phosphate builder (e.g., sodium tri-polyphosphate in the range of 0 wt% to less than 10 wt%), citric acid, citrate salts and nitrilotriacetic acid, any silicate salt (e.g., sodium or potassium silicate or sodium meta-silicate in the range of 0 wt% to less than 10 wt%); any carbonate salt (e.g., sodium carbonate and/or sodium bicarbonate in the range of 0 wt% to less than 80 wt%), and mixtures thereof; (iii) a bleaching agent, such as any photobleach (e.g., sulfonated zinc phthalocyanines, sulfonated aluminum phthalocyanines, xanthenes dyes, and mixtures thereof), any hydrophobic or hydrophilic bleach activator (e.g., dodecanoyl oxybenzene sulfonate, decanoyl oxybenzene sulfonate, decanoyl oxybenzoic acid or salts thereof, 3,5,5-trimethy hexanoyl oxybenzene sulfonate, tetraacetyl ethylene diamine-TAED, nonanoyloxybenzene

sulfonate-NOBS, nitrile quats, and mixtures thereof), any source of hydrogen peroxide (e.g., inorganic perhydrate salts, examples of which include mono or tetra hydrate sodium salt of perborate, percarbonate, persulfate, perphosphate, or persilicate), any preformed hydrophilic and/or hydrophobic peracids (e.g., percarboxylic acids and salts, percarbonic acids and salts, perimidic acids and salts, peroxymonosulfuric acids and salts, and mixtures thereof); and/or (iv) any other components such as a bleach catalyst (e.g., imine bleach boosters examples of which include iminium cations and polyions, iminium zwitterions, modified amines, modified amine oxides, N-sulphonyl imines, N-phosphonyl imines, N-acyl imines, thiadiazole dioxides, perfluoroimines, cyclic sugar ketones, and mixtures thereof), and a metal-containing bleach catalyst (e.g., copper, iron, titanium, ruthenium, tungsten, molybdenum, or manganese cations along with an auxiliary metal cations such as zinc or aluminum and a sequestrate such as EDTA, ethylenediaminetetra(methylenephosphonic acid).

Compositions comprising cellulose as disclosed herein can be in the form of a dishwashing detergent composition, for example. Examples of dishwashing detergents include automatic dishwashing detergents (typically used in dishwasher machines) and hand-washing dish detergents. A dishwashing detergent composition can be in any dry or liquid/aqueous form as disclosed herein, for example. Components that may be included in certain embodiments of a dishwashing detergent composition include, for example, one or more of a phosphate; oxygen- or chlorine-based bleaching agent; non-ionic surfactant; alkaline salt (e.g., metasilicates, alkali metal hydroxides, sodium carbonate); any active enzyme disclosed herein; anti-corrosion agent (e.g., sodium silicate); anti-foaming agent; additives to slow down the removal of glaze and patterns from ceramics; perfume; anti-caking agent (in granular detergent); starch (in tablet-based detergents); gelling agent (in liquid/gel based detergents); and/or sand (powdered detergents).

Dishwashing detergents such as an automatic dishwasher detergent or liquid dishwashing detergent can comprise (i) a non-ionic surfactant, including any ethoxylated non-ionic surfactant, alcohol alkoxylated surfactant, epoxy-capped poly(oxyalkylated) alcohol, or amine oxide surfactant present in an amount from 0 to 10 wt%; (ii) a builder, in the range of about 5-60 wt%, including any phosphate builder (e.g., mono-phosphates, di-phosphates, tri-polyphosphates, other oligomeric-

polyphosphates, sodium tripolyphosphate-STPP), any phosphate-free builder (e.g., amino acid-based compounds including methyl-glycine-diacetic acid [MGDA] and salts or derivatives thereof, glutamic-N,N-diacetic acid [GLDA] and salts or derivatives thereof, iminodisuccinic acid (IDS) and salts or derivatives thereof, carboxy methyl inulin and salts or derivatives thereof, nitrilotriacetic acid [NTA], diethylene triamine penta acetic acid [DTPA], B-alaninediacetic acid [B-ADA] and salts thereof), homopolymers and copolymers of poly-carboxylic acids and partially or completely neutralized salts thereof, monomeric polycarboxylic acids and hydroxycarboxylic acids and salts thereof in the range of 0.5 wt% to 50 wt%, or sulfonated/carboxylated polymers in the range of about 0.1 wt% to about 50 wt%; (iii) a drying aid in the range of about 0.1 wt% to about 10 wt% (e.g., polyesters, especially anionic polyesters, optionally together with further monomers with 3 to 6 functionalities – typically acid, alcohol or ester functionalities which are conducive to polycondensation, polycarbonate-, polyurethane- and/or polyurea-polyorganosiloxane compounds or precursor compounds thereof, particularly of the reactive cyclic carbonate and urea type); (iv) a silicate in the range from about 1 wt% to about 20 wt% (e.g., sodium or potassium silicates such as sodium disilicate, sodium meta-silicate and crystalline phyllosilicates); (v) an inorganic bleach (e.g., perhydrate salts such as perborate, percarbonate, perphosphate, persulfate and persilicate salts) and/or an organic bleach (e.g., organic peroxyacids such as diacyl- and tetraacylperoxides, especially diperoxydodecanedioic acid, diperoxytetradecanedioic acid, and diperoxyhexadecanedioic acid); (vi) a bleach activator (e.g., organic peracid precursors in the range from about 0.1 wt% to about 10 wt%) and/or bleach catalyst (e.g., manganese triazacyclononane and related complexes; Co, Cu, Mn, and Fe bispyridylamine and related complexes; and pentamine acetate cobalt(III) and related complexes); (vii) a metal care agent in the range from about 0.1 wt% to 5 wt% (e.g., benzatriazoles, metal salts and complexes, and/or silicates); and/or (viii) any active enzyme disclosed herein in the range from about 0.01 to 5.0 mg of active enzyme per gram of automatic dishwashing detergent composition, and an enzyme stabilizer component (e.g., oligosaccharides, polysaccharides, and inorganic divalent metal salts).

Compositions comprising cellulose as disclosed herein can be in the form of an oral care composition. Examples of oral care compositions include dentifrices,

toothpaste, mouth wash, mouth rinse, chewing gum, and edible strips that provide some form of oral care (e.g., treatment or prevention of cavities [dental caries], gingivitis, plaque, tartar, and/or periodontal disease). An oral care composition can also be for treating an "oral surface", which encompasses any soft or hard surface within the oral cavity including surfaces of the tongue, hard and soft palate, buccal mucosa, gums and dental surfaces. A "dental surface" herein is a surface of a natural tooth or a hard surface of artificial dentition including a crown, cap, filling, bridge, denture, or dental implant, for example.

An oral care composition herein can comprise about 0.01-15.0 wt% (e.g., ~0.1-10 wt% or ~0.1-5.0 wt%, ~0.1-2.0 wt%) of one or more cellulose materials disclosed herein, for example. One or more cellulose materials herein comprised in an oral care composition can sometimes be provided therein as a thickening agent and/or dispersion agent, which may be useful to impart a desired consistency and/or mouth feel to the composition. One or more other thickening or dispersion agents can also be provided in an oral care composition herein, such as a carboxyvinyl polymer, carrageenan (e.g., L-carrageenan), natural gum (e.g., karaya, xanthan, gum arabic, tragacanth), colloidal magnesium aluminum silicate, or colloidal silica, for example.

An oral care composition herein may be a toothpaste or other dentifrice, for example. Such compositions, as well as any other oral care composition herein, can additionally comprise, without limitation, one or more of an anticaries agent, antimicrobial or antibacterial agent, anticalculus or tartar control agent, surfactant, abrasive, pH-modifying agent, foam modulator, humectant, flavorant, sweetener, pigment/colorant, whitening agent, and/or other suitable components. Examples of oral care compositions to which one or more cellulose materials herein can be added are disclosed in U.S. Patent Appl. Publ. Nos. 2006/0134025, 2002/0022006 and 2008/0057007, which are incorporated herein by reference.

An anticaries agent herein can be an orally acceptable source of fluoride ions. Suitable sources of fluoride ions include fluoride, monofluorophosphate and fluorosilicate salts as well as amine fluorides, including olaflur (N'-octadecyltrimethylendiamine-N,N,N'- tris(2-ethanol)-dihydrofluoride), for example. An anticaries agent can be present in an amount providing a total of about 100-20000 ppm, about 200-5000 ppm, or about 500-2500 ppm, fluoride ions to the composition, for example. In oral care compositions in which sodium fluoride is the sole source of

fluoride ions, an amount of about 0.01-5.0 wt%, about 0.05-1.0 wt%, or about 0.1-0.5 wt%, sodium fluoride can be present in the composition, for example.

An antimicrobial or antibacterial agent suitable for use in an oral care composition herein includes, for example, phenolic compounds (e.g., 4-allylcatechol; p-hydroxybenzoic acid esters such as benzylparaben, butylparaben, ethylparaben, methylparaben and propylparaben; 2-benzylphenol; butylated hydroxyanisole; butylated hydroxytoluene; capsaicin; carvacrol; creosol; eugenol; guaiacol; halogenated bisphenolics such as hexachlorophene and bromochlorophene; 4-hexylresorcinol; 8-hydroxyquinoline and salts thereof; salicylic acid esters such as menthyl salicylate, methyl salicylate and phenyl salicylate; phenol; pyrocatechol; salicylanilide; thymol; halogenated diphenylether compounds such as triclosan and triclosan monophosphate), copper (II) compounds (e.g., copper (II) chloride, fluoride, sulfate and hydroxide), zinc ion sources (e.g., zinc acetate, citrate, gluconate, glycinate, oxide, and sulfate), phthalic acid and salts thereof (e.g., magnesium monopotassium phthalate), hexetidine, octenidine, sanguinarine, benzalkonium chloride, domiphen bromide, alkylpyridinium chlorides (e.g. cetylpyridinium chloride, tetradecylpyridinium chloride, N-tetradecyl-4-ethylpyridinium chloride), iodine, sulfonamides, bisbiguanides (e.g., alexidine, chlorhexidine, chlorhexidine digluconate), piperidino derivatives (e.g., delmopinol, octapinol), magnolia extract, grapeseed extract, rosemary extract, menthol, geraniol, citral, eucalyptol, antibiotics (e.g., augmentin, amoxicillin, tetracycline, doxycycline, minocycline, metronidazole, neomycin, kanamycin, clindamycin), and/or any antibacterial agents disclosed in U.S. Patent No. 5776435, which is incorporated herein by reference. One or more antimicrobial agents can optionally be present at about 0.01-10 wt% (e.g., 0.1-3 wt%), for example, in the disclosed oral care composition.

An anticalculus or tartar control agent suitable for use in an oral care composition herein includes, for example, phosphates and polyphosphates (e.g., pyrophosphates), polyaminopropanesulfonic acid (AMPS), zinc citrate trihydrate, polypeptides (e.g., polyaspartic and polyglutamic acids), polyolefin sulfonates, polyolefin phosphates, diphosphonates (e.g.,azacycloalkane-2,2-diphosphonates such as azacycloheptane-2,2-diphosphonic acid), N-methyl azacyclopentane-2,3-diphosphonic acid, ethane-1-hydroxy-1,1-diphosphonic acid (EHDP), ethane-1-amino-1,1-diphosphonate, and/or phosphonoalkane carboxylic acids and salts thereof (e.g., their alkali metal and ammonium salts). Useful inorganic phosphate and polyphosphate salts include, for

example, monobasic, dibasic and tribasic sodium phosphates, sodium tripolyphosphate, tetrapolyphosphate, mono-, di-, tri- and tetra-sodium pyrophosphates, disodium dihydrogen pyrophosphate, sodium trimetaphosphate, sodium hexametaphosphate, or any of these in which sodium is replaced by potassium or ammonium. Other useful anticalculus agents in certain embodiments include anionic polycarboxylate polymers (e.g., polymers or copolymers of acrylic acid, methacrylic, and maleic anhydride such as polyvinyl methyl ether/maleic anhydride copolymers). Still other useful anticalculus agents include sequestering agents such as hydroxycarboxylic acids (e.g., citric, fumaric, malic, glutaric and oxalic acids and salts thereof) and aminopolycarboxylic acids (e.g., EDTA). One or more anticalculus or tartar control agents can optionally be present at about 0.01-50 wt% (e.g., about 0.05-25 wt% or about 0.1-15 wt%), for example, in the disclosed oral care composition.

A surfactant suitable for use in an oral care composition herein may be anionic, non-ionic, or amphoteric, for example. Suitable anionic surfactants include, without limitation, water-soluble salts of C_{8-20} alkyl sulfates, sulfonated monoglycerides of C_{8-20} fatty acids, sarcosinates, and taurates. Examples of anionic surfactants include sodium lauryl sulfate, sodium coconut monoglyceride sulfonate, sodium lauryl sarcosinate, sodium lauryl isoethionate, sodium laureth carboxylate and sodium dodecyl benzenesulfonate. Suitable non-ionic surfactants include, without limitation, poloxamers, polyoxyethylene sorbitan esters, fatty alcohol ethoxylates, alkylphenol ethoxylates, tertiary amine oxides, tertiary phosphine oxides, and dialkyl sulfoxides. Suitable amphoteric surfactants include, without limitation, derivatives of C_{8-20} aliphatic secondary and tertiary amines having an anionic group such as a carboxylate, sulfate, sulfonate, phosphate or phosphonate. An example of a suitable amphoteric surfactant is cocoamidopropyl betaine. One or more surfactants are optionally present in a total amount of about 0.01-10 wt% (e.g., about 0.05-5.0 wt% or about 0.1-2.0 wt%), for example, in the disclosed oral care composition.

An abrasive suitable for use in an oral care composition herein may include, for example, silica (e.g., silica gel, hydrated silica, precipitated silica), alumina, insoluble phosphates, calcium carbonate, and resinous abrasives (e.g., a urea-formaldehyde condensation product). Examples of insoluble phosphates useful as abrasives herein are orthophosphates, polymetaphosphates and pyrophosphates, and include dicalcium orthophosphate dihydrate, calcium pyrophosphate, beta-calcium pyrophosphate,

tricalcium phosphate, calcium polymetaphosphate and insoluble sodium polymetaphosphate. One or more abrasives are optionally present in a total amount of about 5-70 wt% (e.g., about 10-56 wt% or about 15-30 wt%), for example, in the disclosed oral care composition. The average particle size of an abrasive in certain embodiments is about 0.1-30 microns (e.g., about 1-20 microns or about 5-15 microns).

An oral care composition in certain embodiments may comprise at least one pH-modifying agent. Such agents may be selected to acidify, make more basic, or buffer the pH of a composition to a pH range of about 2-10 (e.g., pH ranging from about 2-8, 3-9, 4-8, 5-7, 6-10, or 7-9). Examples of pH-modifying agents useful herein include, without limitation, carboxylic, phosphoric and sulfonic acids; acid salts (e.g., monosodium citrate, disodium citrate, monosodium malate); alkali metal hydroxides (e.g. sodium hydroxide, carbonates such as sodium carbonate, bicarbonates, sesquicarbonates); borates; silicates; phosphates (e.g., monosodium phosphate, trisodium phosphate, pyrophosphate salts); and imidazole.

A foam modulator suitable for use in an oral care composition herein may be a polyethylene glycol (PEG), for example. High molecular weight PEGs are suitable, including those having an average molecular weight of about 200000-7000000 (e.g., about 500000-5000000 or about 1000000-2500000), for example. One or more PEGs are optionally present in a total amount of about 0.1-10 wt% (e.g. about 0.2-5.0 wt% or about 0.25-2.0 wt%), for example, in the disclosed oral care composition.

An oral care composition in certain embodiments may comprise at least one humectant. A humectant in certain embodiments may be a polyhydric alcohol such as glycerin, sorbitol, xylitol, or a low molecular weight PEG. Most suitable humectants also may function as a sweetener herein. One or more humectants are optionally present in a total amount of about 1.0-70 wt% (e.g., about 1.0-50 wt%, about 2-25 wt%, or about 5-15 wt%), for example, in the disclosed oral care composition.

A natural or artificial sweetener may optionally be comprised in an oral care composition herein. Examples of suitable sweeteners include dextrose, sucrose, maltose, dextrin, invert sugar, mannose, xylose, ribose, fructose, levulose, galactose, corn syrup (e.g., high fructose corn syrup or corn syrup solids), partially hydrolyzed starch, hydrogenated starch hydrolysate, sorbitol, mannitol, xylitol, maltitol, isomalt, aspartame, neotame, saccharin and salts thereof, dipeptide-based intense sweeteners,

and cyclamates. One or more sweeteners are optionally present in a total amount of about 0.005-5.0 wt%, for example, in the disclosed oral care composition.

A natural or artificial flavorant may optionally be comprised in an oral care composition herein. Examples of suitable flavorants include vanillin; sage; marjoram; parsley oil; spearmint oil; cinnamon oil; oil of wintergreen (methylsalicylate); peppermint oil; clove oil; bay oil; anise oil; eucalyptus oil; citrus oils; fruit oils; essences such as those derived from lemon, orange, lime, grapefruit, apricot, banana, grape, apple, strawberry, cherry, or pineapple; bean- and nut-derived flavors such as coffee, cocoa, cola, peanut, or almond; and adsorbed and encapsulated flavorants. Also encompassed within flavorants herein are ingredients that provide fragrance and/or other sensory effect in the mouth, including cooling or warming effects. Such ingredients include, without limitation, menthol, menthyl acetate, menthyl lactate, camphor, eucalyptus oil, eucalyptol, anethole, eugenol, cassia, oxanone, Irisone®, propenyl guaiethol, thymol, linalool, benzaldehyde, cinnamaldehyde, N-ethyl-p-menthan-3-carboxamine, N,2,3-trimethyl-2-isopropylbutanamide, 3-(1-menthoxy)-propane-1,2-diol, cinnamaldehyde glycerol acetal (CGA), and menthone glycerol acetal (MGA). One or more flavorants are optionally present in a total amount of about 0.01-5.0 wt% (e.g., about 0.1-2.5 wt%), for example, in the disclosed oral care composition.

An oral care composition in certain embodiments may comprise at least one bicarbonate salt. Any orally acceptable bicarbonate can be used, including alkali metal bicarbonates such as sodium or potassium bicarbonate, and ammonium bicarbonate, for example. One or more bicarbonate salts are optionally present in a total amount of about 0.1-50 wt% (e.g., about 1-20 wt%), for example, in the disclosed oral care composition.

An oral care composition in certain embodiments may comprise at least one whitening agent and/or colorant. A suitable whitening agent is a peroxide compound such as any of those disclosed in U.S. Patent No. 8540971, which is incorporated herein by reference. Suitable colorants herein include pigments, dyes, lakes and agents imparting a particular luster or reflectivity such as pearling agents, for example. Specific examples of colorants useful herein include talc; mica; magnesium carbonate; calcium carbonate; magnesium silicate; magnesium aluminum silicate; silica; titanium dioxide; zinc oxide; red, yellow, brown and black iron oxides; ferric ammonium ferrocyanide; manganese violet; ultramarine; titaniated mica; and bismuth oxychloride.

One or more colorants are optionally present in a total amount of about 0.001-20 wt% (e.g., about 0.01-10 wt% or about 0.1-5.0 wt%), for example, in the disclosed oral care composition.

Additional components that can optionally be included in an oral composition herein include one or more enzymes (above), vitamins, and anti-adhesion agents, for example. Examples of vitamins useful herein include vitamin C, vitamin E, vitamin B5, and folic acid. Examples of suitable anti-adhesion agents include solbrol, ficin, and quorum-sensing inhibitors.

In certain embodiments of the present disclosure, a composition comprising cellulose can be a film or coating.

A film or coating can be a dried film or coating in some aspects, comprising less than about 3, 2, 1, 0.5, or 0.1 wt% water, for example. The amount of cellulose comprised in a film or coating herein can be about, or at least about, 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19, 20, 21, 22, 23, 24, 25, 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, 40, 41, 42, 43, 44, 45, 46, 47, 48, 49, 50, 51, 52, 53, 55, 56, 57, 58, 59, 60, 61, 62, 63, 64, 65, 66, 67, 68, 69, 70, 71, 72, 73, 74, 75, 76, 77, 78, 79, 80, 81, 82, 83, 84, 85, 86, 87, 88, 89, 90, 91, 92, 93, 94, 95, 96, 97, 98, 99, 99.5, or 99.9 wt%, for example.

A film or coating herein can have a uniform thickness of at least about 4 nm, for instance. Such thickness in other aspects can be about, or at least about, 4.0, 4.5, 5.0, 5.5, 6.0, 7.0, 8.0, 9.0, 10, 25, 50, 100, 250, 500, 750, 1000, 1500, 2000, 2500, 3000, 3500, 4000, 4500, or 5000 nm. Uniform thickness characterizes, for example, a contiguous area that (i) is at least 20% of the total film/coating area, and (ii) has a standard deviation of thickness of less than about 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 20, 30, 40, or 50 nm. A film or coating herein can be characterized as thin in some aspects.

A film or coating herein can exhibit low permeability to, or is impermeable to, an aqueous composition, lipophilic composition, or gaseous composition, for example. A film or coating can be characterized as being of low permeability to a particular substance if the film/coating permeability to the substance is below a threshold value commonly assigned in the art of interest. To illustrate, the threshold value for styrene permeability in the SMC (super-multicoated) release film field is 200×10^{-9} g cm/cm^2/h, such as measured using the method described in *American Institute of Chemical*

Engineer, 53rd National Meeting, Preprint No.32d (Bixler and Michaels, 1964). The threshold value for a particular substance (e.g., aqueous composition, a lipophilic composition, or a gaseous composition) is a function of the technical field of concern. A film or coating can be characterized as being impermeable to an aqueous composition, lipophilic composition, and/or a gaseous composition if it does not permit passage of such composition over an extended period of time (e.g., at least 1, 2, 3, or more days).

A film or coating herein can exhibit various degrees of transparency as desired. For example, a film/coating can be highly transparent (e.g., high optical transparency, and/or low haze). Optical transparency as used herein can refer to a film or coating allowing at least about 10-99% light transmission, or at least about 50%, 60%, 70%, 80%, 90%, 95%, 96%, 97%, 98%, or 99% light transparency. High transparency can optionally refer to a film/coating having at least about 90% optical transmittance. Transparency of a film or coating herein can be measured following test ASTM D 1746 (2009, *Standard Test Method for Transparency of Plastic Sheeting*, ASTM International, West Conshohocken, PA), for example, which is incorporated herein by reference.

An aqueous composition or lipophilic composition is preferably in liquid form as applied to a film/coating disclosed herein. An aqueous composition can be as disclosed elsewhere herein, for example. Examples of lipophilic compositions herein include non-aqueous liquids such as oil, organic liquids that do not exhibit hydrogen bonding (e.g., aliphatic and/or aromatic hydrocarbons such as hexane, octane, benzene and other alkyl benzenes), or organic liquids that exhibit only moderate hydrogen bonding (e.g., alkyl and aryl esters, ketones and ethers such as ethyl acrylate, butyl acrylate, dimethyl ketone, butyl glycol ether, diglycol ethyl ether). Examples of gaseous compositions herein include air, water vapor, and gases comprising at least .001%, .01%, .1%, 1%, 10%, 25%, 50%, 75%, 90%, 95%, 100% of one or more of the following: nitrogen (N_2), oxygen (O_2), argon (Ar), carbon dioxide (CO_2), neon (Ne), helium (He), methane (CH_4), krypton (Kr), hydrogen (H_2), nitrous oxide (N_2O), radon (Rn), xenon (Xe), ozone (O_3), carbon monoxide (CO), sulfur dioxide (SO_2), nitrogen dioxide (NO_2), ammonia (NH_3).

A film or coating herein can be on a material such as paper. Types of paper to which a film/coating herein can be applied include offset, vellum bristol, ledger, cover, index, tag, railroad board, reply card, forms bond, laser bond, OCR bond, MICR bond, safety bond, carbonless CB, carbonless CFB, carbonless CF, newsprint, and kraft, for example. The paper weight herein can be rated at about 10-150 pound (e.g., 10, 20,

21, 22, 23, 24, 25, 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, 40, 50, 60, 70, 80, 90, 100, 110, 120, 130, 140, or 150 pound), for example. Further ingredients and formulation information regarding preparation of paper coatings are disclosed in U.S. Patent No. 7625441, for example, which is incorporated herein by reference. Examples of other materials that can be coated with a film or coating herein include plastic or rubber material, plant material (e.g., fruit, vegetables, seeds), pharmaceuticals (e.g., pills, tablets), glass material, ceramic material, metal material, and electrical equipment/devices/components.

Embodiments of the present disclosure further concern a method for increasing the viscosity of an aqueous composition. This method comprises contacting cellulose with the aqueous composition, wherein the cellulose is insoluble in the aqueous composition and has:

(i) a weight-average degree of polymerization (DP_w) of about 10 to about 1000, and

(ii) a cellulose II crystal structure.

The contacting step in this method results in increasing the viscosity of the aqueous composition, in comparison to the viscosity of the aqueous composition before the contacting step.

An aqueous composition can be as disclosed elsewhere herein such as water (e.g., de-ionized water), an aqueous solution, or a colloidal dispersion, for example. The viscosity of an aqueous composition before the contacting step, measured at about 20-25 °C, can be about 0-10000 cPs (or any integer between 0-10000 cPs), for example. It should be apparent that very large percent increases in viscosity can be obtained with the disclosed method when the aqueous composition has little viscosity before the contacting step.

Contacting cellulose of the present disclosure with an aqueous composition increases the viscosity of the aqueous composition in certain embodiments. This increase in viscosity can be an increase of at least about 1%, 10%, 100%, 1000%, 100000%, or 1000000% (or any integer between 1% and 1000000%), for example, compared to the viscosity of the aqueous composition before the contacting step. It should be apparent that very large percent increases in viscosity can be obtained with the disclosed method when the aqueous composition has little to no viscosity before the

contacting step. An increase in viscosity can be determined, for example, by comparing the viscosity of the aqueous composition obtained by the method (i.e., after the contacting step) with the viscosity of the aqueous composition as it had existed before the method (i.e., before the contacting step).

Contacting cellulose of the present disclosure with an aqueous composition increases the shear thinning behavior of the aqueous composition in certain embodiments. Thus, the cellulose rheologically modifies the aqueous composition in these embodiments. The increase in shear thinning behavior can be an increase of at least about 1%, 10%, 100%, 1000%, 100000%, or 1000000% (or any integer between 1% and 1000000%), for example, compared to the shear thinning behavior of the aqueous composition before the contacting step. It should be apparent that very large percent increases in rheologic modification can be obtained with the disclosed method when the aqueous composition has little or no rheologic behavior before the contacting step.

The contacting step in a method for increasing the viscosity of an aqueous composition can be performed by mixing any cellulose of the present disclosure in the aqueous composition by any means known in the art. For example, mixing can be performed manually or with a machine (e.g., industrial mixer or blender, orbital shaker, stir plate, homogenizer, sonicator, bead mill). Mixing can comprise a homogenization step in certain embodiments. Homogenization (as well as any other type of mixing) can be performed for about 5 to 60, 5 to 30, 10 to 60, 10 to 30, 5 to 15, or 10 to 15 seconds (or any integer between 5 and 60 seconds), or longer periods of time as necessary to mix cellulose with the aqueous composition. A homogenizer can be used at about 5000 to 30000 rpm, 10000 to 30000 rpm, 15000 to 30000 rpm, 15000 to 25000 rpm, or 20000 rpm (or any integer between 5000 and 30000 rpm), for example.

After cellulose herein is mixed with or dissolved into an aqueous composition, the resulting aqueous composition may be filtered, or may not be filtered. For example, an aqueous composition prepared with a homogenization step may or may not be filtered.

Certain embodiments of the above method can be used to prepare an aqueous composition disclosed herein, such as any food product, pharmaceutical product, household product, personal care product, or industrial product disclosed herein.

Cellulose used in a viscosity modification method can have any of the features disclosed herein. For example, any of the features of water-insolubility, DP_w (e.g., DP_w

of 10-30) and/or M_w, glycosidic linkage profile, backbone structure (e.g., linearity), cellulose II structural content, and/or solubility in certain non-aqueous compositions as disclosed elsewhere herein can characterize cellulose used in various embodiments of a viscosity modification method.

Embodiments of the present disclosure further concern a method of treating a material. This material treatment method comprises:

(a) contacting a material with an aqueous composition comprising cellulose, wherein the cellulose is insoluble in the aqueous composition and has:

(i) a weight-average degree of polymerization (DP_w) of about 10 to about 1000, and

(ii) a cellulose II crystal structure; and

(b) drying the aqueous composition,

wherein the drying step leaves a deposit of the cellulose on the surface of the material. This method in certain aspects can be characterized as a method of coating a material (a coating method).

An aqueous composition for use in a coating method in certain aspects is preferably a colloidal dispersion of cellulose as disclosed elsewhere herein. Such a colloidal dispersion can comprise about, or at least about, 0.01, 0.05, 0.1, 0.2, 0.3, 0.4, 0.5, 0.6, 0.7, 0.8, 0.9, 1.0, 1.2, 1.4, 1.6, 1.8, 2.0, 2.2, 2.5, 3.0, 3.5, 4.0, 4.5, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19, 20, 21, 22, 23, 24, 25, 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, or 40 wt% of cellulose as presently disclosed.

A contacting step of a coating method herein can comprise applying a colloidal dispersion comprising cellulose at any of the above weight percentages, for example, to a material. Such application can be to provide onto a material a coat or layer of the colloidal dispersion at a thickness (not yet dried) of about 1, 2, 3, 4, , 5, 6, 7, 8, 9, or 10 mil (1 mil = .001 inch), for example. All of, or a portion of (e.g., at least about 1-99%), the surface(s) of a material can be coated as desired. Coating a material can be for purposes of preparing a removable film in certain embodiments, in which case a colloidal dispersion can be applied in any manner of film casting known in the art (e.g., comprising using a blade coater or casting rod).

After applying an aqueous material such as a colloidal dispersion of cellulose to a material, the aqueous material is dried. Such drying can be performed by allowing the

aqueous material to dry out at room temperature (20-25 °C) (i.e., air drying), or at any other suitable drying temperature such as at any temperature between 15-70 °C, for example. Drying can be done with or without application of forced air or vacuum.

Cellulose used in a coating method can have any of the features disclosed herein. For example, any of the features of water-insolubility, DP_w (e.g., DP_w of 10-30) and/or M_w, glycosidic linkage profile, backbone structure (e.g., linearity), and/or cellulose II structural content as disclosed elsewhere herein can characterize cellulose used in various embodiments of a coating method.

Material that can be coated in a method herein includes paper (e.g., any type as disclosed elsewhere herein), plastic or rubber material, plant material (e.g., fruit, vegetables, seeds), pharmaceuticals (e.g., pills, tablets), glass material, ceramic material, metal material, and electrical equipment/devices/components.

A deposit of cellulose on the surface of a material subject to a coating method herein can be characterized as a film, coating, or film coating in certain embodiments. Such a coating can have any of the features disclosed herein. For example, a coating can (i) have a uniform thickness (e.g., at least about 4 nm), (ii) have low permeability, or be impermeable to, certain compositions (e.g., liquids), and/or (iii) be optically transparent (e.g., highly transparent), as disclosed elsewhere herein.

A film or coating resulting from a coating method herein can is dry. Typically, a dried film or coating has less than 3, 2, 1, 0.5, or 0.1 wt% water comprised therein. The amount of cellulose of the present disclosure in a dried film or coating can be about, or at least about, 1, 2, 3, 4, 5, 6, 7, 8, 9, 10, 11, 12, 13, 14, 15, 16, 17, 18, 19, 20, 21, 22, 23, 24, 25, 26, 27, 28, 29, 30, 31, 32, 33, 34, 35, 36, 37, 38, 39, 40, 41, 42, 43, 44, 45, 46, 47, 48, 49, 50, 51, 52, 53, 55, 56, 57, 58, 59, 60, 61, 62, 63, 64, 65, 66, 67, 68, 69, 70, 71, 72, 73, 74, 75, 76, 77, 78, 79, 80, 81, 82, 83, 84, 85, 86, 87, 88, 89, 90, 91, 92, 93, 94, 95, 96, 97, 98, 99, 99.5, or 99.9 wt%, for example.

Non-limiting examples of compositions and methods disclosed herein include:

1. A composition comprising cellulose, wherein the cellulose:

(i) has a weight-average degree of polymerization (DP_w) of about 10 to about 1000,

(ii) has a cellulose II crystal structure, and

(iii) is insoluble in an aqueous composition.

2. The composition of embodiment 1, wherein the DP_w of the cellulose is about 10 to about 100.

3. The composition of embodiment 1 or 2, wherein the cellulose is a product of a cellodextrin phosphorylase enzyme comprising an amino acid sequence that is at least 90% identical to SEQ ID NO:2 or SEQ ID NO:6, wherein the substrates for the enzyme comprise cellodextrin and glucose-1-phosphate.

4. The composition of embodiment 3, wherein the cellulose as produced by the enzyme has not been subjected to a mercerization or derivatization process.

5. The composition of embodiment 1, 2, 3, or 4, wherein the composition is a film or coating.

6. The composition of embodiment 5, wherein the film or coating has a uniform thickness of at least about 4 nm.

7. The composition of embodiment 5 or 6, wherein the film or coating exhibits low permeability to, or is impermeable to, an aqueous composition, lipophilic composition, or gaseous composition.

8. The composition of embodiment 5, 6, or 7, wherein the film or coating is on paper.

9. The composition of embodiment 1, 2, 3, or 4, wherein the composition is an aqueous composition, optionally having a viscosity of at least about 100 cPs.

10. The composition of embodiment 9, wherein the aqueous composition is a colloidal dispersion.

11. The composition of embodiment 9 or 10, wherein the concentration of the cellulose in the aqueous composition is less than about 10 wt%.

12. The composition of any one of embodiments 1-11, wherein the composition is a food product, personal care product, pharmaceutical product, household product, or industrial product.

13. The composition of embodiment 1, 2, 3, or 4, wherein the cellulose is soluble in a solvent comprising DMSO and/or DMAc.

14. A method for increasing the viscosity of an aqueous composition, the method comprising:

contacting cellulose with the aqueous composition, wherein the cellulose is insoluble in the aqueous composition and has:

(i) a weight-average degree of polymerization (DP_w) of about 10 to about 1000, and

(ii) a cellulose II crystal structure,

wherein the viscosity of the aqueous composition is increased by the cellulose compared to the viscosity of the aqueous composition before the contacting step.

15. The method of embodiment 14, wherein the shear thinning behavior of the aqueous composition is increased by the cellulose compared to the shear thinning behavior of the aqueous composition before the contacting step.

16. A method of treating a material, the method comprising:

(a) contacting a material with an aqueous composition comprising cellulose, wherein the cellulose is insoluble in the aqueous composition and has:

(i) a weight-average degree of polymerization (DP_w) of about 10 to about 1000, and

(ii) a cellulose II crystal structure; and

(b) drying the aqueous composition,

wherein the drying step leaves a deposit of the cellulose on the surface of the material.

EXAMPLES

The present disclosure is further exemplified in the following Examples. It should be understood that these Examples, while indicating certain preferred aspects herein, are given by way of illustration only. From the above discussion and these Examples, one skilled in the art can ascertain the essential characteristics of the disclosed embodiments, and without departing from the spirit and scope thereof, can make various changes and modifications to adapt the disclosed embodiments to various uses and conditions.

EXAMPLE 1

Expression and Analysis of a *Vibrio ruber* Cellodextrin Phosphorylase

This Example describes expression of a putative *Vibrio ruber* cellodextrin phosphorylase enzyme in *E. coli.* Also, this Example demonstrates that this enzyme is indeed a cellodextrin phosphorylase through analysis of enzyme specific activity.

A putative cellodextrin phosphorylase, VruCdp1 (also referred to herein as "CRC03362-VruCdp1"), was identified in *Vibrio ruber* DSM14379. The nucleic acid

sequence encoding VruCdp1 was predicted based on a genomic sequence, and is presented as SEQ ID NO:1. The amino acid sequence of VruCdp1 encoded by SEQ ID NO:1 is presented as SEQ ID NO:2.

Putative VruCdp1 cellodextrin phosphorylase was next heterologously expressed in *E. coli*, as follows. A polynucleotide sequence encoding VruCdp1 was codon-optimized for expression in *E. coli*. This sequence (SEQ ID NO:3) was inserted into the pET30a (Novagen) expression vector at the NdeI and XhoI sites by Generay (Shanghai, China), resulting in plasmid pZZH634. SEQ ID NO:3 contains the codon-optimized open reading frame as well as sequence encoding two extra amino acids (Leu-Glu) and a 6x His-tag at the C-terminus. The amino acid sequence encoded by SEQ ID NO:3 is presented as SEQ ID NO:4. The pZZH634 plasmid was transformed into *E. coli* strain BL21(DE3) (Novagen), which was plated on LB agar plates supplemented with 50 ppm kanamycin. Correctly transformed colonies, as confirmed by PCR and sequencing, were inoculated into 5 ml LB medium supplemented with 50 ppm kanamycin and cultivated in 37 °C with shaking for about 16 hours. About 1 mL of the culture was then inoculated into 25 mL LB medium supplemented with 50 ppm kanamycin and cultivated in 37 °C with shaking until the OD_{600} reached about 0.4-1.0. IPTG was then added into the culture at a final concentration at 100 mM to induce VruCdp1 expression. The culture was then cultivated at 16 °C for 12-16 hours.

After this period of inducing VruCdp1 expression, the *E. coli* cells were pelleted, resuspended in lysis buffer (50 mM Tris pH 7.0, 500 mM NaCl, 10% glycerol, 0.1% Tween-20), and lysed on ice via ultra-sonication for 10 min (35% power, 20 min, 2 sec on/2 sec off) (SCIENT2-II D, Ningbo Scientz Biotechnology Co., Ltd). The lysate was cleared by centrifugation at 13000 rpm for 30 min (BECKMAN COULTER, Avanti™ JE). The clarified lysate was applied onto a His Trap™ HP (5 mL) (GE Healthcare) pre-equilibrated with 50 mM Tris pH 7.0, 500 mM NaCl, and 10% glycerol. The target protein (VruCdp1) was eluted from the column with a linear gradient from 0 to 250 mM imidazole in equilibration buffer. The fractions containing the target protein were pooled, concentrated and exchanged to equilibration buffer using 10K Amicon Ultra devices, and stored in 40% glycerol at -20 °C until usage.

The activity of VruCdp1 (isolated above) was measured using 10 mM G-1-P (Sigma G7000, α-D-Glucose 1-phosphate disodium salt hydrate) and 5 mM cellobiose (Sigma C7252, D-(+)-cellobiose) as substrates. The assay was performed in 25 mM

Tris-HCl buffer, pH 7.0 at 37 °C for 10 minutes. Phosphorus release from the enzyme reaction was quantified using PiBlue™ reagent (BioAssay Systems, US). One unit of cellodextrin phosphorylase activity was defined as the amount of enzyme that releases 1 µmol of inorganic phosphorus per minute under the assay conditions. The specific activity of the isolated VruCdp1 was determined to be 18.4 units/mg. Based on this observation, VruCdp1 was determined to be a cellodextrin phosphorylase (EC 2.4.1.49) belonging to glycosyl hydrolase family 94 (GH94, CAZy number).

Thus, an enzyme comprising SEQ ID NO:2 (VruCdp1) was expressed, isolated and shown to have cellodextrin phosphorylase activity.

EXAMPLE 2

Expression and Analysis of a *Ruminococcus champanellensis* Cellodextrin Phosphorylase

This Example describes expression of a putative *Ruminococcus champanellensis* cellodextrin phosphorylase enzyme in *E. coli.* Also, this Example demonstrates that this enzyme is indeed a cellodextrin phosphorylase through analysis of enzyme specific activity.

A putative cellodextrin phosphorylase, RchCdp1 (also referred to herein as "CRC03359-RchCdp1"), was identified in *Ruminococcus champanellensis* 18P13. The nucleic acid sequence encoding RchCdp1 (positions 2373141 to 2375537 of GENBANK Accession No. NC_021039.1) is presented as SEQ ID NO:5. The amino acid sequence of RchCdp1 encoded by SEQ ID NO:5 is presented as SEQ ID NO:6.

Putative RchCdp1 cellodextrin phosphorylase was next heterologously expressed in *E. coli*, as follows. A polynucleotide sequence encoding RchCdp1 was codon-optimized for expression in *E. coli*. This sequence (SEQ ID NO:7) was inserted into the pET30a (Novagen) expression vector at the NdeI and XhoI sites by Generay (Shanghai, China), resulting in plasmid pZZH631. SEQ ID NO:7 contains the codon-optimized open reading frame as well as sequence encoding two extra amino acids (Leu-Glu) and a 6x His-tag at the C-terminus. The amino acid sequence encoded by SEQ ID NO:7 is presented as SEQ ID NO:8. The pZZH631 plasmid was transformed into *E. coli* strain BL21(DE3) (Novagen), which was plated on LB agar plates supplemented with 50 ppm kanamycin. Correctly transformed colonies, as confirmed by PCR and sequencing, were inoculated into 5 ml LB medium supplemented with 50 ppm kanamycin and cultivated in 37 °C with shaking for about 16 hours. About 1 mL of

the culture was then inoculated into 25 mL LB medium supplemented with 50 ppm kanamycin and cultivated in 37 °C with shaking until the OD_{600} reached about 0.4-1.0. IPTG was then added into the culture at a final concentration at 100 mM to induce RchCdp1 expression. The culture was then cultivated at 16 °C for 12-16 hours.

After this period of inducing RchCdp1 expression, the *E. coli* cells were pelleted, resuspended in lysis buffer (50 mM Tris pH 7.0, 500 mM NaCl, 10% glycerol, 0.1% Tween-20), and lysed on ice via ultra-sonication for 10 min (35% power, 20 min, 2 sec on/2 sec off) (SCIENT2-II D, Ningbo Scientz Biotechnology Co., Ltd). The lysate was cleared by centrifugation at 13000 rpm for 30 min (BECKMAN COULTER, Avanti™ JE). The clarified lysate was applied onto a His Trap™ HP (5 mL) (GE Healthcare) pre-equilibrated with 50 mM Tris pH 7.0, 500 mM NaCl, and 10% glycerol. The target protein (RchCdp1) was eluted from the column with a linear gradient from 0 to 250 mM imidazole in equilibration buffer. The fractions containing the target protein were pooled, concentrated and exchanged to equilibration buffer using 10K Amicon Ultra devices, and stored in 40% glycerol at -20 °C until usage.

The activity of RchCdp1 (isolated above) was measured using 10 mM G-1-P (Sigma G7000, α-D-Glucose 1-phosphate disodium salt hydrate) and 5 mM cellobiose (Sigma C7252, D-(+)-cellobiose) as substrates. The assay was performed in 25 mM Tris-HCl buffer, pH 7.0 at 37 °C for 10 minutes. Phosphorus release from the enzyme reaction was quantified using PiBlue™ reagent (BioAssay Systems, US). One unit of cellodextrin phosphorylase activity was defined as the amount of enzyme that releases 1 µmol of inorganic phosphorus per minute under the assay conditions. The specific activity of the isolated RchCdp1 was determined to be 15.4 units/mg. Based on this observation, RchCdp1 was determined to be a cellodextrin phosphorylase (EC 2.4.1.49) belonging to glycosyl hydrolase family 94 (GH94, CAZy number).

Thus, an enzyme comprising SEQ ID NO:6 (RchCdp1) was expressed, isolated and shown to have cellodextrin phosphorylase activity.

EXAMPLE 3

Using *V. ruber* and *R. champanellensis* Cellodextrin Phosphorylases to Produce Low Molecular Weight, Insoluble Cellulose

This Example describes using the cellodextrin phosphorylases described in Examples 1 and 2 to produce cellulose when applied in reactions containing G-1-P and cellodextrin.

A reaction comprising G-1-P and cellobiose in the presence of a *V. ruber* cellodextrin phosphorylase (VruCdp1, refer to Example 1) produced insoluble polysaccharide. To generate enough insoluble polysaccharide for analysis, a scale-up reaction was conducted by adding 1 g G-1-P, 0.25 g cellobiose, and 400 µg (~7.4 units) isolated VruCdp1 to a glass bottle containing 80 mL of 25 mM Tris buffer pH 7.0. The reaction was incubated overnight at 37 °C. Insoluble polysaccharide product was collected by centrifugation at 3000 rpm for 20 minutes. This material was determined to be low molecular weight cellulose (refer to Example 4 below).

A reaction comprising G-1-P and cellobiose in the presence of an *R. champanellensis* cellodextrin phosphorylase (RchCdp1, refer to Example 2) produced insoluble polysaccharide. To generate enough insoluble polysaccharide for analysis, a scale-up reaction was conducted by adding 1 g G-1-P, 0.25 g cellobiose, and 400 µg (~6.2 units) isolated RchCdp1 to a glass bottle containing 80 mL of 25 mM Tris buffer pH 7.0. The reaction was incubated overnight at 37 °C. Insoluble polysaccharide product was collected by centrifugation at 3000 rpm for 20 minutes. This material was determined to be low molecular weight cellulose (refer to Example 4 below).

Thus, enzymes comprising SEQ ID NO:2 (VruCdp1) or SEQ ID NO:6 (RchCdp1) produce low molecular weight, insoluble cellulose when provided in a reaction comprising G-1-P and cellodextrin (e.g., cellobiose) substrates. It is noteworthy that these enzymes had this particular cellulose synthesis activity, given that sixteen other cellodextrin phosphorylases that were similarly expressed and analyzed did not have this capability (data not shown).

EXAMPLE 4

Analysis of Insoluble Polysaccharides Produced by *V. ruber* and *R. champanellensis* Cellodextrin Phosphorylases

This Example describes various analyses of the insoluble polysaccharide products obtained in the reactions described in Example 3. These analyses indicate that the products comprise low molecular weight, insoluble cellulose.

^{1}H-NMR analysis was conducted on the insoluble materials produced by *V. ruber* and *R. champanellensis* cellodextrin phosphorylases (Example 3). Briefly, 13.8 mg of each sample was dissolved by stirring in 0.8 ml of DMSO-d6, 3 wt% LiCl for 1 hour at 60 °C. NMR was run on the dissolved samples using an AVANCE III HD NMR device equipped with a 5-mm CPC Q1 cryoprobe. This analysis indicated that the insoluble

materials are polymers of glucose with beta-1,4 linkage, which is the characteristic linkage of cellulose. Thus, the insoluble materials produced by *V. ruber* and *R. champanellensis* cellodextrin phosphorylases comprise insoluble cellulose.

Each insoluble cellulose material was further analyzed using triple-detector SEC (size exclusion chromatography) to determine its molecular weight (M_w). Briefly, each sample was dissolved at 0.1-0.3 wt% in DMSO, 2 wt% LiCl and run through SEC. The M_w for each sample was found to be about 3-4 kDa (DP_w ~18-24) (Table 2).

Table 2

Molecular Weight of Cellulose Produced by RchCdp1 and VruCdp1 Enzymes

Cellulose Product of:	Mn[a] (kDa)	Mp[b] (kDa)	Mw[c] (kDa)	Mz[d] (kDa)	DPw[e]	Calculated mass (μg)	IV[f] (mL/g)	Uncertainty in IV
RchCdp1	2.94	2.99	2.95	3	18.2	140.81	6.441	1.47%
VruCdp1	3.82	3.9	3.83	3.8	23.6	133.8	6.277	1.89%

[a] Mn, number average molecular weight.
[b] Mp, peak molecular weight.
[c] Mw, mass average molecular weight.
[d] Mz, z-average molecular weight.
[e] DPw, mass average degree of polymerization.
[f] IV, intrinsic viscosity.

Thus, the cellulose samples produced by each of the RchCdp1 and VruCdp1 enzymes were of much lower molecular weight compared to cellulose obtained from cotton, wood pulp and microbial sources.

The low molecular weight cellulose samples were readily soluble and filterable in DMSO/LiCl (preparations as provided for SEC analysis above) and DMAc/LiCl (5 wt% LiCl in DMAc) at room temperature. This is noteworthy, since cellulose obtained from wood pulp, for example, typically cannot be dissolved in DMSO/LiCl, and requires elevated temperatures (e.g., about 100 °C) and times (e.g., 1 or more days) to dissolve in DMAc/LiCl. Since there was a clear viscometer peak observed with each of the samples (data not shown), it appears that enzymatically produced low molecular weight cellulose molecules behave as rigid rods.

Both as-made (produced as in Example 3 and stored in water, but never dried) and dried cellulose material (as synthesized by both RchCdp1 and VruCdp1 enzymes) exhibited a reflection indicative of cellulose II crystal under wide angle X-ray scattering (WAXS) analysis, which is the most stable crystal form of cellulose. It is noteworthy that, although the as-made samples were provided in an abundance of water after

enzymatic production (98.5 wt% and 97.5 wt% water, respectively, for cellulose products of RchCdp1 and VruCdp1 enzymes), a clear reflection was still observed, superimposed to a broad amorphous diffraction from the water. This observation of cellulose II structure is interesting, since it is believed that cellulose II is typically obtained after cellulose has undergone certain chemical processing steps (e.g., mercerization; derivatization followed by recovery of non-derivatized cellulose) (Kroon-Batenburg and Kroon, *Glycoconjugate J.* 14:677-690). In contrast, the present Example demonstrates that cellulose as directly produced in reactions containing RchCdp1 and VruCdp1 enzymes has a cellulose II crystal structure, without application of any post-synthesis chemical treatments.

Atomic force microscopy (AFM) was used to analyze a thin film made from drying a colloidal dispersion of insoluble cellulose synthesized by either RchCdp1 or VruCdp1 enzymes. Briefly, a film was casted from a ~2 wt% dispersion of insoluble cellulose in water using a blade coater with a 3-mil thickness. The coated wet film was allowed to dry by slow water evaporation at room temperature. AFM analysis (FIGs. 1A and 1B) of dried coatings showed a unique morphology of sheets with a highly uniform thickness of about 5 nm and width of hundreds of nanometers. It is believed that such a two-dimensional, graphene-like cellulose coating has never previously been demonstrated. Typically rather, cellulose materials such as nano-crystalline cellulose and those from microbial sources form rod-like colloids, not two-dimensional flake-like structures. Flake-like two dimensional structures are contemplated to have a number of advantages. For example, cellulose material with such structural properties likely has enhanced oxygen- and/or water-barrier properties. Moreover, the highly crystalline nature of the cellulose materials provided herein should allow increased mechanical properties of traditional thermoplastic polymers.

The above-prepared colloidal dispersions of insoluble cellulose could easily be coated to yield highly transparent, continuous films. Such film had a very thin thickness ranging between 1 and 2 microns, with a roughness of about 300 nm (data not shown). Thus, the low molecular weight, insoluble cellulose material provided herein is contemplated to be useful in water-based coating systems that can enable a number of applications. Examples of such applications include oxygen- and water vapor-barrier coatings on packaging plastics, as well as edible coating on fruits and vegetables to increase product shelf life. Moreover, the disclosed coatings can be useful for seed

coating applications and for enabling active ingredient release in pharmaceutical compositions.

Colloidal dispersions in water containing 1.7-2.5 wt% of insoluble cellulose synthesized by either RchCdp1 or VruCdp1 enzymes were analyzed for their degree of viscosity. Briefly, a Brookfield rheometer was used to obtain viscosity versus shear rate data, where the viscosity was measured at 10 (1/s) shear rate from the curves. It was found that both colloidal dispersions exhibit high viscosity that was 10000 times higher than the viscosity of water (FIG. 2). Also, the dispersions exhibited shear thinning behavior (where viscosity decreases as a function of shear rate), which is desired in many thickening applications. It is noteworthy to have obtained such high viscosity levels, given that each insoluble cellulose sample was of low DPw (less than 25 DPw, Table 2). In fact, commercially available carboxymethyl-derivatized cellulose (water-soluble) required significantly higher DPw (about 1000 or higher) to increase viscosity in water to the same extent as the viscosity observed when using the insoluble cellulose samples provided herein (FIG. 3).

Thus, the insoluble polysaccharide materials produced by *V. ruber* and *R. champanellensis* cellodextrin phosphorylases comprise low molecular weight, insoluble cellulose. This cellulose has a DP_w of about 18-24 and exhibits a cellulose II crystal structure. The cellulose II crystal structure is not a result of chemical processing such as mercerization or derivatization/un-derivatization processes, but rather characterizes the insoluble cellulose material as it is directly produced enzymatically. The unique properties of the insoluble cellulose provided herein gives this material broad utility, such as use in viscosity- and rheology-modification applications, and film/barrier applications.

CLAIMS

What is claimed is:

1. A composition comprising cellulose, wherein said cellulose:
(i) has a weight-average degree of polymerization (DP_w) of about 10 to about 1000,
(ii) has a cellulose II crystal structure, and
(iii) is insoluble in an aqueous composition.

2. The composition of claim 1, wherein the DP_w of the cellulose is about 10 to about 100.

3. The composition of claim 1, wherein said cellulose is a product of a cellodextrin phosphorylase enzyme comprising an amino acid sequence that is at least 90% identical to SEQ ID NO:2 or SEQ ID NO:6, wherein the substrates for said enzyme comprise cellodextrin and glucose-1-phosphate.

4. The composition of claim 3, wherein the cellulose as produced by the enzyme has not been subjected to a mercerization or derivatization process.

5. The composition of claim 1, wherein the composition is a film or coating.

6. The composition of claim 5, wherein the film or coating has a uniform thickness of at least about 4 nm.

7. The composition of claim 5, wherein the film or coating exhibits low permeability to, or is impermeable to, an aqueous composition, lipophilic composition, or gaseous composition.

8. The composition of claim 5, wherein the film or coating is on paper.

9. The composition of claim 1, wherein the composition is an aqueous composition, optionally having a viscosity of at least about 100 cPs.

10. The composition of claim 9, wherein the aqueous composition is a colloidal dispersion.

11. The composition of claim 9, wherein the concentration of the cellulose in the aqueous composition is less than about 10 wt%.

12. The composition of claim 9, wherein the composition is a food product, personal care product, pharmaceutical product, household product, or industrial product.

13. The composition of claim 1, wherein the cellulose is soluble in a solvent comprising DMSO and/or DMAc.

14. A method for increasing the viscosity of an aqueous composition, the method comprising:
contacting cellulose with the aqueous composition, wherein said cellulose is insoluble in the aqueous composition and has:
(i) a weight-average degree of polymerization (DP_w) of about 10 to about 1000, and
(ii) a cellulose II crystal structure,
wherein the viscosity of the aqueous composition is increased by said cellulose compared to the viscosity of the aqueous composition before the contacting step.

15. The method of claim 14, wherein the shear thinning behavior of the aqueous composition is increased by said cellulose compared to the shear thinning behavior of the aqueous composition before the contacting step.

16. A method of treating a material, said method comprising:
(a) contacting a material with an aqueous composition comprising cellulose, wherein the cellulose is insoluble in the aqueous composition and has:
(i) a weight-average degree of polymerization (DP_w) of about 10 to about 1000, and
(ii) a cellulose II crystal structure; and

(b) drying the aqueous composition,

wherein the drying step leaves a deposit of said cellulose on the surface of the material.

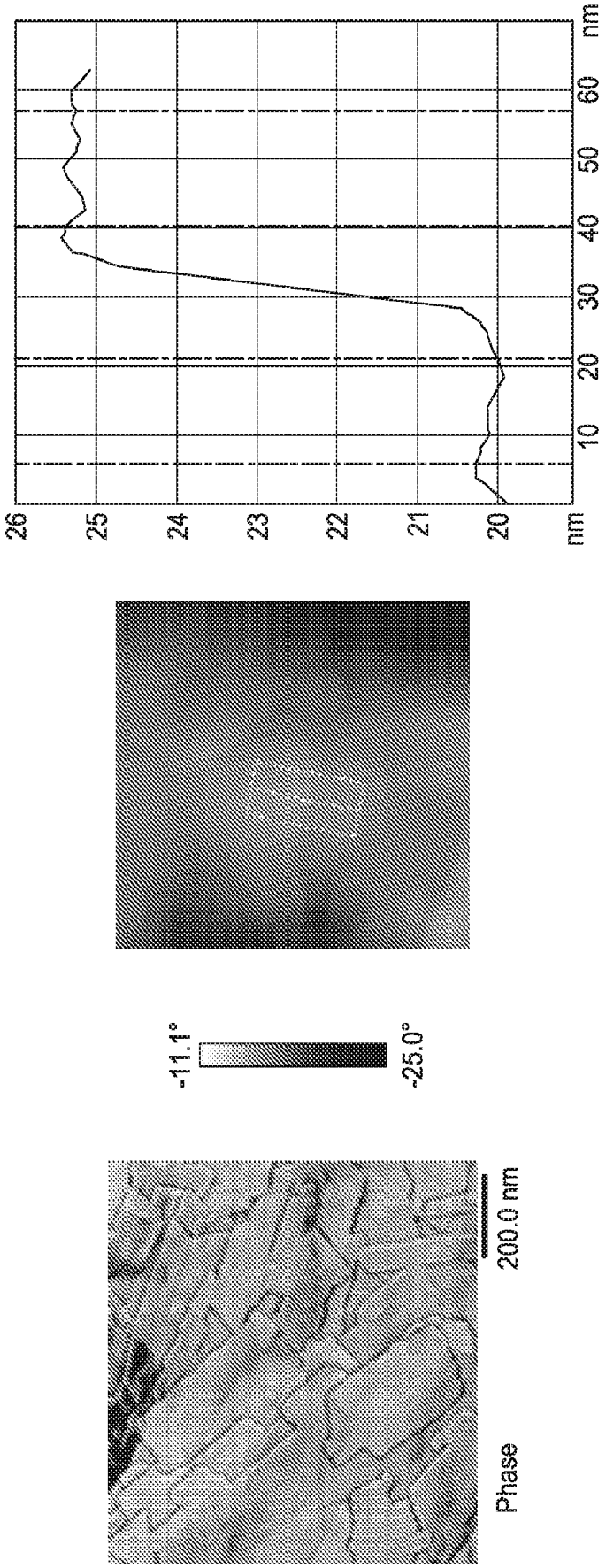


FIG. 1A

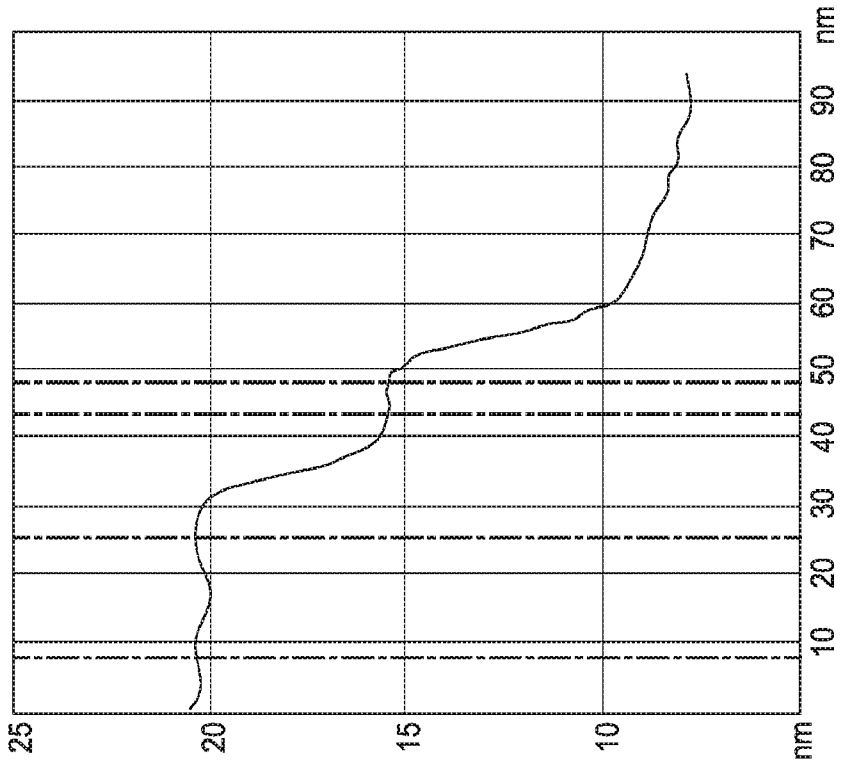
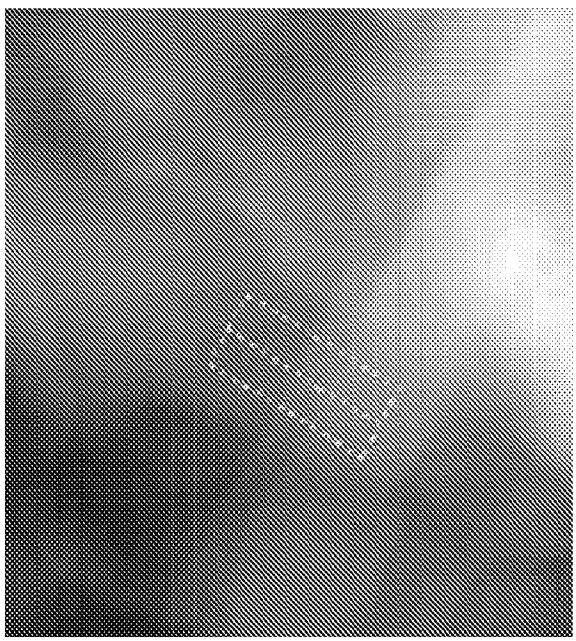


FIG. 1B



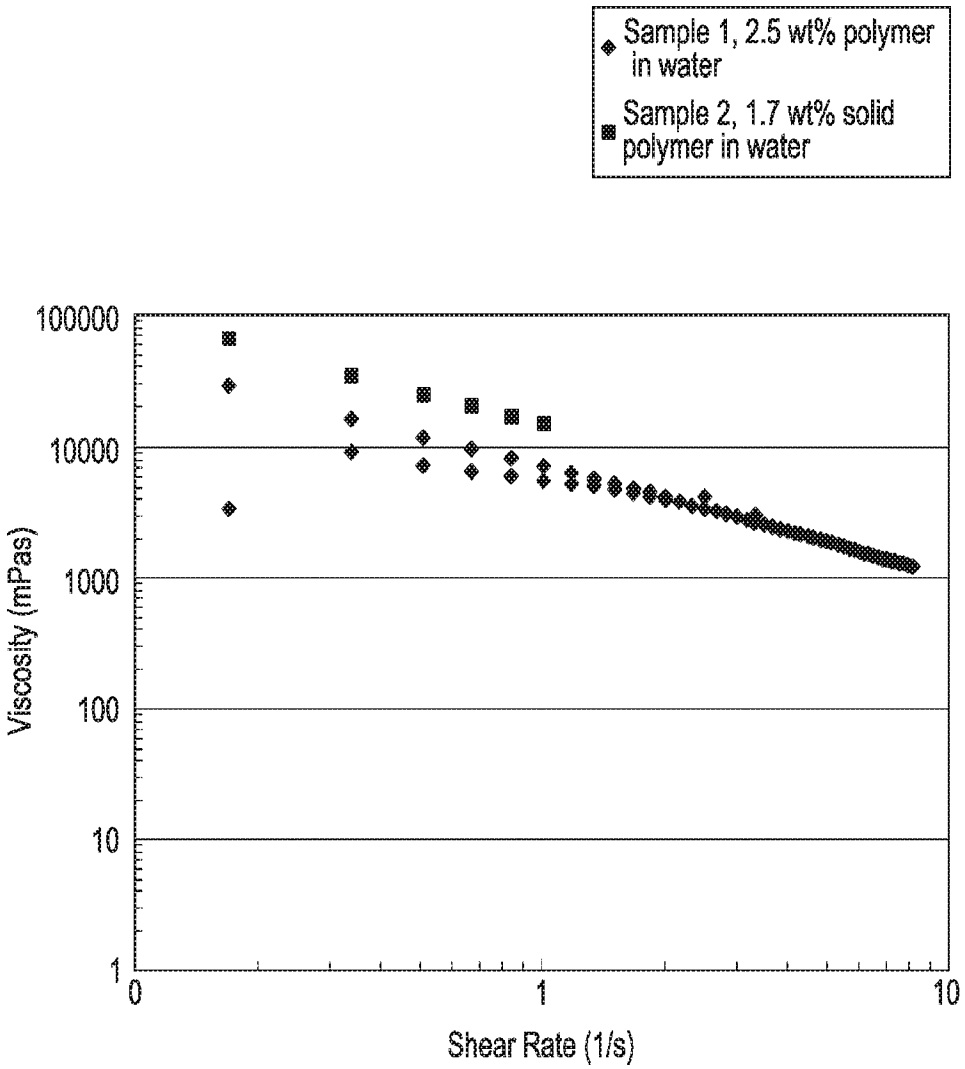

Sample 1, 2.5 wt% polymer in water
Sample 2, 1.7 wt% solid polymer in water
100000
10000
1000
100
10
1
Viscosity (mPas)
0
1
10
Shear Rate (1/s)

FIG. 2

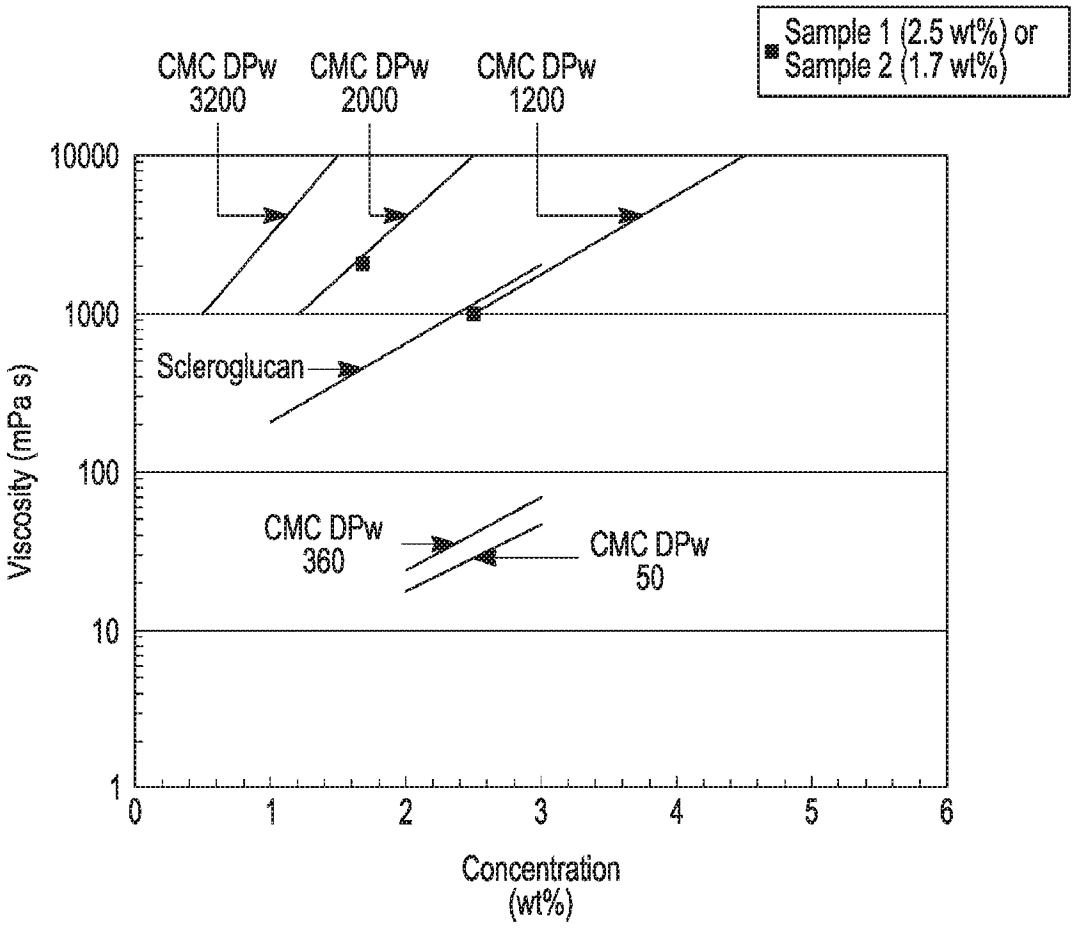

Sample 1 (2.5 wt%) or
Sample 2 (1.7 wt%)
CMC DPw
3200
CMC DPw
2000
CMC DPw
1200
10000
1000
100
10
1
Scleroglucan
CMC DPw
360
CMC DPw
50
Viscosity (mPa s)
0
1
2
3
4
5
6
Concentration
(wt%)

FIG. 3

INTERNATIONAL SEARCH REPORT

International application No
PCT/US2015/065699

A. CLASSIFICATION OF SUBJECT MATTER
INV. C12P19/04 C08L1/02
ADD.

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
C12P C08L

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practicable, search terms used)
EPO-Internal, FSTA, BIOSIS, EMBASE

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	HIRAISHI ET AL: "Synthesis of highly ordered cellulose II in vitro using cellodextrin phosphorylase", CARBOHYDRATE RESEARCH, vol. 344, 2009, pages 2468-2473, XP055005255, * See pages 2471-2472 (sections 4.1-4.3 and Scheme 1) *	1-16

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[X] Further documents are listed in the continuation of Box C. [ ] See patent family annex.

* Special categories of cited documents :

"A" document defining the general state of the art which is not considered to be of particular relevance

"E" earlier application or patent but published on or after the international filing date

"L" document which may throw doubts on priority claim(s) or which is cited to establish the publication date of another citation or other special reason (as specified)

"O" document referring to an oral disclosure, use, exhibition or other means

"P" document published prior to the international filing date but later than the priority date claimed

"T" later document published after the international filing date or priority date and not in conflict with the application but cited to understand the principle or theory underlying the invention

"X" document of particular relevance; the claimed invention cannot be considered novel or cannot be considered to involve an inventive step when the document is taken alone

"Y" document of particular relevance; the claimed invention cannot be considered to involve an inventive step when the document is combined with one or more other such documents, such combination being obvious to a person skilled in the art

"&" document member of the same patent family

Date of the actual completion of the international search	Date of mailing of the international search report
4 February 2016	17/02/2016

Name and mailing address of the ISA/	Authorized officer
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INTERNATIONAL SEARCH REPORT

International application No
PCT/US2015/065699

C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	SAWANO ET AL: "Characterization of Ruminococcus albus cellodextrin phosphorylase and identification of a key phenylalanine residue for acceptor specificity and affinity to the phosphate group", THE FEBS JOURNAL, vol. 280, 2013, pages 4463-4473, XP002753751, * See page 4463 (Abstract) *	1-16
A	CHASSARD ET AL: "Ruminococcus champanellensis sp. nov., a cellulose-degrading bacterium from human gut microbiota", INTERNATIONAL JOURNAL OF SYSTEMATIC AND EVOLUTIONARY MICROBIOLOGY, vol. 62, 2012, pages 138-143, XP002753752, * See page 142 (Description...) *	1-16
A	KADOKAWA ET AL: "Synthesis of new polysaccharide materials by phosphorylase-catalyzed chain elongation", POLYMER PREPRINTS, vol. 52, 2011, pages 59-60, XP055103508, * See page 60 (Experimental/ materials) *	1-16
A,P	PETROVIC ET AL: "Characterization of oligocellulose synthesized by reverse phosphorolysis using different cellodextrin phosphorylases", ANALYTICAL CHEMISTRY, vol. 87, 20 August 2015 (2015-08-20), pages 9639-9646, XP002753753, * See page 9645 (Conclusions) *	1-16
X	HATTORI ET AL: "Enzymatic synthesis of cellulose II-like substance via cellulolytic enzyme-nediated transglycosylation in an aqueous medium", CARBOHYDRATE RESEARCH, vol. 353, 2012, pages 22-26, XP002753865, * See page 22 (Abstract) *	1,2
A	PINEDA ET AL: "Técnicas de fermentación y aplicaciones de la celulosa bacteriana: una revisión", INGENERÍA Y CIENCIA, vol. 8, 2012, pages 307-335, XP002753866, * See section 2.1 *	1-16

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