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(54) Title: MOLD RELEASE COMPOSITION AND PROCESS THEREWITH

(57) Abstract: A composition that can be used as mold release agent is disclosed. The composition comprises a solvent; a silicone resin, gum, or fluid; and optionally a catalyst, a co-solvent, or both in which the solvent is a volatile siloxane. Also provided is a process for applying a thin, continuous film coating on or onto a mold. The process comprises combining a solvent, a silicone resin or silicone gum or silicone fluid, and optionally a catalyst, a co-solvent, or both to produce a mold release composition; applying the mold release composition on or onto a mold, and optionally curing the composition in which the solvent is a volatile siloxane.



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## MOLD RELEASE COMPOSITION AND PROCESS THEREWITH

### FIELD OF THE INVENTION

This invention relates to a composition that can be used as mold release agent and to a process for applying a thin, continuous film coating of the release agent on a mold.

### BACKGROUND OF THE INVENTION

In commercial operations, mold release agent, generally a polymer or a combination of polymers, can be an emulsion or dispersion in a solvent. If dispersed in a solvent, the solvent is used as a vehicle to wet the surface of a shape-determining mold, onto which the release agent is applied. However, silicone resins, which are often used as release agents, often do not coat surfaces well when dispersed in a typical hydrocarbon solvent. The silicone resins, bead or puddle on the surface to which they have been applied, thus preventing a thin, continuous film from being achieved.

Therefore, there is an increasing need to develop a new release agent that can produce a thin, continuous film coating on the surface of a mold.

### SUMMARY OF THE INVENTION

A composition comprising a solvent; a silicone resin, gum, or fluid; and optionally a catalyst, a co-solvent, or both in which the solvent is a volatile siloxane.

Also provided is a process for applying a thin, continuous film coating on or onto a mold, which comprises combining a solvent, a silicone resin or silicone gum or silicone fluid, and optionally a catalyst, a co-solvent, or both to produce a mold release composition; applying the mold release composition on or onto a mold, and optionally curing the composition in which the solvent is a volatile siloxane.

### DETAILED DESCRIPTION OF THE INVENTION

According to the invention, the term "mold" means one or more shape-determining surfaces. Any volatile siloxane can be used as the solvent of the invention's composition. The term "volatile siloxane" refers to a rapidly evaporating siloxane under the temperature and pressure of use. Typically, it can

have an evaporation rate of  $> 0.01$  relative to n-butyl acetate which has an assigned value of 1.

A suitable solvent can have the formula of  $R(R_2SiO)_xSiR_3$  or  $(R_2SiO)_y$  where each R can be the same or different and is preferably, an alkyl group, 5 alkoxy group, a phenyl group, a phenoxy group, or combinations of two or more thereof; having 1 to about 10, preferably 1 to about 8 carbon atoms per group. R can also be a halogen. The most preferred R is a methyl group and can be substituted with a halogen, an amine, or other functional group. Subscript x can be a number from 1 to about 20, preferably 1 to 10. Subscript y can be a number 10 from 3 to about 20, preferably 3 to about 10. A preferred solvent has a molecular weight in the range between about 50 and about 1,000 and a boiling point less than about 300°C, preferably lower than 250°C, more preferably lower than 200°C, and most preferably lower than 150°C.

Examples of suitable methyl siloxanes include, but are not limited to, 15 hexamethyldisiloxane, hexamethylcyclotrisiloxane, 2,5-dichloro-1,1,3,3,5,5,- hexamethyltrisiloxane, 1,3-dimethyltetramethoxydisiloxane, 1,1,1,3,5,5,5,- heptamethyltrisiloxane, 3-(heptafluoropropyl)trimethylsiloxane, octamethyltrisiloxane, octamethyltetrasiloxane, octamethylcyclotetrasiloxane, decamethyltetrasiloxane, decamethylcyclopentasiloxane, 20 dodecamethylpentasiloxane, and dodecamethylcyclohexasiloxane, and combinations of two or more thereof.

Any silicone resin, gum, or fluid that is compatible with a volatile siloxane disclosed above can be used. It is generally a polyorganosiloxane. For example, methoxy-terminated polyalkylsiloxanes and hydroxy-terminated 25 polydimethylsiloxane can be used. Suitable silicone resin, gum, or fluid can be a resin, gum or fluid. Examples of suitable polyorganosiloxanes include polydimethylsiloxanes, polymethylhydrogensiloxanes, polysilsesquioxanes, polytrimethylsiloxanes, polydimethylcyclosiloxanes, and combinations of two or more thereof. Each silicone resin can also contain function groups such as halide, 30 amine, hydroxy, epoxy, carbinol, carboxylate, acetoxy, alkoxy, acrylate, and combinations of two or more thereof. The molecular weight can be in the range of from about 500 to about 1,000,000. A preferred silicone resin is

polyorganosiloxane having terminal silicon-bonded hydroxyl groups, which is well known and is commercially available.

The above-disclosed siloxanes and silicone resins, gums, or fluids are generally available commercially, for example, from Dow Corning Chemicals, Midland, Michigan, and General Electric, Fairfield, Connecticut.

Any organic solvent, preferably substantially free of water such as, for example, a hydrocarbon or halogenated hydrocarbon, that is inert towards other components of the composition, is compatible with the volatile siloxane and is volatile to evaporate rapidly when applied to the mold surface can be used as co-solvent. A co-solvent can also reduce the viscosity of the composition and promote the release of a polymer from a mold. Preferably, a co-solvent has a normal boiling point below about 300°C, preferably below 200°C, and most preferably below 150°C, depending on the temperature of the mold release composition to be applied to a mold. The lower the temperature the mold release composition to be allied, the lower the boiling point solvent is preferred and vice versa. Examples of suitable co-solvents include, but are not limited to, octane, decane, cyclohexane, toluene, xylene, methylene chloride, methylene dichloride, ethylene dichloride, carbon tetrachloride, chloroform, perchloroethylene, acetone, methylethyl ketone, ethyl acetate, tetrahydrofuran, dioxane, white spirit, mineral spirits, naphtha, and combinations of two or more thereof.

The mold release composition can also comprise additional silicone compound such as modified fumed silica, surfactants, fluoropolymers such as polytetrafluoroethylene, waxes, fatty acids such as stearic acid, fatty acid salts such as metal stearates, finely dispersed solids such as talc, emulsifiers, biocides, corrosion inhibitors.

Each component disclosed above can be present in the composition in an effective amount sufficient to effect the suitable release of a molded article. For example, based on the total weight of the composition, the solvent can be present in the composition in the range of from about 10 to about 99%; a silicone resin can be present in the composition in the range of from about 0.1 to about 90%. A co-solvent, if used, can be present in the composition in such range that the sum of solvent and co-solvent is about 10 to about 99%, provided that the solvent is

present at least about 10 %, preferably at least 20%. Other components, if present, can be in the range of from about 0.01 to about 10%.

Any catalyst that can catalyze or enhance the curing of a composition comprising a volatile siloxane, a silicone resin, and a solvent can be used herein.

5 A preferred catalyst is an organic titanium compound. Titanium tetrahydrocarbyloxides, also referred to as tetraalkyl titanates herein, are most preferred organic titanium compounds because they are readily available and effective. Examples of suitable titanium compounds include those expressed by the formula  $Ti(OR)_4$  where each R is individually selected from an alkyl, 10 cycloalkyl, alkaryl, hydrocarbyl radical containing from 1 to about 30, preferably 2 to about 18, and most preferably 2 to 12 carbon atoms per radical and each R can be the same or different. Titanium tetrahydrocarbyloxides in which the hydrocarboxyl group contains from 2 to about 12 carbon atoms per radical which is a linear or branched alkyl radical are most preferred because they are relatively 15 inexpensive, more readily available, and effective in curing the composition. Suitable titanium compounds include, but are not limited to, titanium tetraethoxide, titanium tetrapropoxide, titanium tetraisopropoxide, titanium tetra-n-butoxide, titanium tetrahexoxide, titanium tetra 2-ethylhexoxide, titanium tetraoctoxide, and combinations of two or more thereof.

20 Other suitable catalysts include a compound or element of VIII group of the periodic table of the elements such as platinum, palladium, iron, zinc, rhodium, and nickel as well as a tin or zirconium compound. Examples of other suitable catalysts include, but are not limited to, dibutyltin diacetate, dibutyl dilaurate, zinc acetate, zinc octoate, zirconium octoate, and combinations of two 25 or more thereof. For example, dibutyltin diacetate, can be used independently or in combination with a titanium compound.

These catalysts are believed commercially available. For examples, TYZOR<sup>®</sup> TPT and TYZOR<sup>®</sup> TBT (tetra isopropyl titanate and tetra n-butyl titanate, respectively) available from E. I. du Pont de Nemours and Company, 30 Wilmington, Delaware, U.S.A.

Each of the catalysts disclosed above can be used in the composition in the range of from about 0.01 to about 10 weight % relative to the silicone polymers.

The composition can be produced by any means known to one skilled in the art such as, for example, mixing each component disclosed above. It is preferred that the catalyst be introduced after the silicone resin, solvent, and the optional co-solvent are combined.

5           A process for applying a thin, continuous film coating on or onto a mold, which comprises (1) combining a solvent, a silicon resin or silicone gum or silicone fluid, and optionally a co-solvent, a catalyst, or both to produce a mold release composition, (2) applying the mold release composition on or onto a mold, and optionally (3) curing the release composition. The mold release composition  
10       can be the same as the composition disclosed above. Application of the mold release composition can be carried out by any means known to one skilled in the art such as, for example, spraying, brushing, wiping, dipping, and combinations of two or more thereof. Any surface of a shape-determining mold can be applied with the release composition. Curing can be carried out by any means known to  
15       one skilled in the art such as curing at ambient temperature such as from about 25°C to about 200°C under a pressure that accommodates the temperature range such as, for example, atmospheric pressure for about one second to about 2 hours. Generally, curing is carried out at the temperature and pressure at which the molding is being carried out.

20           For producing a molded article, a molding material or a material to be molded can be introduced into or onto the mold by any means known to one skilled in the art such as, for example, pumping, extruding, blending, or other suitable means known to one skilled in the art. Thereafter, a molded article is produced, which can be removed or released from the mold by any means known  
25       to one skilled in the art. Because the means for making a molded article is well known in the art, the description of which is omitted herein for the interest of brevity.

          The molding material can be any material that can be molded including, but not limited to, plastics, polymers, glass, ceramics, and metals. Examples of  
30       polymers include a thermosetting resin or a thermoplastic resin. Specific examples include acrylonitrile-butadiene-styrene, acrylonitrile-chlorinated polyethylene-styrene, acrylic-styrene-acrylonitrile, polyacetal homopolymers and

copolymers, acrylics, cellulose, fluoropolymers, polyamides, polyacrylates, polybutylene, polycarbonates, polyesters, polyethylenes, ethylene acid copolymers, ethylene-ethyl acrylates, ethylene-methyl acrylates, polymethyl acrylate, polymethyl methacrylate, polybutyl methacrylate, ethylene-vinyl acetates, ethylene vinyl alcohol copolymers, ionomers, polymethylpentene, polyphenylene oxides, polypropylene, ethylene-propylene copolymers, polypropylene impact copolymers, polypropylene random copolymers, polystyrenes, styrene-acrylonitrile, styrene-butadiene copolymers, styrene-ethylene/butylene-styrene, styrene-maleic anhydride copolymers, polyvinyl chloride, polyvinyl chloride, vinylidene chloride homopolymers and copolymers, styrenic block copolymers, polyolefin blends, elastomeric alloys, thermoplastic urethanes, polyethylene terephthalate, polybutylene terephthalate, thermoplastic copolyesters, polyethers, thermoplastic polyamides, polyether-polyamide block copolymers, allyl molding compounds, bis-maleimides, epoxy resins, phenolic resins, polyesters, ethylene-propylene diene terpolymers such as EPDM rubber, polyimides, ionomers, polyurethanes, segmented polyurea/urethanes, reaction injection molded polyurethanes, silicones, urea-melamine formaldehyde resins, polyacetals, polyesters, polyamides, ionomers, and combinations of two or more thereof. These polymers are well known to one skilled in the art.

## EXAMPLES

The following examples are illustrative of the invention and should not be construed as to unduly limit the scope of the invention.

Example 1. This example demonstrates that thinner continuous silicone resin coatings can be produced when using a volatile methylsiloxane than when using petroleum ether.

A release agent composition comprising 80% solvent (either petroleum ether or octamethylcyclotetrasiloxane (volatile methyl)), 20% silicone resins and fluids, and about 0.2% of catalyst (based on total weight of solvent and resin and fluid; TYZOR<sup>®</sup> TBT and dibutyltin diacetate) were sprayed onto pre-weighed 1 inch by 3 inch (2.54 cm by 7.62 cm) glass microscope slides heated to 65°C with quantity shown in the table below. After allowing the solvent sufficient time to evaporate in a hood, the release agent was cured for 2 minutes at 65°C. The slides

were then re-weighed and visually examined under a microscope to estimate the coverage of the slide's surface with silicone resin. The results are shown in Table 1.

Table 1

Coating Wt (mg)	Petroleum Ether	Methyl Siloxane
0.7		100% coverage
0.9	20% coverage	
1.9	40%	
2.8	65%	100%
3.1	70%	
3.5		100%
8.4	95%	
19.4	98%	
38.2	100%	

5

The coating was very irregular and bumpy on the slides coated using petroleum ether as the solvent. The coating using methyl siloxane as the solvent was very smooth. Replacing the petroleum ether with mineral spirits aggravated the beading of the silicone resins on the glass surface.

10 Example 2. This example serves to demonstrate how the use of various volatile methyl siloxane solvents promote the formation of smooth, continuous coatings of various silicone resins, fluids and functional fluids applied at elevated temperature.

15 Silicone resins, fluids and functional fluids were dispersed in various hydrocarbon solvents and volatile methyl siloxanes at a concentration of 5 wt % polymer solids and 95 wt % solvent. The silicone polymers and solvent were sprayed, using a Preval aerosol sprayer, onto pre-weighed, 1" x 3" (2.54 cm x 7.62 cm) glass microscope slides heated to 65C. After the solvent evaporated, the slides were re-weighed to determine coating weight and visually examined for the smoothness and coverage of the coating, as shown in Table 2.

20

Table 2

SP <sup>1</sup>	Solvent <sup>2</sup>	Coating			SP <sup>1</sup>	Solvent <sup>2</sup>	Coating		
		Wt (mg)	Description	Coverage			Wt (mg)	Description	Coverage
A	Toluene	1.8	spotty	incomplete	A	OMS	0.8	smooth	complete
B	PE	2.7	rough	incomplete	B	DMS	4.1	smooth	complete
B	MS	1.6	beaded	incomplete	B	---			
B	acetone	3.0	rough	incomplete	B	---			
C	MS	3.3	beaded	incomplete	C	OMS	2.6	smooth	complete
C					C	DMS	3.2	smooth	complete
D	S	1.2	beaded	incomplete	D	OMS	1.7	smooth	complete
D					D	OMTS	3.5	smooth	complete

<sup>1</sup> SP denotes silicone polymer; Polymer A is Hydroxy terminated, methyl silsesquioxane-dimethyl siloxane copolymer; polymer B is Methoxy terminated, methyl silsesquioxane-dimethyl siloxane copolymer; polymer C is 14,000 CSt (centiStoke) hydroxy terminated polydimethyl siloxane; and polymer D is 60,000 CSt polydimethyl siloxane.

<sup>2</sup> PE is petroleum ether; MS is mineral spirits; S is Stoddard solvent; OMS denotes octamethylcyclotetrasiloxane; DMS is decamethyltetrasiloxane; and OMTS is octamethyltrisiloxane.

Results show the silicone polymers dispersed in VMS fluids produced smooth, continuous films whereas the same silicone polymers dispersed in hydrocarbon solvents produced rough, incomplete films.

Example 3. This example demonstrates how the use of volatile methyl siloxane solvents promote the formation of smooth, continuous coatings of silicone polymers when applied at ambient temperature.

A sample of a silicone gum was dispersed in petroleum ether while a second sample of the same silicone gum was dispersed in VMS fluids at a concentration of 5 wt % polymer solids and 95 weight % solvent. The silicone polymer and solvent were sprayed, using a Preval aerosol sprayer, onto pre-weighed, 1" x 3" (2.54 cm x 7.62 cm) glass microscope slides at ambient temperature. After the solvent evaporated, the slides were re-weighed to determine coating weight and visually examined for the smoothness and coverage of the coating as shown in Table 3.

Table 3

SP <sup>1</sup>	Solvent <sup>2</sup>	Coating			SP <sup>1</sup>	Solvent <sup>2</sup>	Coating		
		Wt (mg)	Description	Coverage			Wt (mg)	Description	Coverage
D	PE	4.1	rough	incomplete	D	75/25 wt % OMS/OMTS	2.5	smooth	complete

<sup>1</sup> SP denotes silicone polymer; Polymer D is 60,000 CSt polydimethyl siloxane.

<sup>2</sup> PE is petroleum ether; OMS denotes hexamethyldisiloxane and OMTS is octamethyltrisiloxane.

5 Results show the silicone polymers dispersed in VMS fluids produced a smooth, continuous film whereas the same silicone polymers dispersed in a hydrocarbon solvent produced rough, incomplete film when applied at ambient temperature.

10 Example 4. This example serves to demonstrate how the use of a volatile methyl siloxane can promote the formation of a smooth, thin coating of a fully formulated silicone release agent.

A commercial release agent for plastics, adhesives and elastomeric products was purchased. The manufacturer describes the product as having a 50% active material content comprising trimethylated silica and tetra(trimethylsiloxy) silane and utilizing a hydrocarbon solvent mixture composed of Stoddard solvent, xylene and ethylbenzene. The manufacturer's brochure recommends diluting 15 parts of the release agent with 80 parts isopropanol and 10 parts toluene to improve surface wetting. Following the manufacturer's recommendation, the release coating was diluted to 5 wt % polymer solids utilizing an 10/80/10 wt % mixture of release agent, isopropanol and toluene. The release coating was then 20 applied using a Preval aerosol sprayer, onto pre-weighed, 1" x 3" (2.54 cm x 7.62 cm) glass microscope slides heated to 65C. After the solvent evaporated, the slides were re-weighed to determine coating weight and visually examined for the smoothness and coverage of the coating. As a comparison, the release agent was 25 diluted in octamethyltrisiloxane (OMTS), again to 5 weight % polymer solids and applied to a glass slide using the described manner. The results showed that manufacturer's recommended hydrocarbon solvent produced a spotty, incomplete coating whereas the use of the VMS solvent produced a smooth, continuous coating. The results are shown in Table 4 where IPA is isopropyl alcohol.

Table 4

Solvent	Coating			solvent	Coating		
	Wt (mg)	Description	Coverage		Wt (mg)	Description	Coverage
80/10 IPA/Toluene	3.0	spotty	incomplete	OMTS	3.3	smooth	complete

Example 5. This example serves to demonstrate how formation of a smooth, continuous coating improves the performance of a release agent.

- 5 A release coating utilizing a methyl silsesquioxane-dimethyl siloxane copolymer and a polydimethyl siloxane fluid were dispersed in a various solvents at a solids concentration of 5 weight %. The coating was applied using a Preval aerosol sprayer to a pre-weighed 9" x 12" x 3" (22.86 cm x 30.48 cm x 7.62 cm) aluminum box mold heated to 65C. After the solvent had evaporated, the mold
- 10 was cooled to room temperature and re-weighed. The mold was then reheated and filled with toluene diisocyanate based polyurethane flexible foam resin. The mold lid was secured and the expanding foam was allowed to cure for 6 minutes while being maintained at 65C in an oven. The foam was then removed from the mold by hand. If the foam was successfully removed from the mold without tearing,
- 15 the mold was refilled with polyurethane resin again without recoating the mold with release agent. This cycle was repeated until tearing of the foam was observed. The results are shown in Table 5 where the abbreviations are the same as those in Table 1.

Table 5

Solvent <sup>1</sup>	Weight of Cured Coating	Number of Releases Before Tearing of Foam
OMS	0.12 g	14
50/50 wt % mix OMTS/DMS	0.12 g	10
10/20/10/60 wt % mix, 244/1.0/1.5/75 OMS/OMTS/DMS/MS	0.14 g	12
mineral spirits	0.13 g	1

- 20 <sup>1</sup> See previous tables for solvent abbreviations.

The results show that a release coating dispersed in a VMS fluid or a combination of VMS and hydrocarbon solvent provide significantly improved performance compared to using a hydrocarbon solvent alone.

CLAIMS

1. A composition comprising a solvent; a silicone resin, gum, or fluid; and optionally a catalyst, a co-solvent, or both wherein said solvent is a volatile siloxane.
- 5 2. A composition according to claim 1 comprising said catalyst, said co-solvent, or both.
3. A composition according to claim 1 or 2 wherein said solvent has the formula of  $R(R_2SiO)_xSiR_3$  or  $(R_2SiO)_y$ ; each R is independently an alkyl group, alkoxy group, a phenyl group, a phenoxy group, or combinations of two or more  
10 thereof having 1 to about 10 carbon atoms per group.
4. A composition according to claim 1, 2, or 3 wherein said solvent is hexamethyldisiloxane, hexamethylcyclotrisiloxane, 2,5-dichloro-1,1,3,3,5,5,- hexamethyltrisiloxane, 1,3-dimethyltetramethoxydisiloxane, 1,1,1,3,5,5,5,- heptamethyltrisiloxane, 3-(heptafluoropropyl)trimethylsiloxane,  
15 octamethyltrisiloxane, octamethylcyclotetrasiloxane, decamethyltetrasiloxane, decamethylcyclopentasiloxane, dodecamethylpentasiloxane, dodecamethylcyclohexasiloxane, or combinations of two or more thereof; and is preferably octamethyltrisiloxane, octamethylcyclotetrasiloxane, decamethyltetrasiloxane, or combinations of two or more thereof.
- 20 5. A composition according to claim 1, 2, 3, or 4 wherein said catalyst is a titanium compound, dibutyltin diacetate, or combinations thereof.
6. A composition according to claim 1, 2, 3, 4, or 5 wherein said silicone resin, gum, or fluid is a polyorganosiloxane; preferably a methoxy-terminated polyalkylsiloxane, a hydroxy-terminated polydimethylsiloxane, or combinations of  
25 two or more thereof; and more preferably a polydimethylsiloxane, a polymethylhydrogensiloxane, a polysilsesquioxane, a polytrimethylsiloxane, a polydimethylcyclsiloxane, or combinations of two or more thereof.
7. A composition according to claim 6 further comprising modified fumed silica, a surfactant, a fluoropolymer, a wax, a fatty acid, a fatty acid salt, a finely  
30 dispersed solid, an emulsifier, a biocide, a corrosion inhibitor, or combinations of two or more thereof.

8. A process comprising combining a solvent, a silicone resin or silicone gum or silicone fluid, and optionally a catalyst, a co-solvent, or both to produce a mold release composition; applying said composition on or onto a mold; and optionally curing said composition wherein said solvent, said silicone resin or silicone gum  
5 or silicone fluid, said catalyst, and said co-solvent are the same as characterized in claim 1, 2, 3, 4, 5, 6, or 7.
9. A process according to claim 8 further comprising curing said composition.
10. A process according to claim 8 or 9 further comprising introducing a  
10 molding material into or onto said mold, converting said material into a molded article, and recovering said article.

# INTERNATIONAL SEARCH REPORT

International Application No  
PCT/US2004/015266

## A. CLASSIFICATION OF SUBJECT MATTER

IPC 7 B29C33/60 B29C33/64 C08L83/04

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)

IPC 7 B29C C08L

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

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal

## C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category *	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
X	US 3 684 756 A (BROOKS HOWARD LARRY) 15 August 1972 (1972-08-15) the whole document -----	1-10
X	US 6 403 163 B1 (FISHER PAUL DAVID ET AL) 11 June 2002 (2002-06-11) the whole document -----	1-7
X	US 2002/193511 A1 (LIN ZUCHEN) 19 December 2002 (2002-12-19) the whole document -----	1-7

☐ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

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# INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No  
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