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(57) Abstract: Disclosed herein are compositions comprising blends of at least one triglyceride; at least one fatty acid; and at least one fatty acid derivative, whereby said compositions exhibit improved processing and flame/burn characteristics. The compositions may, optionally, further comprise fillers, extenders, modifiers, binders, flame and article colorants, and perfumes and are useful in the manufacture of, for example, fireplace logs, fire starter blocks, candles, and the like.

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## HYDROCARBON-FREE, NON-POLYMERIC FORMULATIONS AND ARTICLES

### BACKGROUND OF THE INVENTION

#### 1. Field of Invention

The present invention relates to moldable and extrudable compositions that are useful for producing articles, such as candles and artificial firelogs. The compositions, which are flammable and combustible, comprise additives, e.g. oils, fats, and their derivatives, for improving processing and burn characteristics.

#### 2. Description of Related Art

Fatty triglycerides and fatty acids derived from them by hydrolysis and removal of glycerin are known to be employed as fuels in candles, fire starter blocks, fireplace logs, and similar articles. They are mixed with polymeric and /or hydrocarbon (waxy-type) fuels or formulated into composites with various forms of fiber or other solid species, usually flammable materials.

U.S. Patent No. 3,297,419 discloses a synthetic fuel log produced from a flame-supporting material that is solid at room temperature. A filler or extender of a combustible type is combined with the flame-supporting material. The flame-supporting material and the extender are combined with a binder to provide an elongate synthetic fuel log that is said to remain intact during burning.

U.S. Patent No. 3,843,336 discloses a self supporting log having about seventy percent of its fuel value derived from petroleum derivatives contained therein. The log has an extruded thermoset resin skeleton highly filled with particulate vegetable matter. The woody particles are preferably initially impregnated with one or more petroleum derivatives, and subsequently added to an aqueous solution of a prepolymer resin mix. The mixture is then

extruded with preferably a screw extruder having a heated die barrel to concentrate the thermoset resin at the outer surface of the extrusion, forming a continuous casing.

U.S. Patent No. 4,040,796 discloses an artificial firelog comprised of a flammable body typically formed of a particulate wood and wax mixture preferably with salts added for flame color. In a preferred embodiment, by a coextrusion process, a groove is formed along the outer surface of the body and extends substantially its entire length. The groove is filled with gelled starter fuel. The log is preferably formed with at least one substantially flattened side with the gelled fuel filled groove located just above the flattened side whereby the log can be situated with its flattened side on the fireplace grate or andirons and the gelled fuel filled groove located along the lower preferably front, face of the log thereby facilitating the log's fast lighting characteristics. The log is provided with an over-wrapper having an inner low or medium density polyethylene coating and which over-wrapper is heat sealed about the log. When lighting, the over-wrapper is initially lit and this also ignites the gelled fuel. The heat and pressure from the hot expanding combustion gases then softens and ruptures the over-wrapper and allows the flame to spread along the gelled fuel in the notch thereby fully enveloping the firelog in flame.

U.S. Patent No. 4,049,680 discloses organic amide waxes having at least two amide groups per molecule are prepared by reacting monocarboxylic acids preferably fatty acids with organic di- or poly-isocyanates.

U.S. Patent No. 4,326,854 discloses a synthetic firelog made by binding cellulosic particles with a combustible binder produced by the interaction of a liquid combustible by-product or waste product and a modifier, the resulting material being such as to be carbonizable on combustion to form a porous skeleton which maintains the shape of the log

during burning. The by-product may be molasses, various waste oils or pitches, or sulfite lye and the modifier interacts physically or chemically therewith to form a plastic, thixotropic binder material. The cellulosic material may be sawdust, paper, or any of a variety of cellulosic residues from the processing of vegetable materials.

U.S. Patent No. 4,333,738 discloses an improved synthetic fire log that contains wood fibers or sawdust, coal liquid, molasses and paraffin wax in appropriate quantities. Green sawdust is first heated to a temperature of from 250 to 300°F. to reduce the moisture content thereof and provide relatively dry sawdust. To the dry sawdust is added a quantity of coal liquid while the sawdust is at an elevated temperature of at least 200°F. Next, molasses is added to the sawdust, coal liquid combination while the combination is in a temperature range of from 115-150°F. Thereafter, paraffin wax in block form is added while the foregoing mixture is at an elevated temperature at least sufficient to melt the paraffin. Upon thorough incorporation of the materials, the resulting mixture is poured into a mold and compressed to form a stable, solid, compacted mass after which the mass is removed from the mold thus forming a synthetic fire log.

U.S. Patent No. 4,813,975 discloses a composition of matter, which is suitable for manufacturing candles by pressing/extruding, which is based on stearic/palmitic acid and also comprises: 0.2-10% (w.w) of a crystal modifier and optionally 0.5-15% (w.w) of another, as to carbon chain length adjacent, natural fatty acid and/or such fatty acid derivative. Preferably the stearic/palmitic acid is a mixture of stearic acid and palmitic acid in the weight range of 20-80:80-20. The crystal modifier is usually of the ester type and is derived from a polyol with 2-4 hydroxyl groups and higher fatty acid and/or dimeric fatty acid.

U.S. Patent No. 5,868,804 discloses artificial firelogs that provide a natural visual and audible sensation so that the artificial firelogs have both a soothing flickering flame and a crackling or popping sound normally observed only with natural wood logs. The crackling additive can be mustard seeds, sesame seeds and synthetic hollow spheres.

U.S. Patent No. 6,017,373 discloses an artificial firelog that contains 2% to about 6% coriander seed added to create a crackling sound that mimics the sounds produced during the burning of natural logs. The random crackling sound continues for approximately the same time period as observed with the burning of natural wood firelogs and has an amplitude and frequency of crackling sound that mimics burning natural wood logs.

U.S. Patent No. 6,086,644 discloses a scented candle that includes a candle manufacturing material, a fragrance provider, and at least one of polypropylene glycol and polypropylene glycol monoalkyl ether of a specified general formula.

U.S. Patent No. 6,136,054 discloses a synthetic firelog made by mixing specially processed waxed-cardboard or other wax coated papers of varying proportions with a binder/fuel. The binder/fuel consists of a petroleum wax or a mixture of waxes, the nature and extent of which is suitably modified by in situ admixture with the paraffin wax already present in the waxed-cardboard. Sawdust or other wood fillers may also be added. Upon thorough incorporation of the materials, the resulting mixture is extruded, molded, compressed, or otherwise formed, such that the resulting mass is sufficiently solid to hold its shape at normal room temperature.

U.S. Patent No. 6,268,466 discloses a low molecular weight, tertiary amide terminated polyamide that may be blended with a liquid hydrocarbon to form a transparent composition having gel consistency. The tertiary amide terminated polyamide may be prepared by

reacting "x" equivalents of dicarboxylic acid wherein at least 50% of those equivalents are from polymerized fatty acid, "y" equivalents of diamine such as ethylene diamine, and "z" equivalents of a monofunctional reactant having a secondary amine group as the only reactive functionality. The gel contains about 5-50% tertiary amide terminated polyamide, with the remainder preferably being pure hydrocarbon. The gels are said to be useful in formulating personal care products and other articles wherein some degree of gel-like or self-supporting consistency is desired.

U.S. Patent No. 6,503,077 discloses a tertiary amide-terminated dimer acid-based polyamide that may be blended with a solvent to form a gel. The solvent may be flammable, and a wick may be added to the resulting gel so as to form a candle. Depending on the composition, the candle may be formed into a free-standing pillar, or may be better suited to being placed in a container. The solvent may, for example, be mineral oil or triglyceride. A solid coating may be placed around the candle, for advantages including enhancement of the mechanical stability of the gelled body, and elimination of the tendency of a gel to have an oily feel and to accept noticeable fingerprints. The solvent which, in combination with the tertiary amine-terminated dimer acid-based polymer forms a gel, may be or include a fragrance material. The gelled composition may also include fatty acid and/or a compound containing one, two, or more ester groups. In one aspect, the article does not contain a wick, and is intended to function as a fragrance-releasing product.

U.S. Patent No. 6,521,002 discloses a clear transparent candle, which can be scented, made of a composition comprising at least one polyamide resin; at least one aliphatic acid alkyl ester having 16 to 40 carbon atoms total in the acid and alcohol moieties thereof; at least one unsaturated alcohol having 11 to 20 carbon atoms; at least one polyether diol ester; at

least one drying agent selected from the group consisting of saturated alcohols having 14 to 22 carbon atoms, fatty acid amides, and fatty acid bis-amides in specified ranges of proportions, 0 to 3 parts by weight of at least one emulsifier, 0 to 10 parts by weight of at least one fragrance and 0 to 1 part by weight of at least one preservative. The candle can stand alone or be in a container.

U.S. Patent No. 6,544,303 discloses a composite scented candle that has a high load scented shell and a core candle with a similar or lesser fragrance load. The inside candle can be wax, paraffin, gel oil or polyamide-based and can be scented or unscented. The outside shell can be made of paraffin wax and a reinforcement having a melting point of at least 70°C. and is scented at a level so that fragrance is continuously released without burning and is further activated by lighting the core candle.

U.S. Patent No. 6,602,306 discloses an artificial firelog comprising a mixture of combustible materials and a combustible binder and further containing particulate coke comprising about 1% to about 35% by weight of the firelog, the coke particles having a size less than about 4 mm, to create a realistic crackling sound that mimics the sounds produced during the burning of natural logs. The particulate coke has a preferred moisture content between about 2% and about 20% by weight. The particulate coke may be combined in predetermined ratios with other established crackle additives including natural additives such as coriander seed to extend the time period during which the burning artificial firelog crackles. Preferably, the particulate coke comprises metallurgical coke.

U.S. Patent No. 6,706,942 discloses environmentally degradable molded or extruded articles comprising a polyhydroxyalkanoate C4C6 copolymer composition having short annealing cycle times. Such short annealing cycle times are achieved by compositions having

a percentage of C6 units of 2-8%.

U.S. Patent No. 6,712,865 discloses a colored flame candle and the manufacture thereof. The colored flame candle comprises a candle body and a candle combustion wick, wherein the candle body comprises a primary combustion agent, a higher fatty acid amide and a higher fatty acid triglyceride as the strength- and flexibility-modifier, a color-forming agent and conventional adjuvant such perfumes and pigments.

U.S. Patent No. 6,719,816 discloses an artificial firelog comprising combustible cellulosic material, a combustible wax binder and an amount of sodium bicarbonate effective to reduce emissions of particulate matter (PM) and carbon monoxide (CO). The sodium bicarbonate comprises about 1% to about 10% by weight of the artificial firelog. Besides substantially reducing PM and CO emissions during combustion of the artificial firelog, the undesirable odor associated with combustion of the wax binder is decreased and the firelog's total burn time is extended. Moreover, the sodium bicarbonate is said to increase the "poke resistance" of the firelog making the log suitable for outdoor use and a natural appearing white ash develops during the course of combustion.

A number of polymeric materials can be used instead of or in addition to waxes primarily for candle manufacturing. They are polyamides (U.S. Patent No. 6,544,303; PCT WO 2001095871 and PCT Int. Application WO 2001095871), ester-terminated polyamides or tertiary-amide terminated polyamides (U.S. Patent Nos. 6,268,466 and 6,503,077), in-situ polymerized vegetable oils and unsaturated carboxylic acids or anhydrides (German Patent 19619396), polyhydroxy alcoholate copolymers (U.S. Patent No. 6,706,942), polyether diol ester (U.S. Patent No. 6,521,002) and vinyl acetate-ethylene copolymers (U.S. Patent No. 6,712,865).



Several non-polymeric materials have been used as the main component of the formulation. They are ethyl carbamate (Chinese Patent Application Publication 1043340A), polybasic acid esters (Chinese Patent Application Publication 1073201A), trimethyl citrate, di-alkyl oxalate, mono-alkyl succinate, paraformaldehyde, ethyl hydroxyformate, urothropine, and stearic acid (U.S. Patent No. 6,712,865).

Fatty acids, amides, including bis-amides, esters, and calcium stearate are also known as lubricants for numerous plastic materials (primarily thermoplastics). Such lubricants are added at up to ten weight percent to the polymeric materials to ease their processing [See *Plastics Additives Handbook*, 5<sup>th</sup> edition by Hans Zweifel, Hanser Publishers, Munich, 2000].

The disclosures of the foregoing are incorporated herein by reference in their entirety.

### **SUMMARY OF THE INVENTION**

As noted above, fatty triglycerides and fatty acids derived from them by hydrolysis and removal of glycerin are known to be employed as fuels in candles, fire starter blocks, fire logs, and similar articles. They are mixed with polymeric and /or hydrocarbon (waxy-type) fuels or formulated into composites with various forms of fiber or other solid species, usually flammable materials.

Altering the fatty materials with the use of additives for meeting required processing characteristics (hardness, softening point, ease of forming the object in a standard process, strength of the object immediately upon production and at a later time, moldability, mold release, extrudability and the like) of the burnable product and/or changing the nature of the way the combinations burn (flame height, time of burning, flame brightness and color, the ability of the object to hold together while burning etc.) can result in unique and novel compositions, the production of which is not obvious.

Additionally, various interactions among the additives and the fuels can result in both desirable and undesirable effects. Therefore, altering the compositions to achieve all the desired characteristics, while at the same time avoiding the use of both polymeric and traditional hydrocarbons, is especially challenging.

In accordance with the present invention, natural fats or oils are typically used as they occur, generally in a liquid or paste form at ambient temperature. Fatty acids, derived from triglycerides by hydrolysis and removal of the glycerin can also be used as they occur. Some of the properties of each of these can be improved by modifying the fuel stock used. Hydrogenation can be used either on the triglycerides or on the fatty acids to change their physical characteristics by making them harder and more crystalline in nature. The fats and fatty acids can also be blended either in hardened or unhardened state to achieve the desirable properties. Lastly, fats and oils of differing carbon chain distributions can also be utilized in these formulations to produce an even wider variety of properties. Such techniques are known in the art; however, none of these techniques, alone, is sufficient to meet all the required and above-described desirable characteristics of the products involved.

Artificial fire logs (hereinafter referred to as "fireplace logs") are composites that contain cellulosic materials, such as sawdust, and a binder that is produced from certain waxes that may also optionally contain combustible additives that impart processing, combustion characteristics, and flame appearance.

Hydrocarbon-free, non-polymeric formulations based on natural oils, fats, and their derivatives with or without cellulose-containing components are not believed to have been reported as moldable and/or extrudable owing to their very low softening points and poor burning characteristics: short burn time and low flame.

According to the present invention, there are provided non-polymeric, hydrocarbon-free formulations that are based on triglyceride-fatty acid blends (both soft and hard), serving as fuels. The formulations are suitable for manufacturing moldable and/or extrudable compositions and useful articles, such as candles and cellulosic material-filled artificial fire logs or fire starter blocks. Improved processing and flame/burn characteristics are achieved by combining these blends with fatty acid derivatives, such as metal stearates and ethylene bisamides.

Thus, the present invention is directed to a moldable and/or extrudable composition that is suitable for manufacturing useful shaped articles and is based on triglyceride-fatty acid blends that also comprise fatty acid derivatives to achieve improved processing and flame-burning characteristics.

More particularly, the present invention is directed to a composition comprising a blend of:

- (A) at least one triglyceride;
- (B) at least one fatty acid; and
- (C) at least one fatty acid derivative.

In another aspect, the present invention is directed to an article of manufacture comprising a composition comprising a blend of:

- (A) at least one triglyceride;
- (B) at least one fatty acid; and
- (C) at least one fatty acid derivative.

In still another aspect, the present invention is directed to a method for improving the processing and flame-burning characteristics of articles comprising a blend of at least one triglyceride and at least one fatty acid, wherein the method comprises adding to said blend at least one fatty acid derivative.

### DESCRIPTION OF THE PREFERRED EMBODIMENTS

As specified above, the present invention is directed to, inter alia, a composition comprising a blend of:

- (A) at least one triglyceride;
- (B) at least one fatty acid; and
- (C) at least one fatty acid derivative.

The fatty acid(s) of the blends of the present invention is (are) preferably of the formula  $R_1\text{COOH}$ , wherein  $R_1$  is an aliphatic hydrocarbon radical of 5 to 25 carbon atoms, including, but are not limited to, such acids as caprylic, oleic, capric, linoleic, lauric, linolenic, myristic, eicosenoic, palmitic, lauroleic, margaric, myristoleic, stearic, palmitoleic, arachidic, gadoleic, behenic, erucic, pelargonic, elaeostearic, isostearic, licanic, neodecanoic, arachidonic, 2-ethyl hexanoic, lignoceric, caproic, pentadecanoic, hydroxystearic, phenylstearic, and the like. The acids employed can, if desired, be hardened by hydrogenation where they are not saturated to begin with. Mixtures of such acids can, of course, be employed.

The triglyceride(s) of the blends of the present invention is (are) an ester, or esters, of glycerol with one or more fatty acids that are preferably of the formula  $R_1\text{COOH}$ , wherein  $R_1$  is an aliphatic hydrocarbon radical of 5 to 25 carbon atoms, as described above.

The fatty acid derivative(s) of the blends of the present invention are preferably selected from the group consisting of metal salts of fatty acids, fatty acid amides, fatty acid bisamides, and mixtures thereof. Preferably, the fatty acids from which the derivatives are prepared will, again, be of the formula  $R_1\text{COOH}$ , wherein  $R_1$  is an aliphatic hydrocarbon radical of 5 to 25 carbon atoms, as described above.

The metallic moiety of the metal salts of the fatty acids is preferably selected from the group consisting of alkali metals and alkaline earth metals, for example, sodium, potassium, magnesium, calcium, and the like. Sodium and calcium are preferred, calcium most preferred. Again, the fatty acid moiety is preferably of the formula  $R_1\text{COOH}$ , wherein  $R_1$  is an aliphatic hydrocarbon radical of 5 to 25 carbon atoms, as described above. The fatty acid moiety most preferably comprises stearate and/or mixtures of stearate with other naturally occurring and commercially available fatty acids. Again, mixtures of such salts can be employed.

The bisamide waxes, ethylene bisstearamide and methylene bisphenylstearamide, are known and have a number of uses, for example they are used either alone or in admixture with other materials as lubricants for various applications including the compaction of metal powders; the drawing of wire; the extrusion of plastic pipe; sand shell molding; the processing of polystyrene; and as mould release and detackifying agents for synthetic rubbers.

In addition, it is known that such bisamide waxes can be used as additives for a number of materials including paper to improve resistance to water and oil; paraffin waxes and asphalts to increase the melting point thereof; adhesives to reduce viscosity and eliminate cold block and tack. Such bisamide waxes have also been used as anti-static agents for cellophane.

These bisamide waxes are commercially available and can be manufactured by a process in which a fatty acid is reacted with a diamine at a temperature above the melting point of the resulting amide wax; the reaction proceeds with the evolution of water.

The most widely used bisamide wax is ethylene bisstearamide, which can be made by reacting one mole of ethylenediamine with two moles of stearic acid. Similarly, methylene bisphenylstearamide can be prepared by the reaction of stearic acid with methylenedianiline. Alternatively, these products can be made by the reaction of a fatty acid of the formula  $R_1\text{COOH}$ , wherein  $R_1$  is an aliphatic hydrocarbon radical of 5 to 25 carbon atoms, as described above, with an organic isocyanate, as disclosed in U.S. Patent No. 4,049,680.

For use in the practice of the present invention, ethylene bisamides are preferred. Most preferred is N,N'-ethylene bis-stearamide (EBS).

Those skilled in the art will understand that the fatty acid amides that can be employed in the practice of the present invention can be prepared from such fatty acids and monoamines.

The total additive content, i.e., the content of the fatty acid derivative(s), (C), preferably is in the range of from about 3 to about 99 percent by weight based on the total weight of (A), (B), and (C); more preferably, from about 5 to about 20 percent by weight; most preferably, from about 10 to about 15 percent by weight.

The preferred weight ratios between the triglycerides and the fatty acid-containing fuels in the blends of the present invention are in the range of 60/40 to 40/60 for most applications, although wider ranges, such as 90/10 to 10/90, may apply when specific properties, e.g., burn time and/or flame height, are weighted more heavily than others.

In addition to the fatty acids, triglycerides, and fatty acid derivatives of which the compositions and articles of the present invention are comprised, such compositions and articles may, if desired, further comprise additional fillers, extenders, modifiers, binders, flame and article colorants, perfumes, and the like. In particular, where such articles are fireplace logs or fire starter blocks, fillers that are cellulose-containing materials will normally be included. Such cellulose-containing materials can include, but are not limited to, cellulose fiber, cellulose derivatives, wood chips, wood fibers, wood flour, flax, kenaf, sisal, jute, rice hulls, cotton, hemp, and the like.

Suitable cellulose-containing fillers for use in the practice of the present invention may be derived from any of a number of available sources, such as ground wood, sawdust, wood flour, ground newsprint, magazines, books, cardboard, wood pulps (mechanical, stone ground, chemical, mechanical-chemical, refined, bleached or unbleached, virgin or recycled, sludge, waste fines), and various agricultural wastes (rice hulls, wheat, oat, barley and oat chaff, coconut shells, peanut shells, walnut shells, straw, corn husks, corn stalks, jute, hemp, bagasse, bamboo, flax, and kenaf).

Fibrous wood material, which is preferred because of its abundance and suitability can be derived from either soft woods or evergreens or from hard woods commonly known as broad leaf deciduous trees. While soft wood is the primary source of fiber for use in the practice of the present invention, additional fiber make-up can be derived from a number of secondary or fiber reclaim sources, including bamboo, rice, sugar cane, flex, kenaf and recycled fibers from newspapers, boxes, computer printouts, and the like.

Where fillers are included in the compositions of the present invention, they are preferably present in the range from about 1 to about 60 percent by weight, based on the weight of the total composition.

Various features and aspects of the present invention are illustrated further in the examples that follow. While these examples are presented to show one skilled in the art how to operate within the scope of the invention, they are not intended in any way to serve as a limitation upon the scope of the invention.

### **Examples**

#### **Softening Point Improvement**

An important characteristic of a fuel, whether or not combined with a fibrous material, is the ability to hold together the formed article in a cohesive unit that doesn't melt and flow off or slump into an unusable pile. This property is dependent upon more than just the melting point. A test method, the Ring and Ball Test, was used to determine the suitability of formulations in this regard.

The Ring and Ball Test measures strength as the melting point of the fuel is approached: a  $\frac{1}{4}$  inch thick disc made up of the sample, e.g., a composite, must support a steel ball  $\frac{3}{8}$  of an inch in diameter and weighing 3.5 to 3.6 grams. This is a variation of other ring and ball softening point tests, such as ASTM D 36, and uses the same equipment. The sample disc is  $\frac{5}{8}$  of an inch in diameter in the bottom 0.1 inch of thickness, and  $\frac{3}{4}$  of an inch in diameter in the top 0.15 inch of thickness, thus resting on a lip in the sample holder. The steel ball sits on top of the sample disc, held roughly centered by a retainer with three prongs that keep the ball from rolling. Two of the sample holders, sample discs, balls, and retainers are mounted in a framework and immersed in water in a 1000 mL beaker, with the samples



suspended about midway up the beaker. The water is heated on a heating plate at a rate of about 5°F ( about 2.75°C) per minute, with moderate stirring of the water by a 30 mm Teflon coated magnetic stirring bar. The temperature is read from a thermometer with its bulb suspended at about the sample level, between the two samples. The temperature at which each ball falls through the sample disc is recorded, and the average value is used as the softening point of that composition.

### **Sample Preparation**

Samples are prepared by weighing appropriate amounts of the fuel and additives into a 250 mL beaker, then adding a 30 mm magnetic stirring bar. The beaker is then placed on a heater/stirrer, and a thermometer is suspended in the beaker so that its bulb does not touch the bottom. The heater is then turned on to a level to produce a surface temperature in the range of 300 to 350°F (149 to 177°C) and the stirrer turned on when enough of the sample is molten to allow mixing. The sample is heated and stirred until it is a clear solution - normally above 220 °F (104°C).

For example, 97.0 grams of hard tallow (Iodine value below 5) were weighed into a tared 250 mL beaker along with 3.0 grams of ethylene bisstearamide in prill form. Heating is started and by the time a temperature of about 170°F (76°C) is reached, there is enough melted hard tallow to begin stirring. Heating and stirring are continued until a clear solution is achieved at about 270°F (132°C). In the case of stearates, a slight haze may be left in the solution. Two sample holders were placed on a clean ceramic surface, and the resulting composition poured carefully into them until they just ran over. The samples were allowed to cool into solid discs and stabilized for at least three hours at room temperature. They were then tested as described above.

## **Burn Time and Flame Height**

### **1. Fire Log-type Composites**

In order to measure the effects of various additives on the burning characteristics of articles, such as burn time and flame height, samples of fuels modified by the additives were mixed with wood chips on a 50/50 basis (by weight) and formed by finger pressure into 1.5 inch diameter by a 3/4 inch high conical piles. These comprised 5 grams of wood chips and 5 grams of a fuel/additive blend, each on a 70 mm aluminum weighing cup, arranged in a matrix on a metal surface. Each was ignited with the use of flame about a 3/4 inch long from a Harbor Freight brand butane microtorch that was applied for one second. Flame characteristics were observed and recorded as shown in Table 3. Burn times were recorded in minutes until the flame went out. Flame heights were recorded in centimeters, measured as average heights during stabilized burn by placing a ruler behind the sample and visually measuring the middle of the flickering height.

### **2. Candle Formulations**

Burn time and flame height of the candle-type formulations and articles were determined on samples cast into cylindrical discs in ring and ball sample holders with wicks suspended in their centers, with the wicks cut off at a height of 0.5 inch above the porcelain surface. Samples were allowed to harden, removed from the rings, and the wicks were ignited as described above. Flame height was measured from candle top to middle of the flickering range, against a ruler as described above. Burn time was recorded in minutes until the flame went out.

The data presented in Table 1 demonstrate the unique ability of ethylene bisamides, especially N,N'-ethylene bis-stearamide (EBS), to increase substantially the ring and ball softening points of triglyceride fuels. Metal stearates, of which calcium stearate (CaSt) was the most efficient, resulted in a less pronounced effect for tallow, hard tallow, and tallow fatty acids. In the case of soft tallow fatty acids, the EBS provides the only effective treatment and even then the effect is small. CaSt was more efficient than EBS for improving the softening point of hard tallow fatty acids, while sodium stearate showed no increase at all in either triglycerides or fatty acids.

Table 1

Effect of Additives on Softening Point of Individual Fuels in Wood-containing Composites

<b>Fuel type</b>	<b>Additive</b>	<b>Additives Concentration, %</b>	<b>Softening Point, °F</b>
Tallow (T)	None	0	103
Tallow	EBS	3	118
Tallow	EBS	5	198
Tallow	EBS	7	205
Tallow	CaSt	2	115
Tallow	CaSt	4	118
Hard Tallow (HT)	None	0	141
Hard Tallow	EBS	3	143
Hard Tallow	EBS	5	170
Hard Tallow	EBS	7	212+
Hard Tallow	CaSt	4	142
Hard Tallow	NaSt	4	141
Tallow Fatty Acids (TFA)	None	0	112
Tallow Fatty Acids	EBS	3	118
Tallow Fatty Acids	EBS	5	126
Tallow Fatty Acids	CaSt	4	114
Tallow Fatty Acids	CaSt	6	116

Hard Tallow Fatty Acids (HTFA)	None	0	144
Hard Tallow Fatty Acids	EBS	7	148
Hard Tallow Fatty Acids	CaSt	2	150
Hard Tallow Fatty Acids	CaSt	3	153
Hard Tallow Fatty Acids	CaSt	4	161
Hard Tallow Fatty Acids	CaSt	5	164
Hard Tallow Fatty Acids	CaSt	6	185
Hard Tallow Fatty Acids	NaSt	4	142

The results in Table 2 show an unexpected, synergistic effect of the fuel compositions [hard triglyceride (HT), soft triglyceride (T), hard triglyceride fatty acid (HTFA), or soft triglyceride fatty acid (TFA)] and the presence of EBS and/or CaSt on increasing the softening point of the formulations.

**Table 2**

**Effect of Additives on Softening Point of Combined Fuels in Wood-containing Composites**

<b>Fuel 1, %</b>	<b>Fuel 2, %</b>	<b>Additive 1, %</b>	<b>Additive 2, %</b>	<b>Softening Point, °F</b>
HT, 50	TFA, 50	0	0	128
HT, 47.5	TFA, 47.5	EBS, 5	0	142
HT, 47.5	TFA, 47.5	CaSt, 5	0	144
HT, 45	TFA, 45	EBS, 5	CaSt, 5	153

T, 50	HTFA, 50	0	0	135
T, 47.5	HTFA, 47.5	EBS, 5	0	156
T, 47.5	HTFA, 47.5	CaSt, 5	0	172
T, 45	HTFA, 45	EBS, 5	CaSt, 5	176

The effect of calcium stearate, an increase in burn time with only a slight reduction in flame height in each set, was unexpected (see Table 3). Both CaSt and EBS showed a tendency to reduce flame height only slightly for all fuel types. Tallow fatty acids burned the most quickly. All fatty acid-containing formulations burned with a higher flame than the triglycerides.

**Table 3****Effect of Additives on Burn Time and Flame Height of Wood-containing Composites**

<b>Fuel Type</b>	<b>Additive</b>	<b>Concentration, %</b>	<b>Flame Height, cm</b>	<b>Burn time, min.</b>
Tallow (T)	None	0	7.5	9.0
Tallow	EBS	5	6.3	10.0
Tallow	CaSt	5	6.3	13.5
Hard Tallow (HT)	None	0	7.5	11.0
Hard Tallow	EBS	5	6.3	11.0
Hard Tallow	CaSt	5	5.8	14.0
Tallow Fatty Acids (TFA)	None	0	11.3	8.0
Tallow Fatty Acids	EBS	5	10.0	8.5
Tallow Fatty Acids	CaSt	5	8.8	11.0
Hard Tallow Fatty Acids (HTFA)	None	0	10.0	9.5
Hard Tallow Fatty Acids	EBS	5	9.5	10.0
Hard Tallow Fatty Acids	CaSt	5	8.8	12.0

It was surprising to discover that the careful balancing of the composition of the fuel (fatty acids or triglycerides, soft or hydrogenated) with the proper selection of additives promotes suppressing and/or improving undesirable properties in order to attain the required performance characteristics of both formulations and the articles produced from them.

Thus, in attempting to make a suitable candle material, one might combine either hardened tallow or tallow (to get a base fuel that would set up into a suitably firm, but somewhat flexible, shape) with either tallow fatty acids or hard tallow fatty acids (to increase the height of the flame) and, notwithstanding, still produce an article that slumps at too low a temperature, has an insufficient visible flame, burns for too short a time, and/or takes too long to harden in the mold.



Proper application of the EBS additive caused rapid crystallization of the candle-type formulation in the mold and provided superior strength and stiffness of the produced article even when it was still warm. As shown in Table 4, CaSt increased the length of time the material burns.

Sodium stearate provided a highly visible, bright flame. It is known in the art that the addition of sodium stearate to a fuel contributes to a bright yellow flame.

**Table 4**

**Effect of Additives and Fuel Compositions on Properties of Candle-type Formulations**

<b>Fuel 1,%</b>	<b>Fuel 2,%</b>	<b>EBS,%</b>	<b>CaSt,%</b>	<b>NaSt,%</b>	<b>Softening Point, °F</b>	<b>Flame height, cm</b>	<b>Burn time, min.</b>
HT, 50	TFA, 50	0	0	0	128	1.3	4.0
HT, 35	TFA, 54	5	5	1	149	2.0	4.5
HT, 44	TFA, 45	5	5	1	152	1.3	8.0
HT, 55	TFA, 34	5	5	1	154	1.0	9.0
T, 50	HTFA, 50	0	0	0	135	1.5	7.0
T, 35	HTFA, 54	5	5	1	170	2.0	6.0
T, 44	HTFA, 45	5	5	1	175	1.3	10.0
T, 55	HTFA, 34	5	5	1	178	1.3	11.0

In the case of a composite structure, such as a fire log based on wood chips and fatty fuel mixtures (see Table 5), the combination of additives added strength and stiffness, held the structure together while burning, controlled the length of burn, and provided "quick

strength" during post-production while the crystallization of the fuel mixture into a suitable waxy structure takes place.

Therefore, complex blends of soft and hardened hydrocarbon-free, non-polymeric triglyceride-fatty acids (used as fuels) were found to be suitable for making useful articles, such as fire logs and candles, in the presence of fatty acid-derived additives, such as metal stearates and ethylene bisamides. The use of such compositions yielded articles having the necessary processing and burn characteristics.

**Table 5**

**Effect of Additives and Fuel Compositions on the Characteristics of Wood-containing Composites**

<b>Fuel 1,%</b>	<b>Fuel 2,%</b>	<b>EBS,%</b>	<b>CaSt, %</b>	<b>NaSt,%</b>	<b>Softening Point, °F</b>	<b>Flame height, cm</b>	<b>Burn time, min.</b>	<b>Visible Color</b>
HT, 50	TFA, 50	0	0	0	128	8.8	9.0	Blue
HT, 35	TFA, 54	5	5	1	149	8.8	12.5	Yellow
HT, 44	TFA, 45	5	5	1	152	7.5	13.0	Yellow
HT, 55	TFA, 34	5	5	1	154	6.3	13.5	Yellow
T, 50	HTFA, 50	0	0	0	135	8.8	9.5	Blue
T, 35	HTFA, 54	5	5	1	170	8.8	12.0	Yellow
T, 44	HTFA, 45	5	5	1	175	7.5	13.0	Yellow
T, 55	HTFA, 34	5	5	1	178	7.0	14.0	Yellow

In view of the many changes and modifications that can be made without departing from principles underlying the invention, reference should be made to the

appended claims for an understanding of the scope of the protection to be afforded the invention.

## CLAIMS

### What is claimed is:

1. A composition comprising a blend of:
  - (A) at least one triglyceride;
  - (B) at least one fatty acid; and
  - (C) at least one fatty acid derivative.
2. The composition of claim 1 wherein the triglyceride/fatty acid ratio is between 90/10 and 10/90 by weight.
3. The composition of claim 1 wherein the fatty acid derivative(s) is (are) selected from the group consisting of metal salts of fatty acids, fatty acid amides, fatty acid bisamides, and mixtures thereof.
4. The composition of claim 3 wherein fatty acid derivatives are selected from the group consisting of calcium stearate, sodium stearate, N,N'-ethylene bisstearamide, and mixtures thereof.
5. The composition of claim 1 wherein the content of the fatty acid derivative(s), (C), is from about 3 to about 99 percent by weight, based on the total weight of components (A), (B), and (C).

6. The composition of claim 1 further comprising at least one additive selected from the group consisting of fillers, extenders, modifiers, binders, flame and article colorants, and perfumes.
7. An article of manufacture comprising a composition comprising a blend of:
  - (A) at least one triglyceride;
  - (B) at least one fatty acid; and
  - (C) at least one fatty acid derivative.
8. The article of claim 7 wherein the composition further comprises at least one additive selected from the group consisting of fillers, extenders, modifiers, binders, flame and article colorants, and perfumes.
9. The article of claim 7 wherein the fatty acid derivative(s) is (are) selected from the group consisting of metal salts of fatty acids, fatty acid amides, fatty acid bisamides, and mixtures thereof.
10. The article of claim 9 wherein fatty acid derivatives are selected from the group consisting of calcium stearate, sodium stearate, N,N'-ethylene bisstearamide, and mixtures thereof.

11. The article of claim 7 wherein the content of the fatty acid derivative(s), (C), is from about 3 to about 99 percent by weight, based on the total weight of components (A), (B), and (C).
12. The article of claim 8 wherein a filler is present, said filler being selected from the group consisting of cellulose fiber, cellulose derivatives, wood chips, wood fibers, wood flour, flax, kenaf, sisal, jute, rice hulls, cotton, and hemp.
13. The article of claim 7 wherein said article is a fireplace log or fire starter block.
14. The article of claim 12 wherein said article is a fireplace log or fire starter block.
15. The article of claim 7 wherein said article is a candle.
16. A method for improving the processing and flame-burning characteristics of articles comprising a blend of at least one triglyceride and at least one fatty acid, wherein the method comprises adding to said blend at least one fatty acid derivative.
17. The method of claim 16 wherein the fatty acid derivative(s) is (are) selected from the group consisting of metal salts of fatty acids, fatty acid amides, fatty acid bisamides, and mixtures thereof.

18. The method of claim 17 wherein fatty acid derivatives are selected from the group consisting of calcium stearate, sodium stearate, N,N'-ethylene bisstearamide, and mixtures thereof.
19. The method of claim 16 wherein the content of the fatty acid derivative(s) is from about 3 to about 99 percent by weight, based on the total weight of the triglyceride(s), the fatty acid(s), and the fatty acid derivative(s).
20. The method of claim 16 wherein the blend further comprises at least one additive selected from the group consisting of fillers, extenders, modifiers, binders, flame and article colorants, and perfumes.

# INTERNATIONAL SEARCH REPORT

International application No

PCT/US2007/024989

## A. CLASSIFICATION OF SUBJECT MATTER

INV. C11C5/00 C10L5/44 C10L11/04 C10L9/10 C10L10/00

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)

C11C C10L

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 practical, search terms used)

EPO-Internal, WPI Data, COMPENDEX, FSTA, BIOSIS

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No
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X	GB 1 387 711 A (AVON PROD INC) 19 March 1975 (1975-03-19) page 1, column 2, paragraph 2-4; example 4	1-20
X	US 22 592 A (TATUM, JOEL, H.) 11 January 1859 (1859-01-11) the whole document	1-15
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☒ Further documents are listed in the continuation of Box C.

☒ See patent family annex.

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# INTERNATIONAL SEARCH REPORT

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C(Continuation). DOCUMENTS CONSIDERED TO BE RELEVANT

Category*	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
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X	KOWALSKI ET AL: "Chemical and enzymatic interesterification of a beef tallow and rapeseed oil equal-blend" EUROPEAN JOURNAL OF LIPID SCIENCE AND TECHNOLOGY, WILEY VCH VERLAG, WEINHEIM, DE, vol. 106, 13 October 2004 (2004-10-13), pages 655-664, XP002347668 ISSN: 1438-7697 table 4	1-11
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Information on patent family members

International application No

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