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PROPELLENT POWDER AND PROCESS OF MANUFACTURE

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10 Claims. (Cl. 52—13)

This invention relates to the production of non-solvent propellent powders. For the preparation of the socalled solventless propellent explosives, it has been customary to make use of the chemical substance diethyl diphenyl urea, as a gelatinizing agent. In conjunction with nitroglycerine and under the influence of heat and pressure, it is possible to produce a gelatinized nitroglycerine nitrocellulose propellent explosive in the form of flakes or cords as desired, without the use of any volatile solvent such as acetone, the diethyl-diphenyl-urea acting as combined gelatinizing agent, stabilizing agent and moderant. Such a propellent explosive possesses many well known advantages.

Diethyl-diphenyl-urea in practice suffers from the disadvantage that on account of its low solubility in nitroglycerine at ordinary or room temperature, it is impossible to dissolve a sufficient amount of the diethyl-diphenyl-urea in nitroglycerine to form the usually required composition of the propellent explosive. Thus, taking proportions commonly employed, it is impossible to dissolve at room temperature 9 parts of diethyl-diphenyl-urea in 41 parts of nitroglycerine for mixing with 50 parts of nitrocellulose. It is true that this proportion of diethyl-diphenyl-urea can be dissolved in nitroglycerine by heating the latter to a temperature of 30° C. to 35° C., but such a proceeding is objectionable as a manufacturing operation on account of the increased risk of accident, and further, on lowering the temperature of the nitroglycerine-diethyl-diphenyl-urea solution, the excess of diethyl-diphenyl-urea separates out as crystals.

A general method of introducing the diethyl-diphenyl-urea is in the form of powdered crystals added to the nitrocellulose either before, during, or after mixing with the nitroglycerine. This method is objectionable on account of the difficulty of ensuring uniform distribution of the three constituents of the mixture, namely, fibrous nitrocellulose, liquid nitroglycerine and solid diethyl-diphenyl-urea. Such imperfect mixing causes irregularities in gelatinization which cannot be completely removed, even by an increased number of operations in the later stages of production, such as prolonged treatment between heated rolls.

Despite this serious objection, the use of diethyl-diphenyl-urea as a gelatinizing agent in the production of the so-called solventless propellent powders has been well established, chiefly because of the many advantages which this compound possesses for the production of propellent explosives.

The object of the present invention is to provide a process which, while producing solventless propellent explosives that retain the desirable properties conferred by diethyl-diphenyl-urea, at the same time affords a means whereby the gelatinizing agent can be entirely dissolved in the nitroglycerine, and this solution uniformly mixed with the nitrocellulose.

We have now discovered that phenyl urethane, when mixed in the correct proportions, with diethyl-diphenyl-urea, forms a mixture that remains liquid at room temperature and is miscible with nitroglycerine at room temperature with no separation of crystals even on long standing. The phenyl urethane has a melting point of 52° C., and in our researches has been found to possess gelatinizing and stabilizing properties to an outstanding and very high degree.

The following summarizes the properties of the mixtures:—

% Phenyl urethane	% Diethyl-diphenyl-urea	Setting point or state at 15° C.
62.5	37.5	24.0° C.
58.8	41.2	Liquid at 15° C.
55.5	44.5	Liquid at 15° C.
52.6	47.4	Liquid at 15° C.
50.0	50.0	Liquid at 15° C.
47.6	52.4	26.5° C.

This formation with centralite of a mixture that is liquid at room temperature is a particular property of the phenyl urethane that could not be foreseen, as phenyl benzyl urethane, which is a liquid itself at ordinary temperatures, and has other suitable properties, only forms a liquid mixture with diethyl-diphenyl-urea when the proportion of phenyl benzyl urethane exceeds 75%. Further, when the liquid mixture of diethyl-diphenyl-urea and phenyl urethane is added to nitroglycerine, no precipitation of centralite takes place. The result was not to be expected, as it could not be foreseen that the presence of a relatively small quantity of phenyl urethane in the nitroglycerine would be sufficient so to alter the dissolving powers of the latter as to keep the diethyl-diphenyl-urea in solution.

As an illustration of the properties of a composition of diethyl-diphenyl-urea and phenyl urethane the following is given:—

Equal quantities of phenyl urethane and diethyl-diphenyl-urea are melted together, and

the mixture is cooled to room temperature, at which it remains in the liquid state. Eighteen (18) parts by weight of the mixture, equivalent to 9 parts by weight of diethyl-diphenyl-urea plus 9 parts by weight of phenyl urethane, are added to 41 parts by weight of nitroglycerine at room temperature. This results in a homogeneous liquid mixture from which no crystals separate, whereas a mixture of 9 parts diethyl-diphenyl-urea and 41 parts nitroglycerine at room temperature consists of a saturated solution containing a considerable amount of undissolved diethyl-diphenyl-urea, and is therefore not homogeneous.

It is therefore merely necessary to add, after cooling, the requisite amount of the phenyl urethane and diethyl-diphenyl-urea (melted toegther in the correct proportions) to the nitroglycerine at room temperature, and to mix this solution with nitrocellulose suspended in water by any of the well known methods, such as spraying or air agitation. There is thus obtained a uniform mixture of all the constituents, which is in the most favourable condition for the subsequent operation of gelatinization. By this method the gelatinization is then more thorough, and is carried more easily to completion than when diethyl-diphenyl-urea alone is used, thus leading to an increase of output and reduction in cost in conjunction with a considerably improved product. The risk of fire at the rolling stage is also greatly reduced.

As far as the above material is concerned, it may be said briefly therefore that we employ in non-solvent propellent explosives mixtures of diethyl-diphenyl-urea and phenyl urethane that are liquid at normal temperatures.

We have continued our research in this work and have found that we can with safety also use liquid mixtures of diethyl-diphenyl-urea, phenyl urethane and nitroglycerine, whether the mixtures of diethyl-diphenyl-urea and phenyl urethane themselves are liquid or not, provided that when admixed with the nitroglycerine, the mixture is liquid.

All mixtures of phenyl urethane and diethyl-diphenyl-urea containing more than 63 per cent phenyl urethane are solid at room temperature but 9 parts of any of those mixtures will dissolve in 41 parts of nitroglycerine to give a liquid from which no crystals separate at 0° C., even if it is seeded with crystals of phenyl urethane or diethyl-diphenyl-urea or both and allowed to stand with occasional stirring for four hours.

Again, a 48:52 mixture of phenyl urethane and diethyl-diphenyl-urea is solid at room temperature and behaves with nitroglycerine exactly as do the mixtures described above; at 35:65 mixture behaves in the same way.

From mixtures such as the above, but containing appreciably more than 65% of diethyl-diphenyl-urea, crystals separate when the liquid formed by dissolving 9 parts of this gelatinizing mixture in 41 parts nitroglycerine, is cooled to 0° C. or cooled and kept at 0° C. Thus a 30:70 mixture dissolves very readily in nitroglycerine at room temperature, but if the liquid is cooled to 0° C. and seeded as described above with small crystals, within two hours it becomes cloudy; a 20:80 mixture still dissolves at room temperature, but on cooling to 0° C., crystals separate in half an hour.

We can therefore avoid the absence of a solid gelatinizer and facilitate the manufacture of the so-called solventless cordite by employing a liquid mixture of phenyl urethane diethyl-diphenyl-urea and nitroglycerine, whether the mixture of diethyl-diphenyl-urea and phenyl urethane be liquid or solid, provided, as we have said above, that the mixture of the three materials is liquid.

Mixtures of phenyl urethane and diethyl-diphenyl-urea, containing a proportion of diethyl-diphenyl-urea greater than that which would form a liquid mixture with phenyl urethane, are preferable, from the point of view of manufacture to mixtures of phenyl urethane and diethyl-diphenyl-urea containing a proportion of phenyl urethane greater than that which would form with diethyl-diphenyl-urea a liquid mixture, and to the liquid mixtures of phenyl urethane and diethyl-diphenyl-urea themselves. A suitable mixture consists of two parts of diethyl-diphenyl-urea and one part of phenyl urethane. This mixture, since it contains more than 50% of diethyl-diphenyl-urea, is solid at 15° C., but, since it contains little more than 65% of diethyl-diphenyl-urea, crystals do not separate from the mixture with nitroglycerine at 0° C.

By mixing phenyl urethane and diethyl-diphenyl-urea in proportions substantially similar to those given above, we can also utilize the extremely good stabilizing action of phenyl urethane, the good gelatinizing action of diethyl-diphenyl-urea and the liquidity of the gelatinizer-stabilizer nitroglycerine mixture. It might also be added that if the phenyl urethane be dissolved first, this appears to facilitate the solution of the diethyl-diphenyl-urea in the mixture.

The mixture, in brief, consists in a process for the production of non-solvent propellent explosives in which the so-called gelatinizing agent is a mixture of phenyl urethane and diethyl-diphenyl-urea, the mixture being such that it remains liquid at normal temperatures, and is miscible with nitroglycerine at such temperatures with no separation of crystals, even on long standing.

The invention also consists in a modification of the process according to the preceding paragraph, in which the mixture of phenyl urethane and diethyl-diphenyl-urea is not of itself liquid at normal temperatures, but when admixed with the nitroglycerine the mixture is such that no separation of crystals occurs, even on long standing, and the mixture is liquid at normal temperatures.

The invention also consists in methods for the production of non-solvent propellent explosives substantially as herein described, and in non-solvent propellent explosives such as may be made by methods substantially as herein described.

The following example illustrates how the invention may be carried into effect, references to parts being to parts by weight.

Example

Equal parts of phenyl urethane and diethyl-diphenyl-urea are melted together and cooled to room temperature. Nine (9) parts of this liquid are added to 41 parts of nitroglycerine at room temperature, and thoroughly mixed and the homogeneous mixture thus obtained is mixed by air agitation with 50 parts of nitrocellulose suspended in water. The mixture containing all the constituents of the epropellent explosive is separated from the bulk of the water by settling and is then squeezed to effect a further separation of water. The wet lumps are broken down

by hand, and the material is passed through a coarse sieve (4 meshes to the inch). The material is then passed through steam-heated rolls, which express a further quantity of water and yield a product containing about 10% of water. After drying, the material is gelatinized by passage through heated rolls. If desired in flake form, the material is then rolled down to the required thickness, and cut to the required size. If the non-solvent propellant explosive is desired in cords, the sheets are cut into discs, extruded from a hydraulic press heated by a hot water jacket to 75° C., at a pressure of 5000 lbs. per square inch through a die having holes of the appropriate size. The extruded cords are then cut to the desired length.

The resultant propellant explosive is then ready for use.

General

The ballistic properties of the propellent explosives prepared as described above are substantially of the same level as those of a propellent explosive containing the same proportions of nitroglycerine and nitrocellulose with 9 parts of diethyl-diphenyl-urea.

The replacements of a portion of the diethyl-diphenyl-urea by an equivalent amount of phenyl urethane does not reduce the life on prolonged storage of the improved propellent explosive, while its greater homogeneity increases its inherent stability and the regularity and uniformity of its ballistic properties.

The invention is not limited to the method of working outlined in the above examples. Any suitable method of mixing the constituents, separating the water, gelatinizing, rolling or pressing the material may be employed, and the liquid mixture of diethyl-diphenyl-urea, phenyl urethane and nitroglycerine, may be prepared in any desired way, and in any appropriate proportions.

As many apparently widely different embodiments of this invention may be made without departing from the spirit and scope thereof, it is to be understood that we do not limit ourselves to the foregoing examples or descriptions except as indicated in the following patent claims.

We claim:

1. A composition of matter comprises nitrocellulose, nitroglycerin, diethyldiphenylurea and phenyl urethane.

2. A composition of matter comprising nitrocellulose, nitroglycerin, and approximately two parts of diethyldiphenylurea and one part of phenyl urethane.

3. A composition of matter comprising nitrocellulose, nitroglycerin, diethyldiphenylurea and phenyl urethane in liquid form.

4. A composition of matter comprising nitroglycerine, diethyldiphenylurea and phenyl urethane.

5. A composition of matter comprising nitroglycerine, diethyldiphenylurea and phenyl urethane in liquid form.

6. A propellent explosive comprising nitroglycerine, nitrocellulose, diethyldiphenylurea, and phenyl urethane.

7. A propellent explosive comprising approximately nine parts of a homogeneous mixture of phenyl urethane and diethyldiphenylurea, approximately 41 parts of nitroglycerine, and approximately 50 parts of introcellulose.

8. The process of manufacturing propellent explosives which comprises incorporating in nitroglycerine a composition comprising diethyldiphenylurea and phenyl urethane.

9. The process of manufacturing solventless propellent explosives which comprises melting together phenyl urethane and diethyldiphenylurea, adding the liquid components to nitroglycerine at approximately room temperature, and incorporating the resulting product with nitrocellulose.

10. The process of manufacturing propellent explosives which comprises forming a liquid mixture of phenyl urethane and diethyldiphenylurea, cooling the liquid to room temperature, adding the resulting constituents to nitroglycerine, uniformly mixing the nitroglycerine composition to a suspension of nitrocellulose in water, separating the explosive components from the water, and gelatinizing the product.

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CERTIFICATE OF CORRECTION.

Patent No. 1,923,849. **August 22, 1933.**

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It is hereby certified that error appears in the printed specification of the above numbered patent requiring correction as follows: Page 1, line 99, for "centralite" read "diethyl-diphenyl-urea"; page 2, line 58, for "at" read "a"; and line 147, for "epropellent" read "propellent"; and that the said Letters Patent should be read with these corrections therein that the same may conform to the record of the case in the Patent Office.

Signed and sealed this 24th day of October, A. D. 1933.

(Seal) **Acting Commissioner of Patents.**