SILICEOUS FILM WITH SMALLER FLAT BAND SHIFT AND METHOD FOR PRODUCING SAME

TECHNICAL FIELD

[0001] The present invention provides a process for producing a siliceous film. More particularly, the present invention relates to a process for producing a siliceous film having a smaller flat band shift value and excellent insulating properties.

BACKGROUND ART

[0002] In electronic devices such as semiconductor devices, semiconductor elements, for example, transistors, resistors and the like, are generally provided on a substrate. These elements should be electrically insulated from each other. Accordingly, a region for isolating the elements should be provided between these elements. This region is referred to as "isolation region." A siliceous film is generally used as a material for the formation of the isolation region.

[0003] Various processes for producing a siliceous film have been studied. Among them, a technique for producing a siliceous film by firing a thin film of polysilazane has been studied (for example, patent document 1 or 2).

[0004] The siliceous films produced by these processes are disadvantageous in that, due to the stay of nitrogen, carbon, and hydrogen considered attributable to a polysilazane as a starting material and a solvent, the flat band shift in a CV curve is higher than that in a siliceous film formed by high-density plasma CVD. Accordingly, the conventional siliceous films have room for improvement in insulating properties. In this case, particularly when the heating temperature should be low due to limitation on heat resistance and oxidation resistance of the substrate used, there is a tendency toward an increase in the amount of nitrogen, carbon, or hydrogen staying in the siliceous film.

[0005] On the other hand, techniques where an aluminum

compound is added to a polysilazane compound, or aluminum is incorporated in the structure of a polysilazane compound, are also known (patent documents 3 and 4). In these patent documents, however, any study has not been made on the flat band shift of the siliceous film formed by the techniques.

Patent document 1: Patent No. 3511024

Patent document 2: Patent No. 3178412

Patent document 3: Japanese Patent Laid-Open No. 5963/1996

Patent document 4: Japanese Patent Laid-Open No. 105185/1999

Patent document 5: Japanese Patent Laid-Open No. 63833/1992

Patent document 6: Japanese Patent Laid-Open No. 31333/1997

Patent document 7: Japanese Patent Laid-Open No. 176511/1996

Patent document 8: Japanese Patent Laid-Open No. 176512/1996

Patent document 9: Japanese Patent Laid-Open No. 345826/1993

DISCLOSURE OF INVENTION

PROBLEMS TO BE SOLVED BY THE INVENTION

[0006] An object of the present invention is to solve the above problems of the prior art and to provide a process for producing a siliceous film having a smaller flat band shift value, that is, having excellent electrical characteristics such as insulating properties.

MEANS FOR SOLVING THE PROBLEMS

[0007] According to the present invention, there is provided a coating composition comprising: at least one polysilazane compound selected from the group consisting of perhydropolysilazanes and modified perhydropolysilazanes,

having a number average molecular weight of 100 to 50,000; an aluminum compound; and a solvent, said coating composition having an aluminum content of not less than 10 ppb and not more than 100 ppm in terms of the molar ratio of the aluminum atom to the silicon atom contained in the polysilazane compound.

[0008] Further, according to the present invention, there is provided a process for producing a siliceous film comprising coating a coating liquid onto a substrate and firing the coated substrate in an atmosphere containing steam, oxygen, or a mixed gas composed of steam and oxygen, said coating liquid comprising: at least one polysilazane compound selected from the group consisting of perhydropolysilazanes and modified perhydropolysilazanes, having a number average molecular weight of 100 to 50,000; an aluminum compound; and a solvent, said coating composition having an aluminum content of not less than 10 ppb and not more than 100 ppm in terms of the molar ratio of the aluminum atom to the silicon atom contained in the polysilazane compound.

[0009] Furthermore, according to the present invention, there is provided a siliceous film produced by coating a coating liquid comprising a polysilazane compound and an aluminum compound onto a substrate and firing the coated substrate under an oxidizing atmosphere, said siliceous film having an aluminum content of not less than 10 ppb and not more than 100 ppm in terms of the molar ratio of the aluminum atom to the silicon atom.

EFFECT OF THE INVENTION

[0010] According to the present invention, a siliceous film having a smaller flat band shift value can be produced, whereby semiconductor elements having excellent electric characteristics, for example, element isolation films and gate insulation films having excellent insulating properties, can be formed.

BEST MODE FOR CARRYING OUT THE INVENTION

[0011] Coating composition

In the coating composition according to the present invention, the at least one polysilazane compound selected from the group consisting of perhydropolysilazanes or modified perhydropolysilazanes may be any proper polysilazane compound according to need. In the present invention, the perhydropolysilazane is represented by general formula (I):

$$H\text{-}(SiH_2\text{-}NH)_n\text{-}H \qquad (I)$$

wherein n is a number indicating the degree of polymerization.

[0012] In the coating composition according to the present invention, a modified perhydropolysilazane prepared by modifying the perhydropolysilazane represented by general formula (I) with a silazane compound, an alcohol, or an amine may be used. In the present invention, the perhydropolysilazane is preferably used from the viewpoint of reducing the flat band shift of the siliceous film formed using the coating composition. This is because, when a polysilazane compound containing a relatively high organic group content as in the case where an organic group other than hydrogen is contained in the main chain, atoms such as the organic group-derived carbon remains in the siliceous film and, consequently, the effect of the present invention is lowered.

[0013] On the other hand, from the viewpoints of coatability in the formation of the film, storage stability and the like, in some cases, a modified perhydropolysilazane prepared by partially modifying a part of the perhydropolysilazane is preferably used. Compounds usable for the modification of the perhydropolysilazane include compounds such as hexamethyldisilazane, methyl alcohol, ethyl alcohol, and ethylenediamine. The modified perhydropolysilazane is provided by substituting a part of hydrogen atoms, particularly terminal hydrogen, in the perhydropolysilazane using these compound. Such modified perhydropolysilazanes are described in patent documents 5 to 9.

[0014] The weight average molecular weight of the

perhydropolysilazane or the modified perhydropolysilazane used in the coating composition according to the present invention is 100 to 50,000, preferably 500 to 20,000, from the viewpoint of the coatability in coating particularly by spin coating.

[0015] In the coating composition according to the present invention, the polysilazane compound is dissolved in a solvent. The solvent used in this case is preferably an active hydrogen-free inert organic solvent. Solvents usable herein include aromatic hydrocarbon solvents such as benzene, toluene, xylene, ethylbenzene, diethylbenzene, trimethylbenzene, and triethylbenzene; alicyclic hydrocarbon solvents such as cyclohexane, cyclohexene, decahydronaphthalene, ethylcyclohexane, methylcyclohexane, p-menthane, dipentene (limonene), and pinene; saturated hydrocarbon compounds such as n-pentane, i-pentane, n-hexane, i-hexane, n-heptane, i-heptane, n-octane, i-octane, n-nonane, i-nonane, n-decane, and i-decane; ether solvents such as dipropyl ether and dibutyl ether; ketone solvents such as methyl isobutyl ketone; and ester solvents such as propylene glycol monomethyl ether acetate. Among them, solvents capable of satisfactorily dissolving aluminum compounds which will be described later are particularly preferred.

[0016] The aluminum compound used in the present invention is not particularly limited. However, aluminum compounds dissolved in the polysilazane solution are preferred. Accordingly, the aluminum compound can be properly selected depending upon the solvent used in the coating composition.

[0017] The aluminum compound usable in the present invention can be represented, for example, by the following general formula:

$$Al(Z^1)(Z^2)(Z^3) \qquad (A)$$

wherein Z^1, Z^2 and Z^3 each independently represent a group selected from the group consisting of hydrogen, a hydroxyl group, a halogen atom, an alkyl group, a cycloalkyl group, an aryl group, an alkenyl group, a cycloalkenyl group, an alkoxy group, and an acetyl acetonate group,

provided that two or three of Z^1, Z^2 and Z^3 may form a cyclic structure and Z^1, Z^2 and Z^3 may represent a silicon-containing organic group.

[0018] In the present invention, among these aluminum compounds, those represented by general formulae (A-1) to (A-3) are preferred:

$$Al(OR^1)(OR^2)(OR^3) \quad (A\text{-}1)$$

$$Al(R^1)(R^2)(R^3) \quad (A\text{-}2)$$

$$Al(X^1)(X^2)(X^3) \quad (A\text{-}3)$$

wherein R^1, R^2 and R^3 each independently represent hydrogen, an alkyl group having 1 to 20 carbon atoms, a cycloalkyl group, or an aryl group; and X^1, X^2 and X^3 each independently represent fluorine, chlorine, or iodine.

[0019] More specifically, aluminum compounds usable in the present invention include trimethoxyaluminum, triethoxyaluminum, triisopropoxyaluminum, tri-n-propoxyaluminum, trimethylaluminum, triethylaluminum, triisopropylaluminum, tri-n-propylaluminum, aluminum fluoride, aluminum chloride, aluminum iodide, aluminum triacetylacetonate, and aluminum triethylacetoacetate. Further, silicon-containing aluminum compounds, for example, di-s-butoxyaluminoxy-trietoxysilane, may also be used.

[0020] The amount of these aluminum compounds used is not particularly limited so far as the effect of the present invention is not sacrificed. In order to significantly develop the effect of the present invention, the molar ratio of the aluminum atom to the silicon atom contained in the polysilazane compound is preferably not less than 10 ppb, more preferably not less than 100 ppb. Further, from the viewpoint of maintaining good withstanding voltage properties, the molar ratio of the aluminum atom to the silicon atom contained in the polysilazane compound is preferably not more than 100 ppm, more preferably not more than 10 ppm. The above addition amount of the aluminum compound is much smaller than that described in patent document 3 or 4. It is surprising that the properties of the formed siliceous film can be dramatically

improved by using the very small amount of the aluminum compound.

[0021] In the coating composition according to the present invention, the aluminum compound may be added to the coating composition by any desired method. Specifically, the aluminum compound may be added, for example, by a method in which a solid aluminum compound is added to and dissolved in the coating composition, or a method in which an aluminum compound is dissolved in a solvent to prepare a solution which is then mixed into the coating composition. When the aluminum compound is dissolved in the solvent, the solvent may be one usable in the dissolution of the polysilazane compound. In this case, the solvent used in the dissolution of the polysilazane compound may be different from the solvent used in the preparation of the aluminum compound solution. The temperature and pressure at which the aluminum compound is added is not particularly limited. In general, however, the aluminum compound is generally added at 0 to 200°C and at the atmospheric pressure to 10 kg/cm^2G.

[0022] Production process of siliceous film

The production process of a siliceous film according to the present invention comprises coating the above coating composition onto a substrate and firing the coated substrate to form a siliceous film.

[0023] The surface material of the substrate used is not particularly limited, and examples thereof include bare silicon, and silicon wafers with a thermally grown oxide film or a silicon nitride film optionally formed thereon. If necessary, a structure such as a trench isolation groove may be formed on the substrate.

[0024] Methods usable for coating the coating composition onto the surface of the substrate include conventional methods, for example, spin coating, dip coating, spray coating, and transfer methods.

[0025] If necessary, excess organic solvent is removed (by drying) from the coating film formed on the surface of the

substrate. The coating film is then fired in an atmosphere containing steam, oxygen or a mixed gas composed of steam and oxygen, that is, in an oxidizing atmosphere. In this case, an inert gas such as nitrogen or helium may be present as a mixture in such an amount range that is not detrimental to the effect of the present invention.

[0026] In the process according to the present invention, preferably, the firing is carried out under relatively strong oxidizing conditions from the viewpoint of minimizing impurity elements, for example, carbon, hydrogen, and nitrogen, remaining in the siliceous film. In the present invention, firing under an oxygen-containing atmosphere is particularly preferred. The oxygen content is preferably not less than 1%, more preferably not less than 10% by volume.

[0027] Further, in the process according to the present invention, when the firing is carried out in a steam-containing atmosphere, the steam content is preferably not less than 0.1% by volume, more preferably not less than 1% by volume. In the present invention, preferably, firing under a mixed gas atmosphere containing oxygen and steam is particularly preferred.

[0028] The firing temperature should be such that the polysilazane compound can be converted to the siliceous film. The firing temperature is generally 100 to 1,200°C, preferably 300 to 1,000°C. There is a tendency that, as compared with the conventional production process, the effect of improving the flat band shift becomes more significant with decreasing the firing temperature. Further, there is a tendency that a siliceous film having a smaller flat band shift and better electric properties is provided as the firing temperature increases.

[0029] The firing time may be properly selected depending upon the firing temperature but is generally 5 min to 10 hr. The firing time is preferably one hr or shorter from the viewpoint of the efficiency of the production.

[0030] The siliceous film produced by the production process according to the present invention has a smaller flat

band shift value. It is considered that this property is derived from the homogeneous dispersion of the aluminum compound-derived aluminum oxide in the formed siliceous film. By virtue of this property, the siliceous film according to the present invention has excellent electrical characteristics, for example, excellent insulating properties.

[0031] Siliceous film, and substrate with siliceous film

The siliceous film and substrate with a siliceous film according to the present invention may be produced, for example, by the above production process. In general, in the production of the siliceous film, the ratio between the aluminum atom in the aluminum compound as the starting material and the silicon atom in the polysilazane compound as the starting material remains unchanged in the finally obtained siliceous film. Accordingly, the siliceous film according to the present invention can be produced by using the aluminum compound in the formation of the siliceous film so that the ratio of the aluminum atom to the silicon atom is not less than 10 ppb and not more than 100 ppm. Alternatively, in the above method, for example, a siliceous film having a low aluminum content is first formed followed by introduction of an aluminum atom into the film, by another means, for example, ion implantation, to bring the final aluminum content to not less than 10 ppb and not more than 100 ppm.

[0032] The siliceous film and substrate with a siliceous film have excellent electrical characteristics and thus are useful for use in various semiconductor elements, for example, element isolation films, interlayer insulating films such as premetal dielectric films and intermetallic dielectric films, and gate insulating films for liquid crystal display devices and the like.

[0033] Reference Synthesis Example 1 Synthesis of perhydropolysilazane

A reactor comprising a four-necked flask having an internal volume of 2 liters and equipped with a gas blow pipe, a mechanical stirrer, and a Dewar condenser was provided. The

air in the reactor was replaced by dry nitrogen, and 1500 ml of dry pyridine was introduced into the four-necked flask, followed by ice cooling. Next, 100 g of dichlorosilane was added to pyridine in the flask. This resulted in the production of an adduct ($SiH_2Cl_2 \cdot 2C_5H_5N$) as a white solid. This reaction mixture was further ice-cooled, and 70 g of ammonia was blown into the reaction mixture with stirring. Subsequently, dry nitrogen was blown into the reaction mixture for 30 min to remove excess ammonia.

[0034] The reaction mixture was then filtered in a dry nitrogen atmosphere through a Buchner funnel under reduced pressure to give 1200 ml of a filtrate. Pyridine was removed from this filtrate by an evaporator to give 40 g of perhydropolysilazane.

[0035] The molecular weight of the perhydropolysilazane was measured by gel permeation chromatography using $CDCl_3$ as a developing solution and was found to be 800 in terms of number average molecular weight as determined using polystyrene as a standard. Further, an infrared absorption spectrum of the perhydropolysilazane was measured. As a result, absorption attributable to N-H bond was observed at wavenumbers (cm^{-1}) 3350 and 1180; absorption attributable to Si-H bond was observed at wavenumber (cm^{-1}) 2170; and absorption attributable to Si-N-Si bond was observed at wavenumber (cm^{-1}) 1020 to 820.

[0036] Comparative Example 1

The perhydropolysilazane (20 g) prepared in Reference Synthesis Example 1 was dissolved in 80 g of dibutyl ether, which had been dehydrated with a molecular sieve, and the solution was filtered through a PTFE syringe filter with a filtration accuracy of 0.1 μm manufactured by ADVANTEC.

[0037] A substrate comprising a 10 nm-thick thermally grown oxide film formed on a p-type silicon wafer having a diameter of 8 inches was provided. The polysilazane solution was spin coated on the oxide film at a main spin speed of 1000 rpm/20 sec. After coating, the substrate was dried by heating

on a hot plate at 150°C for 3 min. Subsequently, the substrate was heated to form a 500 nm-thick siliceous film. Heating conditions were as follows.

Heating condition 1: Heated at 400°C for 15 min under an atmosphere with steam concentration 80% by volume and oxygen concentration 20% by volume.

Heating condition 2: Heated at 800°C for 15 min under an atmosphere with steam concentration 80% by volume and oxygen concentration 20% by volume.

Heating condition 3: Heated at 400°C for 15 min under an atmosphere with steam concentration 80% by volume and oxygen concentration 20% by volume, followed by heating at 800°C for 30 min in a dry nitrogen atmosphere.

Heating condition 4: Heated at 400°C for 30 min in a nitrogen atmosphere.

[0038] For each of the substrates thus obtained, flat band shift was measured. At the outset, each of the substrates was confirmed to have a film thickness of 500 nm by thickness measurement with an ellipsometer (Model-M44, manufactured by J.A. Woolam). Further, a CV curve and a flat band shift value based on the curve were measured with an automatic mercury probe CV/IV measuring device (Model SSM 495, manufactured by SSM Japan). The voltage applied in the CV measurement was in the range of -100 to +100 V.

[0039] Comparative Example 2

For a high-density plasma CVD film, the flat band value was measured in the same manner as in Comparative Example 1.

[0040] Example 1

Tri(isopropoxy)aluminum (1 g) was mixed into 100 g of dehydrated dibutyl ether to give a tri(isopropoxy)aluminum solution.

[0041] The perhydropolysilazane (20 g) prepared in Reference Synthesis Example 1 was dissolved in 80 g of dibutyl ether, which had been dehydrated with a molecular sieve. The aluminum compound solution (27 mg) containing

tri(isopropoxy)aluminum was added to this solution, and the mixed solution was filtered through a PTFE syringe filter with a filtration accuracy of 0.1 μm manufactured by ADVANTEC.

[0042] The flat band value was measured in the same manner as in Comparative Example 1, except that this mixed solution was used.

[0043] Example 2

The flat band value was measured in the same manner as in Example 1, except that tri(acetyl acetonate)aluminum was used instead of tri(isopropoxy)aluminum and the amount of the aluminum compound solution added was changed to 43 mg.

[0044] Example 3

The flat band value was measured in the same manner as in Example 1, except that (ethyl acetoacetate)di(isopropoxy)aluminum was used instead of tri(isopropoxy)aluminum and the amount of the aluminum compound solution added was changed to 34 mg.

[0045] Example 4

The flat band value was measured in the same manner as in Example 1, except that tri(ethyl acetoacetate)aluminum was used instead of tri(isopropoxy)aluminum and the amount of the aluminum compound solution added was changed to 56 mg.

[0046] Example 5

The flat band value was measured in the same manner as in Example 1, except that aluminum chloride was used instead of tri(isopropoxy)aluminum and the amount of the aluminum compound solution added was changed to 18 mg.

[0047] Example 6

The flat band value was measured in the same manner as in Example 1, except that di-s-butoxyaluminoxy-triethoxysilane was used instead of tri(isopropoxy)aluminum and the amount of the aluminum compound solution added was changed to 47 mg.

[0048] Comparative Example 3

The flat band value was measured in the same manner as in Example 1, except that the amount of tri(isopropoxy)aluminum to be dissolved in dibutyl ether was changed to 0.001 g.

[0049] Comparative Example 4

The flat band value was measured in the same manner as in Example 1, except that the amount of the tri(isopropoxy)aluminum solution to be added to the polysilazane solution was changed to 2.7 g.

[0050] Example 7

Methyl alcohol (10 g) was mixed and thoroughly dissolved into 90 g of dehydrated dibutyl ether. This solution (10 g) was sampled, and 100 g of the perhydropolysilazane solution prepared in Comparative Example 1 was gradually added thereto with stirring. This mixed solution was heated at 80°C for one hr in a nitrogen atmosphere and was allowed to cool. This reaction resulted in the formation of an ethyl alcohol-modified perhydropolysilazane solution.

[0051] The flat band value was measured in the same manner as in Example 1, except that this modified perhydropolysilazane solution was used.

[0052] Results of evaluation

The results obtained for Comparative Examples 1 and 2 and Examples 1 to 6 were as follows.

[Table 1]

	Al con-tent*	Heating condition 1	Heating condition 2	Heating condition 3	Heating condition 4 (comparative)
Comparative Example 1	0	-18.5	-13.0	-22.0	-45.2
Comparative Example 2	0	-7.0 to -15.0			
Example 1	3 ppm	-11.2	-8.5	-9.5	-38.2
Example 2	3 ppm	-10.0	-6.2	-11.0	-44.0
Example 3	3 ppm	-9.3	-9.0	-10.1	-40.5
Example 4	3 ppm	-11.0	-7.8	-10.2	-46.2
Example 5	3 ppm	-9.9	-7.2	-8.9	-39.5

Example 6	3 ppm	-10.4	-8.8	-10.4	-50.2
Comparative Example 3	3 ppb	-19.0	-13.4	-22.4	
Comparative Example 4	300 ppm	Immeas-urable	Immeas-urable	Immeas-urable	
Example 7	3 ppm	-11.4	-6.2	-8.0	

Unit: V

Al content: Proportion of the number of aluminum atoms to the number of silicon atoms

[0053] In Examples 1 and 4, the content of aluminum in siliceous films produced under respective firing conditions was measured with a secondary ion mass spectrometer (Model 6650, manufactured by Physical Electronics) and a tracer-type surface shape measuring device (Model DEKTAK8000, manufactured by SLDAN). As a result, the aluminum content was substantially identical to the aluminum content of the coating liquid, indicating that the content of aluminum in the coating composition is identical to the content of aluminum in the siliceous film produced using the coating composition. Further, it was found that the aluminum content was constant in the thickness-wise direction of the siliceous film, and the aluminum atom was evenly distributed in the siliceous film.

INDUSTRIAL APPLICABILITY

[0054] This invention can provide a production process of a siliceous film having a smaller flat band shift value, that is, a siliceous film having excellent insulating properties. The siliceous film produced by this process is utilizable, for example, in semiconductor elements, for example, element isolation films, interlayer insulating films such as premetal dielectric films and intermetallic dielectric films, and gate insulating films for liquid crystal display devices and the like.

CLAIMS

[1] A coating composition comprising: at least one polysilazane compound selected from the group consisting of perhydropolysilazanes and modified perhydropolysilazanes, having a number average molecular weight of 100 to 50,000; an aluminum compound; and a solvent, said coating composition having an aluminum content of not less than 10 ppb and not more than 10 ppm in terms of the molar ratio of the aluminum atom to the silicon atom contained in the polysilazane compound.

[2] The coating composition according to claim 1, wherein said modified perhydropolysilazane is a modification product of a perhydropolysilazane with a silazane compound, an alcohol, or an amine.

[3] The coating composition according to claim 1 or 2, wherein said aluminum compound is represented by general formula (A):

$$Al(Z^1)(Z^2)(Z^3) \qquad (A)$$

wherein Z^1, Z^2 and Z^3 each independently represent a group selected from the group consisting of hydrogen, a hydroxyl group, a halogen atom, an alkyl group, a cycloalkyl group, an aryl group, an alkenyl group, a cycloalkenyl group, an alkoxy group, and an acetyl acetonate group, provided that two or three of Z^1, Z^2 and Z^3 may form a cyclic structure and Z^1, Z^2 and Z^3 may represent a silicon-containing organic group.

[4] The coating composition according to any one of claims 1 to 3, wherein said coating liquid has an aluminum content of not less than 100 ppb and not more than 10 ppm in terms of the molar ratio of the aluminum atom to the silicon atom contained in the polysilazane compound.

[5] A process for producing a siliceous film comprising coating a coating liquid onto a substrate and firing the coated substrate in an atmosphere containing steam, oxygen, or a mixed gas composed of steam and oxygen, said coating liquid comprising: at least one polysilazane compound selected from the group consisting of perhydropolysilazanes and

modified perhydropolysilazanes, having a number average molecular weight of 100 to 50,000; an aluminum compound; and a solvent, said coating composition having an aluminum content of not less than 10 ppb and not more than 10 ppm in terms of the molar ratio of the aluminum atom to the silicon atom contained in the polysilazane compound.

[6] The process for producing a siliceous film according to claim 5, wherein said firing is carried out under an atmosphere containing oxygen or a mixed gas composed of oxygen and steam.

[7] A siliceous film produced by a process according to claim 5 or 6.

[8] A siliceous film produced by coating a coating liquid comprising a polysilazane compound and an aluminum compound onto a substrate and firing the coated substrate under an oxidizing atmosphere, said siliceous film having an aluminum content of not less than 10 ppb and not more than 10 ppm in terms of the molar ratio of the aluminum atom to the silicon atom.

[9] A substrate with a siliceous film, comprising a substrate and a siliceous film, produced by a process according to claim 5 or 6, provided on at least one surface of said substrate.

[10] The substrate with a siliceous film according to claim 9, wherein said siliceous film is an element isolation film, an interlayer insulating film, or a gate insulating film.