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(54) **FLEXIBLE POLYURETHANE FOAM, PROCESS FOR ITS PRODUCTION, AND SEAT FOR AUTOMOBILE USING THE FLEXIBLE POLYURETHANE FOAM**

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See application file for complete search history.

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(57) **ABSTRACT**

To provide a flexible polyurethane foam having good vibration characteristics and suitable particularly for a seat for an automobile.

A flexible polyurethane foam obtained by reacting a high molecular weight polyoxyalkylene polyol or a polymer-dispersed polyol containing fine polymer particles in the high molecular weight polyoxyalkylene polyol, with a polyisocyanate compound, in the presence of a catalyst, a blowing agent and a foam stabilizer, characterized in that an amino-modified silicone (F) having a silicon atom and a nitrogen atom in its molecule is used in an amount of from 0.00001 to 1 part by mass per 100 parts by mass of all active hydrogen compounds.

14 Claims, No Drawings

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**FLEXIBLE POLYURETHANE FOAM,
PROCESS FOR ITS PRODUCTION, AND SEAT
FOR AUTOMOBILE USING THE FLEXIBLE
POLYURETHANE FOAM**

TECHNICAL FIELD

The present invention relates to a novel flexible polyurethane foam, suitable as a urethane cushion material providing good riding comfort, particularly for a seat for an automobile.

BACKGROUND ART

In recent years, in the field of polyurethane foams useful for various applications, various researches and developments have been made in order to improve the characteristics suitable for the respective applications. For example, along with upgrading of automobile seats in order to improve the riding comfort of seat cushion, it is targeted to improve the rebound resilience, vibration characteristics, durability, etc. With respect to the vibration characteristics, the influence of car body vibration over a human body varies depending upon the vibration frequency. However, it is considered effective for improvement of the riding comfort to take damping of vibration particularly large in a frequency range (for example from 4 to 8 Hz or from 6 to 20 Hz) to which a human is sensitive. Further, it is considered that in order to improve these characteristics, a seat cushion is effective which employs a polyoxyalkylene polyol having a higher molecular weight than one heretofore produced.

On the other hand, as a seat cushion, a combination of a metal spring with a pad material made of a flexible polyurethane foam (hereinafter referred to simply as a flexible foam) has been used in many cases. However, in recent years, there has been a trend of employing an automobile seat so-called a deep foam type whereby a metal spring is abolished by imparting spring characteristics to the flexible foam itself to meet the demand for e.g. cost down, reduction of weight, etc. The deep foam type seat has become thick, since no metal spring is used in combination.

Further, the characteristics of the flexible foam have become a factor substantially influential over the sitting comfort and riding comfort of the seat. Namely, the static characteristics and dynamic characteristics as indices for the sitting comfort and riding comfort, have been regarded as important in the development of flexible foams. Among the static characteristics, it is particularly important to control the supported feeling at the initial stage of sitting and the bottom-hitting feeling at the final stage of sitting.

When a person actually sits on a seat provided with a pad material of flexible foam, the flexible foam will be compressed and deflected, and the position of e.g. the hip will sink to a certain height. As a method for measuring this static characteristic (the static sitting feeling), a test method may be employed wherein the deflection is measured in a load test in accordance with a performance test method for a pad material for an automobile seat according to JASO automobile standard B408-89 (1989), to obtain a load-deflection curve, or the deflection under a load of 500N (Newton Load) may be used which is obtained from a load-deflection curve obtained by the measurement employing a pressure plate in accordance with JIS E7104 (2002). This pressure plate is oval with a long diameter A of 300 mm, a short diameter B of 250 mm and a thickness C of at least 35 mm and is so-called Tekken Plate.

On the other hand, the polyoxyalkylene polyol to be used

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lene oxide, using a sodium/potassium catalyst such as sodium hydroxide or potassium hydroxide and an initiator such as a polyhydric alcohol. By this production method, an unsaturated monool having an unsaturated bond (hereinafter referred to simply as a monool) will be formed as a by-product, and the amount of such a monool produced, will increase with an increase of the molecular weight of the polyoxyalkylene polyol (a decrease of the hydroxyl value).

In the case of a polyoxyalkylene polyol having a hydroxyl value of about 56 mgKOH/g which is commonly used as a material for resilient polyurethane foams, the amount of such a monool produced, is not so large as to bring about a problem. However, in the case of a polyoxyalkylene polyol having a high molecular weight and a low hydroxyl value, the amount of such a monool produced, will be problematic. Namely, in a case where a resilient polyurethane foam is produced by using a polyoxyalkylene polyol having a high monool content (a high total unsaturation value), there will be a problem such as a decrease in hardness or a worse in compression set of the produced foam, or a worse in curing property at the time of production of the foam. Further, even if it is attempted to produce a polyoxyalkylene polyol having a low hydroxyl value by using a sodium/potassium catalyst, the amount of the monool produced will be so large that such production will be practically difficult.

Under the circumstances, a method has been proposed wherein in order to improve the characteristics such as the riding comfort, durability, etc. for an automobile seat, a polyoxyalkylene polyol having a low monool content is used for the production of a resilient polyurethane foam (Patent Document 1).

However, it has been found that a resilient polyurethane foam represented by a deep foam type produced by using a polyoxyalkylene polyol having a low monool content, has an extremely high rebound resilience (rebound resilience of core portion: 71 to 74%), whereby the riding comfort is inadequate from the viewpoint of the occupant posture-stability performance or supporting performance during traveling. In order to solve such problems, an invention has been proposed to suppress the rebound resilience by a combined use of a polyoxyalkylene polyol having a low unsaturation value and a polyoxyalkylene polyol having a low molecular weight with a hydroxyl value of from 90 to 300 mgKOH/g (Patent Document 2), but the hysteresis loss has been relatively large at a level of from 25 to 33%, such being disadvantageous from the viewpoint of the durability.

Further, with a seat of the above-mentioned deep foam type structure, the load-deflection characteristics are substantially influenced by the flexible foam itself, and it will be a seat having a relatively small difference in deflection on pressure side of from 500 N to 900 N, when the deflection under load is measured when it is pressed by the above-mentioned pressure plate from above. A seat having a small difference in deflection presents a bottom-hitting feeling and thus showed a tendency that the evaluation of the riding comfort was poor. Therefore, with a seat of deep foam type, in order to increase the difference in deflection, the thickness of the foam was increased. As a technique to increase the difference in deflection without increasing the thickness of the foam, it was proposed to use a fluorinated surfactant having a perfluoroalkyl group structure (Patent Document 3). However, a problem has been pointed out such that the effects tend to be different depending upon the structure of the fluorinated surfactant.

On the other hand, in a method for producing a polyure-

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polyisocyanate component, it has been proposed to add a certain specific bifunctional secondary amine in order to produce a foam having a reduced inclination of the curve on pressure side at a deflection of 75% as an index for evaluating the bottom-hitting feeling, which has no bottom-hitting feeling, and has all of the flexibility, the sinking degree and the vibration characteristics well balanced. However, the supported feeling was insufficient, and the durability particularly wet set was inadequate (Patent Document 4).

Furthermore, a technique to increase the density of a surface skin layer of the foam by using a known defoaming agent, has been known (Patent Document 5). This technique is particularly applied to an integral skin foam and is substantially different from the present invention.

Further, a technique to increase the distribution of the size of cell by using an amino-modified silicone, has been known (Patent Document 6). This technique relates to a polyurethane foam for a polishing sheet, provides a foam having a specific gravity of 0.87, and is different from the present invention.

Patent Document 1:	JP-A-7-330843
Patent Document 2:	JP-A-11-60676
Patent Document 3:	JP-A-11-322875
Patent Document 4:	JP-A-5-320304
Patent Document 5:	JP-A-6-87945
Patent Document 6:	JP-A-2004-75700

DISCLOSURE OF THE INVENTION

Object to be Accomplished by the Invention

The present invention provides a novel flexible polyurethane foam having good vibration characteristics and excellent in the occupant posture-stability performance.

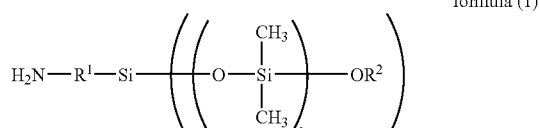
Means to Accomplish the Object

The present invention provides the following.

A flexible polyurethane foam obtained by reacting a high molecular weight polyoxyalkylene polyol or a polymer-dispersed polyol containing fine polymer particles in the high molecular weight polyoxyalkylene polyol, with a polyisocyanate compound, in the presence of a catalyst, a blowing agent and a foam stabilizer, characterized in that an amino-modified silicone (F) having a silicon atom and a nitrogen atom in its molecule is used in an amount of from 0.00001 to 1 part by mass per 100 parts by mass of all active hydrogen compounds.

The above flexible polyurethane foam, wherein the amino equivalent of the amino-modified silicone (F) is from 100 to 100,000 g/mol.

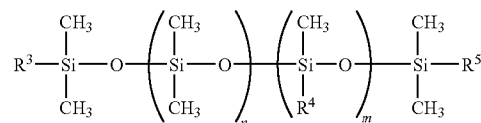
The above flexible polyurethane foam, wherein the amino-modified silicone (F) is a compound represented by the following formula (1) or (2):



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-continued

formula (2)



provided that in the formula (1), R¹ is a C₁₋₁₀ alkylene group, R² is a C₁₋₁₀ alkyl group, and k is an integer of from 1 to 2,000, and

in the formula (2), m is an integer of from 0 to 100; n is an integer of from 1 to 2,000; each of R³, R⁴ and R⁵ which may be the same or different, is a group selected from the group consisting of a C₁₋₁₀ alkyl group, a C₁₋₁₀ alkoxy group and a C₁₋₁₀ alkyl group having a nitrogen atom (provided that when m is 0, at least one of R³ and R⁵ is an alkyl group having a nitrogen atom, and when m is from 1 to 100, at least one of R³, R⁵ and a "m" number of R⁴'s is an alkyl group having a nitrogen atom).

The above flexible polyurethane foam, characterized in that with respect to a foam obtained by foaming into a thickness of 100 mm, the 25% hardness (ILD) X (N/314 cm²) measured in accordance with JIS K6400 (1997) and Y (mm) i.e. the value (difference in deflection on pressure side obtained by subtracting the deflection on 500 N pressure side from the deflection on 900 N pressure side obtained from a load-deflection curve as measured by means of a pressure plate (Tekken Plate) in accordance with JIS E7104 (2002), satisfy a relation formula represented by the following formula (3):

$$Y \geq -0.000370842X^2 + 0.225401X - 10.5013 \quad (3)$$

A seat for an automobile, which is made of the above flexible polyurethane foam.

A process for producing a flexible polyurethane foam, which comprises reacting a high molecular weight polyoxyalkylene polyol or a polymer-dispersed polyol containing fine polymer particles in the high molecular weight polyoxyalkylene polyol, with a polyisocyanate compound, in the presence of a catalyst, a blowing agent and a foam stabilizer, characterized in that an amino-modified silicone (F) having a silicon atom and a nitrogen atom in its molecule is used in an amount of from 0.00001 to 1 part by mass per 100 parts by mass of all active hydrogen compounds.

The above process for producing a flexible polyurethane foam, wherein the amino equivalent of the amino-modified silicone (F) is from 100 to 100,000 g/mol.

The above process for producing a flexible polyurethane foam, wherein the amino-modified silicone (F) is a compound represented by the above formula (1) or (2).

Effects of the Invention

The present invention provides a novel flexible polyurethane foam having good vibration characteristics and excellent in the occupant posture-stability performance.

BEST MODE FOR CARRYING OUT THE INVENTION

(High Molecular Weight Polyoxyalkylene Polyol)

The high molecular weight polyoxyalkylene polyol to be used for producing the flexible polyurethane foam of the present invention, is preferably one obtained by ring opening polymerization of a cyclic ether using, as an initiator, an active hydrogen compound having an average number of hydroxyl groups of from 2 to 6, in the presence of a polymerization catalyst. The molecular weight per hydroxyl group is preferably at least 500, more preferably from 1,000 to 5,000. The molecular weight per hydroxyl group is particularly preferably from 1,500 to 2,500.

The average number of hydroxyl groups in the high molecular weight polyoxyalkylene polyol is the average number of hydroxyl groups in the initiator used. Further, the high molecular weight polyoxyalkylene polyol is regarded as having a molecular weight calculated by the following formula based on the hydroxyl value measured in accordance with JIS K-1557 (1970):

$$\text{Molecular weight} = (56,100 \times \text{the average number of hydroxyl groups in the polyol}) / \text{hydroxyl value}$$

The polymerization catalyst may, for example, be an alkali metal compound or an alkali metal hydroxide such as a potassium compound, e.g. potassium hydroxide or potassium methoxide, a cesium compound, e.g. cesium metal, cesium hydroxide, cesium carbonate or cesium methoxide, a cationic polymerization catalyst such as boron trifluoride, a double metal cyanide complex, or a phosphazanium compound. Among these catalysts, a usual alkali catalyst such as potassium hydroxide, a cesium compound or a double metal cyanide complex is preferred, and in order to obtain a polymer having a high molecular weight, a double metal cyanide complex is particularly preferred.

The double metal cyanide complex catalyst may, for example be one disclosed in JP-B-46-27250. Specifically, it may, for example, be a complex containing zinc hexacyanocobaltate as the main component, preferably an ether and/or an alcohol complex thereof. As the ether, ethylene glycol dimethyl ether (glyme), diethylene glycol dimethyl ether (diglyme), ethylene glycol mono-tert-butyl ether (METB), ethylene glycol mono-tert-pentyl ether (METP), diethylene glycol mono-tert-butyl ether (DETB), tripropylene glycol monomethyl ether (TPME) or the like is preferred. As the alcohol, tert-butyl alcohol or the like is preferred.

The cyclic ether is preferably an alkylene oxide having at least 2 carbon atoms, and specifically, it may, for example, be ethylene oxide, propylene oxide, 1,2-butylene oxide, 2,3-butylene oxide or styrene oxide. Among them, a combination of ethylene oxide and at least one member selected from propylene oxide, 1,2-butylene oxide and 2,3-butylene oxide is particularly preferred.

Further, the high molecular weight polyoxyalkylene polyol preferably has oxyethylene groups, particularly preferably has oxyethylene groups at its terminals. Further, it may be a polyoxyalkylene polyol having oxyethylene groups in its inside. The polyoxyalkylene polyol having oxyethylene groups in its inside may be obtained, for example, by sequentially mixing an alkylene oxide having at least 3 carbon atoms and ethylene oxide, using an initiator, to carry out ring opening polymerization.

The polyoxyalkylene polyol having oxyethylene groups at

polymerization using an initiator, and then subjecting ethylene oxide to ring opening polymerization. Otherwise, it may be obtained by sequentially mixing an alkylene oxide having at least 3 carbon atoms and ethylene oxide to carry out ring opening polymerization using the above-described initiator, and then subjecting ethylene oxide to ring opening polymerization.

The lower limit of the content of the terminal oxyethylene groups in the polyoxyalkylene polyol is preferably 3 mass %, particularly preferably 5 mass %. The upper limit is preferably 25 mass %. If the content of the terminal oxyethylene groups is less than 3 mass %, collapse of the foam, etc. tends to occur. Further, if it exceeds 25 mass %, closed cells in the foam tend to increase, whereby the foam is likely to break at the time of crushing treatment, or shrinkage or the like is likely to occur after the crushing treatment.

The content of all oxyethylene groups is preferably at most 30 mass %.

The initiator may, for example, be ethylene glycol, diethylene glycol, dipropylene glycol, neopentyl glycol, 1,4-butanediol, 1,6-hexanediol, glycerol, trimethylolpropane, pentaerythritol, diglycerol, dextrose, sucrose, bisphenol A or the like, or a compound obtained by adding a small amount of an alkylene oxide to the above initiators. The initiators may be used preferably alone or in combination of two or more of them. If the average number of hydroxyl groups is less than 2, the durability and the riding comfort of the foam may decrease in some cases. On the other hand, if the average number of hydroxyl groups is more than 6, the flexible foam to be produced tends to be hard and tends to have impaired mechanical properties such as elongation.

In the present invention, it is preferred to use a polyoxyalkylene polyol (a) as at least a part of the high molecular weight polyoxyalkylene polyol. The polyoxyalkylene polyol (a) is a high molecular weight polyoxyalkylene polyol, which is a polyoxyalkylene polyol having an unsaturation value of at most 0.07 meq/g (hereinafter sometimes referred to as polyol (a)). Particularly, in the high molecular weight polyoxyalkylene polyol, the amount of the polyol (a) is preferably from 30 to 100 mass %, particularly preferably from 40 to 100 mass %. Further, in a case where the high molecular weight polyoxyalkylene polyol contains fine polymer particles as described hereinafter, the ratio of the high molecular weight polyoxyalkylene polyol and the polyol (a) is calculated on the basis of the mass of the polyols excluding the fine polymer particles.

The average molecular weight per hydroxyl group of the polyol (a) is preferably at least 500, more preferably from 1,000 to 5,000, particularly preferably from 1,500 to 2,500. If the average molecular weight per hydroxyl group is lower than 1,500, the durability or the riding comfort of the polyurethane foam may decrease in some cases. On the other hand, if it is higher than 2,500, the viscosity of the polyol tends to be high, whereby the operation efficiency tends to deteriorate.

Further, the polyol (a) preferably contains oxyethylene groups at its terminals, and the preferred range of the content is as described with respect to the high molecular weight polyoxyalkylene polyol. The polyol (a) is preferably a polyol having a content of oxypropylene groups of at least 70 mass %, particularly preferably at least 75 mass %.

The polyol (a) has an unsaturation value of at most 0.07 meq/g, particularly preferably at most 0.05 meq/g. If the unsaturation value of the polyol (a) is larger than 0.07 meq/g,

In the present invention, a polymer-dispersed polyol containing fine polymer particles in the high molecular weight polyoxyalkylene polyol is used. Such a polymer-dispersed polyol contains fine polymer particles in a polyoxyalkylene polyol matrix, and the fine polymer particles are preferably contained as dispersed. Such fine polymer particles are preferably fine particles of an addition polymerization type polymer or a condensation polymerization type polymer.

The addition polymerization type polymer may, for example, be a homopolymer or copolymer of a vinyl monomer such as acrylonitrile, styrene, a methacrylate or an acrylate. The condensation polymerization type polymer may, for example, be a polyester, a polyurea, a polyurethane or a melamine resin. By the presence of such fine polymer particles, the hydroxyl value of the entire polymer-dispersed polyol may usually be made lower than the hydroxyl value of the matrix polyol.

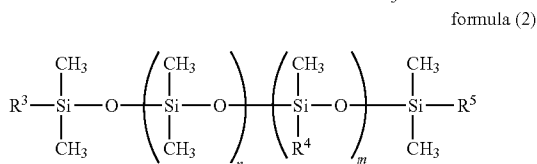
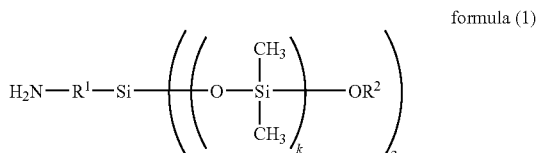
The content of the fine polymer particles contained in the high molecular weight polyoxyalkylene polyol is preferably at most 50 mass %. If the content of the fine polymer particles is higher than 50 mass %, the viscosity tends to be high, such being troublesome. The fine polymer particles are preferably contained in an amount of from 1 to 35 mass % in the high molecular weight polyoxyalkylene polyol.

(Amino-Modified Silicone (F))

In the present invention, an amino-modified silicone (F) having a silicone atom and a nitrogen atom in its molecule is used.

The amino equivalent of the amino-modified silicone (F) is preferably from 100 to 100,000 g/mol. The amino equivalent can be determined by a common neutralization titration method. For example, it can be calculated by the neutralization titration method as specified in JIS K7245 (2000). When the amino equivalent is from 100 to 100,000 g/mol, the above-described flexible foam characteristics are likely to be obtained, and the amino-modified silicone (F) will be stably present in the polyol to be used without being separated.

Further, the amino-modified silicone (F) is preferably a compound represented by the following formula (1) or (2):



In the formula (1), R¹ is a C₁₋₁₀ alkylene group, R² is a C₁₋₁₀ alkyl group (provided that three R²'s may be the same or different) and k is an integer of from 1 to 2,000 (provided that three k's may be the same or different).

In the formula (1), R¹ is an alkylene group which may or may not be branched. R¹ has from 1 to 10, preferably from 1 to 8, more preferably from 1 to 5 carbon atoms. R² is an alkyl group which may or may not be branched. R² has from 1 to 10,

of from 1 to 1,500, more preferably an integer of from 1 to 1,000. When k is within a range of from 1 to 2,000, the foam will be stably foam.

In the formula (2), m is an integer of from 0 to 100; n is an integer of from 1 to 2,000; each of R³, R⁴ and R⁵ which may be the same or different, is a group selected from the group consisting of a C₁₋₁₀ alkyl group, a C₁₋₁₀ alkoxy group and a C₁₋₁₀ alkyl group having a nitrogen atom (provided that when m is 0, at least one of R³ and R⁵ is an alkyl group having a nitrogen atom, and when m is from 1 to 100, at least one of R³, R⁵ and a "m" number of R⁴'s is an alkyl group having a nitrogen atom). When m is at least 2, a "m" number of R⁴'s may be the same or different. Further, the chain comprising a (m+n) number of siloxane units may be a block copolymer chain or a random copolymer chain.

In the formula (2), m is an integer of from 0 to 100, preferably an integer of from 0 to 50. Further, n is an integer of from 1 to 2,000, preferably an integer of from 1 to 1,500, more preferably an integer of from 1 to 1,000. When n is within a range of from 1 to 2,000, the foam will be stably foam.

Each of R³, R⁴ or R⁵ which may be the same or different, is a group selected from the group consisting of an alkyl group, an alkoxy group and an alkyl group having a nitrogen atom, as described hereinafter. When m is 0, at least one of R³ and R⁵ is an alkyl group having a nitrogen atom, and when m is from 1 to 100, at least one of R³, R⁵ and a "m" number of R⁴'s is an alkyl group having a nitrogen atom. This alkyl group may or may not be branched. The alkyl group has from 1 to 10, preferably from 1 to 8, more preferably from 1 to 5 carbon atoms. Further, the alkoxy group may or may not be branched. The alkoxy group has from 1 to 10, preferably from 1 to 8, more preferably from 1 to 5 carbon atoms.

The alkyl group having a nitrogen atom may or may not be branched. The alkyl group having a nitrogen atom has from 1 to 10, preferably from 1 to 8, more preferably from 1 to 5 carbon atoms. The alkyl group having a nitrogen atom is a group having a hydrogen atom of an alkyl group substituted by a primary, secondary or tertiary amine. In the case of a secondary or tertiary amine, such an alkyl group has an additional carbon atom (which may further be substituted by an amine), and such a carbon atom is included in the number of carbon atoms in the alkyl group. The number of the nitrogen atom in the alkyl group i.e. the number of the amine substituent is preferably from 1 to 3. Specifically, such an alkyl group having a nitrogen atom may, for example, be an aminomethyl group, an aminoethyl group, an aminopropyl group, an aminohexyl group, an aminooctyl group, a N,N-dimethylaminoethyl group, a N,N-diethylaminoethyl group, a N,N-dimethylaminopropyl group, a N-(aminoethyl)aminopropyl group (—C₃H₆—NH—C₂H₄—NH₂), or a N-(N'-(aminoethyl)aminoethyl)aminopropyl group (—C₃H₆—NH—C₂H₄—NH—C₂H₄—NH₂). Such an alkyl group having a nitrogen atom is preferably an alkyl group having active hydrogen bonded to a nitrogen atom, i.e. an alkyl group substituted by a primary or secondary amine, in that the amino-modified silicon (F) will not bleed out from the urethane foam to be finally produced.

The amount of the amino-modified silicone (F) to be used is from 0.00001 to 1 part by mass per 100 parts by mass of the total amount of all active hydrogen compounds. If it is smaller than 0.00001 part by mass, the above-described flexible foam characteristics will not be obtained. Further, if it is larger than 1 part by mass, the foam stability at the time of foaming may be impaired. All active hydrogen compounds are compounds having an active hydrogen atom reactive with an isocyanate

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