HYDROGEL COMPOSITION WITH ENHANCED WATER ABSORPTION, AND PREPARATION METHOD THEREFOR AND USE THEREOF

CROSS-REFERENCE TO THE RELATED APPLICATIONS

[0001] The present disclosure claims the priority to the Chinese patent application with the filing No. 2024108373526 filed with the China National Intellectual Property Administration on June 26, 2024, and entitled "HYDROGEL COMPOSITION WITH ENHANCED WATER ABSORPTION, AND PREPARATION METHOD THEREFOR AND USE THEREOF", the entire contents of which are incorporated herein by reference.

TECHNICAL FIELD

[0002] The present disclosure relates to the technical field of food, and in particular, to a hydrogel composition with enhanced water absorption, a preparation method therefor, and a use thereof.

BACKGROUND

[0003] With the development of society and the improvement of living standards, obesity and diabetes have become increasingly serious health problems worldwide. At present, treatment methods for weight loss and diabetes mainly include medication, dietary control, and surgery. Surgery is an effective treatment, but it has high risks and requires a long recovery time. Weight loss via medication is often accompanied by weight rebound after drug withdrawal, and the drugs themselves have certain side effects. Dietary control is a safe and effective non-surgical approach for weight loss, but it requires individuals seeking weight loss to have strong self-control, and thus it is often difficult to achieve effective long-term weight loss. To assist individuals in dietary control, they are usually allowed to consume some products with a strong sense of satiety. Therefore, developing a safe and effective product to aid weight loss is of great significance.

[0004] The prior art has proposed some space-occupying gels in the digestive tract to increase satiety, thereby achieving weight loss. However, to ensure that the existing space-occupying gels in the digestive tract have a relatively high degree of crosslinking, the polymer concentration needs to be strictly controlled. Therefore, the precision requirements for the preparation process are high, which leads to difficulties in quality control of the space-occupying gels in the digestive tract.

[0005] In view of this, the present disclosure is made.

SUMMARY

[0006] The objective of the present disclosure is to provide a hydrogel composition with enhanced water absorption, a preparation method therefor, and a use thereof.

[0007] The present disclosure is implemented as follows.

[0008] In a first aspect, the present disclosure provides a hydrogel composition with enhanced water absorption, wherein the raw materials include carboxymethyl cellulose-based materials, alginic acid-based materials, readily soluble materials, a calcium source, and silicon dioxide, and the mass relationships of the components in the raw materials satisfy:

[0009] $W_1 = m_2/(m_1 + m_2) \times 100\%$, and $10\% \leq W_1 \leq 70\%$;

[0010] $W_2 = m_5/(m_1 + m_2 + m_5 + m_6) \times 100\%$, and $0\% \leq W_2 \leq 40\%$;

[0011] $W_3 = m_6/(m_1 + m_2 + m_5 + m_6) \times 100\%$, and $5\% \leq W_3 \leq 30\%$;

[0012] $W_4 = (m_1 + m_2)/(m_1 + m_2 + m_5 + m_6) \times 100\%$, and $55\% \leq W_4 \leq 90\%$; and

[0013] $f = m_4/m_3$, and $0.1 \leq f \leq 2.0$, wherein

[0014] m_1 is the mass of the carboxymethyl cellulose-based materials, m_2 is the mass of the alginic acid-based materials, m_3 is the mass of carboxyl groups in the alginic acid-based materials, m_4 is the mass of Ca^{2+} in the calcium source, m_5 is the mass of silicon dioxide, and m_6 is the mass of the readily soluble materials.

[0015] In a second aspect, the present disclosure provides a method for preparing the composition according to any one of the preceding embodiments, including dispersing carboxymethyl cellulose-based materials, alginic acid-based materials, and silicon dioxide in a solution, uniformly mixing and drying to obtain Material A, and then mixing Material A with a calcium source and readily soluble materials.

[0016] In a third aspect, the present disclosure provides use of the composition according to any one of the preceding embodiments in the preparation of a space-occupying product in the digestive tract.

[0017] The present disclosure has the following beneficial effects:

[0018] The present disclosure provides a hydrogel composition with enhanced water absorption, a preparation method therefor, and a use thereof. The composition can rapidly form a hydrogel in water, wherein the alginic acid-based materials are physically crosslinked with calcium ions, and the carboxymethyl cellulose-based materials can reduce the sensitivity of the alginic acid-based materials to calcium ions, so that the obtained hydrogel greatly

improves the medium water absorption rate while retaining a certain elastic modulus of the hydrogel. In addition, the raw materials further include readily soluble materials, which are dispersed in the solid hydrogel composition. When the solid hydrogel composition comes into contact with water, the readily soluble materials inside the composition have favorable water absorption capacity, thereby improving the ability of water to enter the composition, and further improving the water absorption performance of the composition. After water absorption, the volume expansion ability of the composition is improved, which is more conducive to space occupation in the digestive tract. After the addition of silicon dioxide, the strength of the hydrogel can also be enhanced. The process of forming the hydrogel from the composition of the present disclosure is entirely by means of physical crosslinking, without chemical reactions. The hydrogel prepared from the composition has high strength, high elastic modulus, and excellent water absorption performance. After entering the digestive tract, it can occupy the gastric volume, reduce food intake, thereby achieving the purposes of weight loss and blood glucose control.

DETAILED DESCRIPTION OF THE EMBODIMENTS

[0019] The embodiments of the present disclosure will be described in detail below with reference to the examples. However, those skilled in the art will appreciate that the following examples are only for illustrating the present disclosure and should not be construed as limiting the scope of the present disclosure. In the examples, where specific conditions are not indicated, the procedures are carried out under conventional conditions or conditions recommended by the manufacturer. The reagents or instruments used, where the manufacturer is not indicated, are all conventional products commercially available.

[0020] The endpoints and any values of the ranges disclosed in the present disclosure are not limited to the precise ranges or values, and these ranges or values should be understood to include values close to these ranges or values. For numerical ranges, the endpoints of the respective ranges, the endpoints and individual point values of the respective ranges, and the individual point values may be combined with one another to obtain one or more new numerical ranges, and these numerical ranges should be considered as specifically disclosed herein.

[0021] The inventor has found through research that in the prior art, citric acid is used to chemically crosslink low molecular weight sodium carboxymethyl cellulose to obtain a hydrogel having certain strength and water absorption performance, and the hydrogel can

increase satiety in the stomach, thereby achieving the purpose of weight loss. However, the preparation process of the hydrogel adopts chemical crosslinking, in which the hydroxyl groups in sodium carboxymethyl cellulose undergo a chemical reaction with citric acid to form ester bonds, thereby achieving connection between molecules and obtaining the hydrogel product. However, to ensure the crosslinking density in this process, the concentration of carboxymethyl cellulose in the reaction process needs to be strictly controlled, resulting in a complicated preparation process, low yield, and high cost.

[0022] Based on the existing intragastric space-occupying hydrogels prepared by chemical crosslinking methods, the inventor has found through research that there is a key problem in the practical application of hydrogels, namely that the medium water absorption rate and elastic modulus of the hydrogel are often contradictory. Generally speaking, when the medium water absorption rate of the hydrogel increases, the hydrogel becomes softer, and the corresponding elastic modulus of the hydrogel is lower, while the mechanical properties are poorer. Under the peristalsis and pressure of the digestive tract, such a hydrogel is prone to failure in maintaining the resulting satiety. When the medium water absorption rate of the hydrogel decreases, the hydrogel becomes harder, with a corresponding higher elastic modulus and better mechanical properties, but the decrease in water absorption rate leads to a smaller volume of the hydrogel, which in turn reduces the satiety of the hydrogel.

[0023] In addition, existing hydrogels generally can only provide good space-occupying effects in the stomach. After the hydrogel reaches the intestine, the higher pH of the intestinal fluid in the intestine causes the gel to further swell and lose a large part of its gel strength, and it cannot exert pressure on the intestine, ultimately resulting in a shortened duration of satiety.

[0024] Therefore, the present disclosure not only needs to develop a method with a simple process, but also needs to simultaneously improve the medium water absorption rate and elastic modulus of the hydrogel, as well as improve the tolerance of the hydrogel under different pH and pressure conditions. Based on this, the inventor proposes the following solution.

[0025] In a first aspect, the present disclosure provides a hydrogel composition with enhanced water absorption, wherein the raw materials include carboxymethyl cellulose-based materials, alginic acid-based materials, readily soluble materials, a calcium source, and silicon dioxide, and the mass relationships of the components in the raw materials satisfy:

[0026] $W_1 = m_2/(m_1 + m_2) \times 100\%$, and $10\% \leq W_1 \leq 70\%$;

[0027] $W_2 = m_5/(m_1 + m_2 + m_5 + m_6) \times 100\%$, and $0\% \leq W_2 \leq 40\%$;

[0028] $W_3 = m_6/(m_1 + m_2 + m_5 + m_6) \times 100\%$, and $5\% \leq W_3 \leq 30\%$;

[0029] $W_4 = (m_1 + m_2)/(m_1 + m_2 + m_5 + m_6) \times 100\%$, and $55\% \leq W_4 \leq 90\%$; and

[0030] $f = m_4/m_3$, and $0.1 \leq f \leq 2.0$, wherein

[0031] m_1 is the mass of the carboxymethyl cellulose-based materials, m_2 is the mass of the alginic acid-based materials, m_3 is the mass of carboxyl groups in the alginic acid-based materials, m_4 is the mass of Ca^{2+} in the calcium source, m_5 is the mass of silicon dioxide, and m_6 is the mass of the readily soluble materials. The units of m_1, m_2, m_3, m_4, m_5, and m_6 are the same, which may all be g or all be kg.

[0032] When the composition of the present disclosure is dispersed in water, the composition can undergo gelation, thereby forming a hydrogel. Specifically, the alginic acid-based materials in the composition can chelate with calcium ions in a solution state to form a hydrogel, undergoing physical crosslinking. However, because alginic acid-based materials have high sensitivity to calcium ions, when alginic acid-based materials and a calcium source are directly mixed, the alginic acid-based materials and calcium ions chelate rapidly in a solution state, forming a hydrogel with high strength and poor water absorption capacity, which cannot meet the requirements for a space-occupying gel in the digestive tract.

[0033] Therefore, in one aspect, the applicant proposes to add carboxymethyl cellulose-based materials to the system of alginic acid-based materials and calcium source. The carboxymethyl cellulose-based materials can reduce the sensitivity of the alginic acid-based materials to calcium ions, avoiding the formation of a hydrogel with excessively high strength and poor water absorption. Therefore, by controlling the values of W_1 and f, the degree of chelation between the alginic acid-based materials and calcium ions can be controlled, thereby controlling the water absorption performance of the composition, so as to achieve controllable adjustment of the water absorption capacity and elastic modulus of the composition during gelation. As a result, the hydrogel formed from the composition provided by the present disclosure has not only good water absorption capacity, but also a relatively high elastic modulus.

[0034] In another aspect, by adding readily soluble materials to the system of alginic acid-based materials and calcium source, when the composition is in a solid particulate state, the readily soluble materials form physical space occupation in the composition. When the composition comes into contact with water to form a hydrogel, the readily soluble materials inside the composition have favorable water absorption capacity, thereby promoting external

water to enter the interior of the composition, thus improving the water absorption performance of the composition. The volume expansion rate of the composition after water absorption increases, which is more conducive to achieving its space occupation in the digestive tract and enhancing the weight loss effect. Therefore, controllable adjustment of the water absorption capacity and elastic modulus of the composition during gelation can also be achieved by adjusting the value of W_3. As a result, the hydrogel formed from the composition provided by the present disclosure has not only good water absorption capacity, but also a relatively high elastic modulus.

[0035] Further, when silicon dioxide is added to the system of the above composition of the present disclosure, the elastic modulus of the hydrogel formed from the composition can also be adjusted. By controlling the value of W_2, the hydrogel formed from the composition of the present disclosure further has relatively high strength.

[0036] Further, the composition of the present disclosure does not undergo chemical reactions during the preparation process, and all are physical crosslinking processes. The main molecular chains are formed by chelation of alginic acid-based materials and the calcium source. Compared with chemical crosslinking processes that require strict control of the crosslinked molecular weight to control parameters such as water absorption capacity and elastic modulus of the hydrogel, the composition provided by the present disclosure can achieve controllable adjustment of the water absorption capacity and elastic modulus of the hydrogel by controlling the proportions of the raw materials, making preparation more convenient, with higher yield and lower development cost.

[0037] When the composition of the present disclosure is dispersed in water, the composition rapidly absorbs water and swells, forming a gelatinous hydrogel. After swallowing the hydrogel, the hydrogel passes through the esophagus into the stomach. Because the hydrogel prepared from the composition of the present disclosure has high strength, high elastic modulus, and excellent water absorption performance, it can occupy the gastric volume in the stomach, reduce food intake, thereby achieving the purposes of weight loss and blood glucose control. When the hydrogel reaches the small intestine, in the high pH environment of the small intestine, the hydrogel swells again, and at the same time, the hydroxide ions of high pH react with silicon dioxide, locally forming microgels, which enhance the elastic modulus at high pH, thereby occupying the volume of the small intestine and/or exerting pressure on the intestinal wall to continuously generate satiety, which is more conducive to weight management.

[0038] In optional embodiments, W_1 can be 10%, 20%, 30%, 40%, 50%, 60%, or 70%, or range values between any two of the above values.

[0039] W_2 can be 0%, 5%, 10%, 15%, 20%, 25%, 30%, 35%, or 40%, or range values between any two of the above values.

[0040] W_3 can be 5%, 7%, 10%, 12%, 15%, 17%, 20%, 22%, 25%, 27%, or 30%, or range values between any two of the above values.

[0041] W_4 can be 55%, 60%, 65%, 70%, 75%, 80%, 85%, or 90%, or range values between any two of the above values.

[0042] f can be 0.1, 0.5, 1.0, 1.5, or 2.0, or range values between any two of the above values.

[0043] To further optimize the strength, water absorption, and elastic modulus of the hydrogel formed after gelation of the composition provided by the present disclosure, the parameter ranges of W_1, W_2, W_3, W_4, and f can be further controlled.

[0044] Specifically, W_1 satisfies: $10\% \leq W_1 \leq 70\%$, W_2 satisfies: $5\% \leq W_2 \leq 30\%$, W_3 satisfies: $5\% \leq W_3 \leq 25\%$, W_4 satisfies: $65\% \leq W_4 \leq 90\%$, and f satisfies: $0.2 \leq f \leq 2.0$.

[0045] More preferably, W_1 satisfies: $12\% \leq W_1 \leq 25\%$, W_2 satisfies: $9\% \leq W_2 \leq 25\%$, W_3 satisfies: $5\% \leq W_3 \leq 20\%$, W_4 satisfies: $70\% \leq W_4 \leq 90\%$, and f satisfies: $0.4 \leq f \leq 2.00$.

[0046] Further, the selection of the source of raw materials is another factor in improving the performance of the hydrogel formed from the composition. Taking sodium alginate as an example, some literature reports that sodium alginate can form a gel at gastric pH, which can prolong gastric emptying time, thereby reducing food intake and preventing obesity. However, due to limitations in the source, molecular weight, and molecular structure (the ratio of mannuronic acid to guluronic acid) of alginates, the weight loss effects reported in different literature vary greatly, greatly limiting the application of alginates in the field of weight loss.

[0047] To further improve the performance of the hydrogel, in optional embodiments, the alginic acid-based materials have a viscosity of ≥ 2000 mPa·s in a solution at a concentration of 2%; preferably, the alginic acid-based materials have a viscosity of ≥ 3000 mPa·s in a solution at a concentration of 2%; more preferably, the alginic acid-based materials have a viscosity of 3000 to 20000 mPa·s in a solution at a concentration of 2%.

[0048] The carboxymethyl cellulose-based materials have a viscosity of ≥ 4000 mPa·s in a solution at a concentration of 2%; preferably, the carboxymethyl cellulose-based materials have a viscosity of ≥ 8000 mPa·s in a solution at a concentration of 2%; more preferably, the

carboxymethyl cellulose-based materials have a viscosity of 8000 to 60000 mPa·s in a solution at a concentration of 2%.

[0049] Since carboxymethyl cellulose-based materials and alginic acid-based materials with different molecular weights and molecular structures exhibit different properties, the present disclosure enables control of the viscosity ranges of the carboxymethyl cellulose-based materials and the alginic acid-based materials, thereby controlling the molecular weights and molecular structures of the carboxymethyl cellulose-based materials and the alginic acid-based materials, and further improving the water absorption and elastic modulus of the hydrogel formed from the composition.

[0050] Preferably, the carboxymethyl cellulose-based materials include but are not limited to carboxymethyl cellulose and salts thereof, and the salts of carboxymethyl cellulose include any one selected from the group consisting of sodium carboxymethyl cellulose, potassium carboxymethyl cellulose, and ammonium carboxymethyl cellulose. Any raw material having a carboxymethyl cellulose structure can be used as a raw material for reducing the sensitivity of sodium alginate and calcium source.

[0051] Preferably, the alginic acid-based materials include but are not limited to alginic acid and salts thereof, and the salts of alginic acid include any one selected from the group consisting of sodium alginate, potassium alginate, and ammonium alginate. Any raw material having an alginic acid and carboxyl group structure thereof can be used as a raw material for chelating with a calcium source and undergoing physical crosslinking.

[0052] Preferably, the calcium source includes but is not limited to at least one selected from the group consisting of calcium carbonate, calcium chloride, calcium citrate, calcium lactate, calcium gluconate, calcium acetate, and calcium phosphate; all products capable of providing a calcium source can serve as the calcium source of the present disclosure. More preferably, the calcium source is a calcium source of low solubility; still more preferably, the calcium source of low solubility is one selected from the group consisting of calcium lactate, calcium carbonate, and calcium phosphate.

[0053] In optional embodiments, the solubility of the readily soluble materials is greater than the solubility of at least one of the carboxymethyl cellulose-based materials and the alginic acid-based materials.

[0054] Preferably, the solubility of the readily soluble materials is greater than the solubility of the material having the highest solubility among the carboxymethyl cellulose-based

materials and the alginic acid-based materials, so as to improve the water absorption performance of the composition.

[0055] Preferably, the solubility of the readily soluble materials is greater than or equal to 100 g/L.

[0056] Preferably, the readily soluble materials include but are not limited to at least one selected from the group consisting of polyvinyl alcohol, polyethylene glycol, cyclodextrin, and sucrose. Any substance containing hydroxyl groups capable of forming hydrogen bonds with water can serve as the readily soluble material of the present disclosure.

[0057] It should be noted that since the composition provided by the present disclosure needs to enter the digestive tract after forming a hydrogel to achieve space occupation in the digestive tract, thereby providing satiety and achieving weight loss, it can be understood that all components in the raw materials of the composition of the present disclosure are food-grade components to ensure that the hydrogel formed from the composition is edible.

[0058] In optional embodiments, the composition has a particle size of 0.1 mm to 3 mm, more preferably 0.3 mm to 2.5 mm, and still more preferably 0.3 mm to 1 mm.

[0059] In a second aspect, the present disclosure provides a method for preparing the hydrogel composition according to any one of the preceding embodiments, including dispersing carboxymethyl cellulose-based materials, alginic acid-based materials, readily soluble materials, and silicon dioxide in a solution, uniformly mixing and drying to obtain Material A, and then mixing Material A with a calcium source.

[0060] By preparing Material A in advance, the carboxymethyl cellulose-based materials, the readily soluble materials, and the silicon dioxide can be added to the alginic acid-based materials in advance for pre-occupation, which is beneficial for carboxymethyl cellulose to play a role in reducing the sensitivity of alginic acid-based materials to calcium ions, for readily soluble materials to pre-occupy space to improve water absorption capacity, and for silicon dioxide to enhance the strength of the composition.

[0061] In Material A, the carboxymethyl cellulose-based materials, alginic acid-based materials, readily soluble materials, and silicon dioxide are uniformly dispersed in the solution, and then dried to obtain a uniformly mixed solid Material A, which is then physically mixed with a calcium source to obtain the composition product. When the composition comes into contact with water, the calcium source rapidly chelates with the alginic acid-based materials to form long-chain molecules. However, due to the pre-dispersion effect of the carboxymethyl cellulose-based materials, the chelating ability of the

calcium source is easy to adjust, avoiding a decrease in the water absorption of the composition. In addition, the readily soluble materials can also exert excellent water absorption performance in the composition, promoting water to enter the interior of the composition, thereby improving the water absorption capacity of the composition. Silicon dioxide improves the strength of the composition, making the volume and strength of the hydrogel easy to control during the process of the composition absorbing water and transforming into a hydrogel.

[0062] Preferably, during the preparation of Material A, the polymer concentration in the solution is 4% to 10%, and the drying temperature is 45°C to 100°C; more preferably, the drying temperature is 45°C to 80°C.

[0063] Here, the polymer in the solution refers to the total concentration of the carboxymethyl cellulose-based materials and the alginic acid-based materials.

[0064] The hydrogel composition provided by the present disclosure is in a solid state before gelation, and after being added to water, it can rapidly undergo gelation to form a hydrogel. After ingestion, the hydrogel can occupy space in the digestive tract, thereby providing a sensation of satiety, reducing meal frequency and food intake, and consequently achieving weight loss and adjunctively ameliorating diabetes symptoms.

[0065] In a third aspect, the present disclosure provides use of the composition according to any one of the preceding embodiments in the preparation of a space-occupying product in the digestive tract.

[0066] In optional embodiments, a method of using the hydrogel composition provided by the present disclosure includes adding the solid composition as a whole to water with stirring, or first adding Material A to water with stirring to obtain a pre-gel suspension, and then adding the remaining raw materials to the pre-gel suspension with stirring.

[0067] Example 1

[0068] The present example provides a hydrogel composition with enhanced water absorption, consisting of Material A and Material B, wherein Material A is composed of sodium carboxymethyl cellulose, sodium alginate, polyethylene glycol, and silicon dioxide, and Material B is composed of calcium lactate. Here, $W_1 = 23\%$, $W_2 = 10\%$, $W_3 = 10\%$, $W_4 = 80\%$, and $f = 0.9$.

[0069] Specifically, in the present example, the mass of sodium carboxymethyl cellulose is 60.00 g, and the viscosity in the solution at a concentration of 2% is 12000 mPa·s.

[0070] The mass of sodium alginate is 17.92 g, and the viscosity in the solution at a concentration of 2% is 4000 mPa·s.

[0071] The mass of silicon dioxide is 9.74 g, and the mass of polyethylene glycol is 9.74 g.

[0072] The present example further provides a method for preparing the above composition, including the following steps:

[0073] Sodium alginate was added to 1500 mL of water in proportion and dispersed under mechanical stirring at 1000 rpm until dissolved, and then polyethylene glycol was added thereto until dissolved with stirring. Sodium carboxymethyl cellulose and silicon dioxide were added and stirred under mechanical stirring at 1000 rpm until a uniform suspension was formed, and no large lumps were observed. The suspension was transferred to a metal tray; the thickness of the suspension in the metal tray was controlled to be below 1 cm, and then the metal tray was placed in an oven at 50°C and dried for 72 h to obtain solid Material A. The solid Material A was placed in a pulverizer for pulverization, passed through 10-mesh and 120-mesh sieves, and sieved to obtain particulate Material A having a particle size of 0.1 mm to 2 mm.

[0074] Calcium lactate was encapsulated as Material B, and the mass of calcium lactate in Material B and the mass of sodium alginate in Material A satisfied $f = 0.9$.

[0075] Example 2

[0076] The present example provides a hydrogel composition with enhanced water absorption, consisting of Material A and Material B, wherein Material A is composed of sodium carboxymethyl cellulose, sodium alginate, α-cyclodextrin, and silicon dioxide, and Material B is composed of calcium lactate. Here, $W_1 = 23\%$, $W_2 = 10\%$, $W_3 = 10\%$, $W_4 = 80\%$, and $f = 0.9$.

[0077] Specifically, in the present example, the mass of sodium carboxymethyl cellulose is 60.00 g, and the viscosity in the solution at a concentration of 2% is 12000 mPa·s.

[0078] The mass of sodium alginate is 17.92 g, and the viscosity in the solution at a concentration of 2% is 5000 mPa·s.

[0079] The mass of silicon dioxide is 9.74 g, and the mass of α-cyclodextrin is 9.74 g.

[0080] The present example further provides a method for preparing the above composition. The preparation method is similar to Example 1, with the difference only in the amounts of the substances used, and the drying temperature is 80°C.

[0081] Example 3

[0082] The present example provides a hydrogel composition with enhanced water absorption, consisting of Material A and Material B, wherein Material A is composed of sodium carboxymethyl cellulose, sodium alginate, sucrose, and silicon dioxide, and Material B is composed of calcium lactate. Here, W_1 = 23%, W_2 = 10%, W_3 = 8%, W_4 = 82%, f = 0.9.

[0083] Specifically, in the present example, the mass of sodium carboxymethyl cellulose is 60.00 g, and the viscosity in the solution at a concentration of 2% is 16000 mPa·s.

[0084] The mass of sodium alginate is 17.92 g, and the viscosity in the solution at a concentration of 2% is 5000 mPa·s.

[0085] The mass of silicon dioxide is 9.50 g, and the mass of sucrose is 7.60 g.

[0086] The present example further provides a method for preparing the above composition. The preparation method is similar to Example 1, with the difference only in the amounts of the substances used, and the drying temperature is 85°C.

[0087] Example 4

[0088] The present example provides a hydrogel composition with enhanced water absorption, consisting of Material A and Material B, wherein Material A is composed of potassium carboxymethyl cellulose, sodium alginate, α-cyclodextrin, and silicon dioxide, and Material B is composed of calcium lactate. Here, W_1 = 23%, W_2 = 15%, W_3 = 10%, W_4 = 65%, f = 0.9.

[0089] Specifically, in the present example, the mass of sodium carboxymethyl cellulose is 90.00 g, and the viscosity in the solution at a concentration of 2% is 8000 mPa·s.

[0090] The mass of sodium alginate is 26.88 g, and the viscosity in the solution at a concentration of 2% is 5000 mPa·s.

[0091] The mass of silicon dioxide is 27.00 g, and the mass of α-cyclodextrin is 18.00 g.

[0092] The present example further provides a method for preparing the above composition. The preparation method is similar to Example 1, with the difference only in the amounts of the substances used, the drying temperature is 70°C, and the sieves used are 20-mesh and 50-mesh, respectively.

[0093] Example 5

[0094] The present example provides a hydrogel composition with enhanced water absorption, consisting of Material A and Material B, wherein Material A is composed of potassium carboxymethyl cellulose, sodium alginate, polyvinyl alcohol, and silicon dioxide, and Material B is composed of calcium lactate. Here, W_1 = 17%, W_2 = 10%, W_3 = 10%, W_4 = 80%, and f = 0.9.

[0095] Specifically, in the present example, the mass of sodium carboxymethyl cellulose is 60.00 g, and the viscosity in the solution at a concentration of 2% is 12000 mPa·s.

[0096] The mass of sodium alginate is 12.30 g, and the viscosity in the solution at a concentration of 2% is 6000 mPa·s.

[0097] The mass of silicon dioxide is 9.00 g, and the mass of polyvinyl alcohol is 9.00 g.

[0098] The present example further provides a method for preparing the above composition. The preparation method is similar to Example 1, with the difference only in the amounts of the substances used, and the sieves used are 20-mesh and 50-mesh, respectively.

[0099] Example 6

[0100] The present example provides a hydrogel composition with enhanced water absorption, consisting of Material A and Material B, wherein Material A is composed of potassium carboxymethyl cellulose, sodium alginate, polyethylene glycol, and silicon dioxide, and Material B is composed of calcium lactate. Here, W_1 = 23%, W_2 = 25%, W_3 = 10%, W_4 = 65%, f = 1.4.

[0101] Specifically, in the present example, the mass of sodium carboxymethyl cellulose is 60.00 g, and the viscosity in the solution at a concentration of 2% is 12000 mPa·s.

[0102] The mass of sodium alginate is 17.92 g, and the viscosity in a solution at a concentration of 2% is 3200 mPa·s.

[0103] The mass of silicon dioxide is 30.00 g, and the mass of polyethylene glycol is 12.00 g.

[0104] The present example further provides a method for preparing the above composition. The preparation method is similar to Example 1, with the difference only in the amounts of the substances used; the sieves used are 20-mesh and 50-mesh, respectively, and the drying temperature is 85°C.

[0105] Example 7

[0106] The present example provides a hydrogel composition with enhanced water absorption, consisting of Material A and Material B, wherein Material A is composed of potassium carboxymethyl cellulose, α-cyclodextrin, and sodium alginate, and Material B is composed of calcium lactate. Here, W_1 = 23%, W_2 = 0%, W_3 = 10%, W_4 = 90%, f = 0.9.

[0107] Specifically, in the present example, the mass of sodium carboxymethyl cellulose is 60.00 g, and the viscosity in the solution at a concentration of 2% is 12000 mPa·s.

[0108] The mass of sodium alginate is 17.92 g, and the viscosity in a solution at a concentration of 2% is 4000 mPa·s.

[0109] The mass of α-cyclodextrin is 8.66 g.

[0110] The present example further provides a method for preparing the above composition. The preparation method is similar to Example 1, with the difference only in the amounts of the substances used, and the drying temperature is 90°C.

[0111] Example 8

[0112] The present example provides a hydrogel composition with enhanced water absorption, consisting of Material A and Material B, wherein Material A is composed of potassium carboxymethyl cellulose, sodium alginate, α-cyclodextrin, and silicon dioxide, and Material B is composed of calcium lactate. Here, W_1 = 23%, W_2 = 15%, W_3 = 10%, W_4 = 65%, f = 1.5.

[0113] Specifically, in the present example, the mass of sodium carboxymethyl cellulose is 60.00 g, and the viscosity in the solution at a concentration of 2% is 14000 mPa·s.

[0114] The mass of sodium alginate is 17.92 g, and the viscosity in a solution at a concentration of 2% is 3200 mPa·s.

[0115] The mass of silicon dioxide is 18.00 g, and the mass of α-cyclodextrin is 12.00 g.

[0116] The present example further provides a method for preparing the above composition. The preparation method is similar to Example 1, with the difference only in the amounts of the substances used; the drying temperature is 75°C, and the sieves used are 18-mesh and 60-mesh, respectively.

[0117] Comparative Example 1

[0118] The present comparative example provides a hydrogel composition, including mixing sodium carboxymethyl cellulose at a concentration of 6% (by weight of water) with citric acid at a concentration of 0.3% (by weight of carboxymethyl cellulose). The mixture was dried in an oven at 45°C, and then ground to form particles having a particle size of 100 μm to 1000 μm. The above particles were crosslinked at 120°C for 4 hours to obtain a hydrogel composition.

[0119] Comparative Example 2

[0120] The present comparative example provides a hydrogel composition, including washing the heat-treated crosslinked hydrogel composition obtained in Comparative Example 1 with deionized water three times, and then drying again to obtain the hydrogel composition of the present comparative example.

[0121] Comparative Example 3

[0122] The present comparative example provides a hydrogel composition, the preparation method of which is the same as Example 1, with the difference only that the raw materials do not contain polyethylene glycol and silicon dioxide.

[0123] Comparative Example 4

[0124] The present comparative example provides a hydrogel composition, the preparation method of which is similar to Example 7, with the difference only that the raw materials do not contain α-cyclodextrin.

[0125] Comparative Example 5

[0126] The present comparative example provides a hydrogel composition, including mixing 10 g of water-absorbent polymer fibers (PAA and PVA, wherein COOH : OH = 3.5:1), 20 g of 3% carboxymethyl cellulose solution, 30 g of 3% sodium alginate, 8 g of α-cyclodextrin, 1 g of citric acid, 0.5 g of silicon dioxide, and 0.5 g of titanium dioxide to form a homogeneous solution, and then crosslinking the above homogeneous solution at 120°C for 3 hours. The product after the crosslinking reaction was dried and ground to obtain a sample of 200 to 5000 micrometers, and then the sample was washed 5 times with purified water at normal temperature and pressure. After washing, it was dried, ground, and sieved to obtain a water-absorbing hydrogel of 200 to 4000 micrometers.

[0127] Experimental Example 1

[0128] 1) Detection in simulated gastric fluid:

[0129] Artificial gastric fluid was prepared according to Part IV of the Chinese Pharmacopoeia 2020, then diluted 8 times, and the pH was adjusted to 2.1 to serve as simulated artificial gastric fluid half an hour before a meal (hereinafter referred to as simulated gastric fluid).

[0130] 40±1 mL of simulated gastric fluid was measured and stirred on a magnetic stirrer at a stirring speed of 100 rpm. 0.25±0.002 g of each of Examples 1 to 8 and Comparative Examples 1 to 5 were weighed, and the calcium source was weighed according to the corresponding f and added together to the simulated gastric fluid with continued stirring (without creating a vortex), and the stirring time was 30±2 min, for simulating the treatment process of gastric fluid.

[0131] A piece of 150-mesh gauze was taken and weighed, recorded as g_1. The above stirred mixture was then filtered through the 150-mesh gauze to remove unabsorbed simulated gastric fluid (during filtration, the liquid on the surface of the gauze was gently scraped with a hand until no visible liquid remained on the hand, indicating the end). The

mixture and the gauze were then weighed together and recorded as g_2, and the state of the hydrogel obtained after filtration was observed.

[0132] The medium water absorption rate MUR of the composition in simulated gastric fluid was calculated as MUR = $(g_2-g_1)/(0.25 \times W_4)$. The elastic modulus G_1' of the filtered hydrogel was measured using a rheometer (uniformly taking the value corresponding to an angular frequency of 10 rad/s). The results are shown in Table 1.

[0133] 2) Detection in simulated intestinal fluid:

[0134] Artificial intestinal fluid was prepared according to Part IV of the Chinese Pharmacopoeia 2020 to serve as simulated small intestinal fluid (hereinafter referred to as simulated intestinal fluid).

[0135] The hydrogel after the detection in simulated gastric fluid in 1) was weighed and recorded as g_0, and then the hydrogel was added to a beaker containing 200 mL of simulated intestinal fluid. The beaker was transferred to a shaker and treated at 37°C and 60 rpm for 120 min, to simulate the treatment process of intestinal fluid.

[0136] A piece of 150-mesh gauze was taken and weighed, recorded as g_3. The above treated mixture was then filtered through the 150-mesh gauze to remove unabsorbed simulated intestinal fluid (during filtration, the liquid on the surface of the gauze was gently scraped with a hand until no visible liquid remained on the hand, indicating the end). The mixture and the gauze were weighed together and recorded as g_4, and the state of the hydrogel obtained after filtration was observed.

[0137] The elastic modulus G_2' of the filtered hydrogel was measured using a rheometer (uniformly taking the value corresponding to an angular frequency of 10 rad/s). The results are shown in Table 1.

Table 1 Performance of compositions and hydrogels

	W_1(%)	W_2(%)	W_3(%)	W_4(%)	f	MUR (g/g)	G_1' (Pa)	G_2' (Pa)
Example 1	23	10	10	80	0.9	83.8	1256	910
Example 2	23	10	10	80	0.9	69.9	1187	865
Example 3	23	10	8	82	0.9	85.8	1206	890
Example 4	23	15	10	65	0.9	59.5	1620	1100
Example 5	17	10	10	80	0.9	75.5	1198	846
Example 6	23	25	10	65	1.4	53.8	2498	2026

Example 7	23	0	10	90	0.9	70.5	920	562
Example 8	23	15	10	65	1.5	48.2	1980	1354
Comparative Example 1	N.A	N.A	N.A	N.A	N.A	27.5	4218	--
Comparative Example 2	N.A	N.A	N.A	N.A	N.A	70.8	950	400
Comparative Example 3	23	0	0	100	0.9	58.2	700	556
Comparative Example 4	23	10	0	90	0.9	60.0	1185	930
Comparative Example 5	N.A	N.A	N.A	N.A	N.A	89.0	500	230

Note: N.A means not applicable; Comparative Example 1 was not subjected to subsequent testing due to its low MUR and poor space-occupying effect.

[0138] As can be seen from Table 1, after the compositions provided in the examples of the present disclosure are transformed into hydrogels, the medium water absorption rate can reach up to 85.8 times the original weight, and after water absorption in simulated gastric fluid, they still maintain a relatively high elastic modulus (greater than 900 Pa), indicating that the compositions provided in the examples of the present disclosure can balance water absorption performance and strength, which is conducive to enhancing the weight loss effect.

[0139] Comparative Examples 1, 2, and 5 all use chemical crosslinking, and the preparation processes are complicated. The hydrogel obtained in Comparative Example 1 has poor water absorption performance and high strength, resulting in insufficient swelling volume of the hydrogel, making it difficult to achieve effective space occupation in the digestive tract. Compared with Comparative Example 1, the hydrogel obtained in Comparative Example 2 has improved water absorption performance, but the elastic modulus decreases, and repeated washing with deionized water is required, which is time-consuming and results in low preparation yield. Comparative Examples 3 and 4 use physical crosslinking similar to the present disclosure to obtain hydrogel compositions. However, Comparative Example 3 does not contain readily soluble materials and silicon dioxide, and its medium water absorption rate and elastic modulus are significantly lower than those of Example 1. Comparative Example 4 does not contain readily soluble materials, and its medium water absorption rate is significantly lower than that of Example 7.

[0140] Experimental Example 2

[0141] 20 volunteers were recruited and randomly divided into 2 groups. The control group took the same amount of sucrose, and the experimental group took 3 g of the hydrogel composition with enhanced water absorption of Example 3 with two meals per day. The administration time was 30 min before meals, twice a day, namely lunch at noon and dinner in the afternoon. At the same time, nutritional intervention was provided to the volunteers, with a daily reduction of 300 kcal in caloric intake. The body weights of the volunteers were recorded at the 2nd month and the 6th month, respectively, and the mean body weights were calculated as shown in Table 2.

Table 2 Body weight change table

	Initial body weight/kg	Body weight at 2 months/kg	Body weight at 6 months/kg
	n = 10	n = 10	n = 8
Control group	85.51	84.94	82.97
Experimental group	86.30	83.33	79.59

Note: n represents the number of subjects.

[0142] As can be seen from Table 2, through nutritional intervention and the use of the composition provided by the present disclosure, good weight loss effects were achieved together. Within 24 weeks, the body weight of the volunteers in the experimental group decreased by 6.71 kg, while that of the control group was only 2.54 kg, indicating that the hydrogel composition provided in the examples of the present disclosure has good weight loss effects.

[0143] The hydrogel composition with enhanced water absorption, the preparation method therefor, and the use thereof provided in the examples of the present disclosure have at least the following advantages:

[0144] 1) The composition provided by the present disclosure is prepared by physical action of food-grade raw materials and can be directly consumed as food.

[0145] 2) The present disclosure uses alginic acid-based materials and a calcium source for physical crosslinking to prepare the composition and form a hydrogel. Compared with hydrogels prepared by chemical crosslinking methods, the preparation process is simpler and the yield is higher, which is conducive to large-scale production.

[0146] 3) By controlling the contents of carboxymethyl cellulose-based materials, readily soluble materials, and silicon dioxide, the water absorption performance and strength of the hydrogel formed from the composition can be directly adjusted, ensuring that the composition can rapidly absorb water and swell, forming space occupation with a larger volume while ensuring its mechanical strength, thereby ensuring the long-lasting satiety effect of the hydrogel in the digestive tract.

[0147] The above are only preferred embodiments of the present disclosure and are not intended to limit the present disclosure. For those skilled in the art, the present disclosure may have various modifications and variations. Any modifications, equivalent substitutions, improvements, etc. made within the spirit and principle of the present disclosure shall fall within the protection scope of the present disclosure.

INDUSTRIAL APPLICABILITY

[0148] The present disclosure provides a hydrogel composition with enhanced water absorption, a preparation method therefor, and a use thereof. The composition is prepared by physical action of food-grade raw materials and can be directly consumed as food. The preparation process is simpler, the yield is higher, and it is conducive to large-scale production.

CLAIMS

1. A hydrogel composition with enhanced water absorption, wherein the raw materials comprise carboxymethyl cellulose-based materials, alginic acid-based materials, readily soluble materials, a calcium source, and silicon dioxide, and mass relationships of the components in the raw materials satisfy:

$W_1 = m_2/(m_1 + m_2) \times 100\%$, and $10\% \leq W_1 \leq 70\%$;

$W_2 = m_5/(m_1 + m_2 + m_5 + m_6) \times 100\%$, and $0\% \leq W_2 \leq 40\%$;

$W_3 = m_6/(m_1 + m_2 + m_5 + m_6) \times 100\%$, and $5\% \leq W_3 \leq 30\%$;

$W_4 = (m_1 + m_2)/(m_1 + m_2 + m_5 + m_6) \times 100\%$, and $55\% \leq W_4 \leq 90\%$; and

$f = m_4/m_3$, and $0.1 \leq f \leq 2.0$, wherein

m_1 is mass of the carboxymethyl cellulose-based materials, m_2 is mass of the alginic acid-based materials, m_3 is mass of carboxyl groups in the alginic acid-based materials, m_4 is a mass of Ca^{2+} in the calcium source, m_5 is a mass of silicon dioxide, and m_6 is mass of the readily soluble materials.

2. The composition according to claim 1, wherein W_1 satisfies: $10\% \leq W_1 \leq 70\%$, W_2 satisfies: $5\% \leq W_2 \leq 30\%$, W_3 satisfies: $5\% \leq W_3 \leq 25\%$, W_4 satisfies: $65\% \leq W_4 \leq 90\%$, and f satisfies: $0.2 \leq f \leq 2.0$; and

more preferably, W_1 satisfies: $12\% \leq W_1 \leq 25\%$, W_2 satisfies: $9\% \leq W_2 \leq 25\%$, W_3 satisfies: $5\% \leq W_3 \leq 20\%$, W_4 satisfies: $70\% \leq W_4 \leq 90\%$, and f satisfies: $0.4 \leq f \leq 2.0$.

3. The composition according to claim 1 or 2, wherein a solubility of the readily soluble materials is greater than a solubility of at least one of the carboxymethyl cellulose-based materials and the alginic acid-based materials;

preferably, the solubility of the readily soluble materials is greater than or equal to 100 g/L; and

preferably, the readily soluble materials comprise at least one selected from the group consisting of polyvinyl alcohol, polyethylene glycol, cyclodextrin, and sucrose.

4. The composition according to claim 1, wherein the carboxymethyl cellulose-based materials have a viscosity of ≥ 4000 mPa·s in a solution at a concentration of 2%; and

preferably, the carboxymethyl cellulose-based materials comprise carboxymethyl cellulose and salts thereof, wherein the salts of carboxymethyl cellulose comprise any one

selected from the group consisting of sodium carboxymethyl cellulose, potassium carboxymethyl cellulose, and ammonium carboxymethyl cellulose.

5. The composition according to claim 1, wherein the alginic acid-based materials have a viscosity of ≥ 2000 mPa·s in a solution at a concentration of 2%; and

preferably, the alginic acid-based materials comprise alginic acid and salts thereof, wherein the salts of alginic acid comprise any one selected from the group consisting of sodium alginate, potassium alginate, and ammonium alginate.

6. The composition according to claim 1, wherein the calcium source comprises at least one selected from the group consisting of calcium carbonate, calcium chloride, calcium citrate, calcium lactate, calcium gluconate, calcium acetate, and calcium phosphate; more preferably, the calcium source is a calcium source of low solubility; still more preferably, the calcium source of low solubility is one selected from the group consisting of calcium lactate, calcium carbonate, and calcium phosphate.

7. The composition according to claim 1, wherein the composition has a particle size of 0.1 mm to 3 mm, more preferably 0.3 mm to 2.5 mm, and still more preferably 0.3 mm to 1 mm.

8. A method for preparing the composition according to any one of claims 1 to 7, characterized by comprising dispersing carboxymethyl cellulose-based materials, alginic acid-based materials, and silicon dioxide in a solution, uniformly mixing and drying to obtain Material A, and then mixing Material A with a calcium source and readily soluble materials.

9. The preparation method according to claim 8, wherein during the preparation of Material A, a polymer concentration in the solution is 4% to 10%, and a drying temperature is 45°C to 100°C; preferably, the drying temperature is 45°C to 80°C.

10. Use of the composition according to any one of claims 1 to 7 in a preparation of a space-occupying product in a digestive tract.