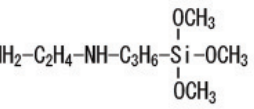
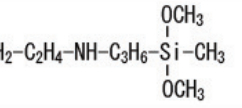
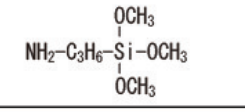
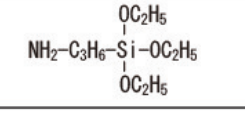
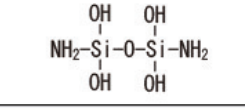
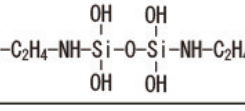
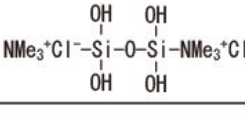
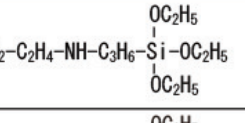
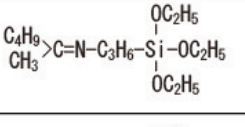
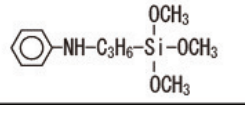
TABLE 1

Mark	Primary particle size (nm)	Secondary particle size (nm)	Degree of association	True density (g/cm^3)	Specific surface area (m^2/g)	Silanol group density (OH/nm^2)	Surface modification
A	34.6	70	2.0	2.2	78.8	5.6	not performed
B	24.4	48	2.0		111.8	4.2	not performed
C	58.4	99	1.7		46.7	3.5	not performed
D	35.1	54	1.5		77.7	5.6	not performed
E	29.7	84	2.8		91.8	4.2	not performed
F	16.5	26	1.6		165.3	2.6	not performed
G	30.6	62	2.0		89.1	1.8	not performed
H	21.9	38	1.7		124.5	1.6	not performed
I	35.6	67	1.9		76.6	–	modified with cation (amino group)
J	32.9	68	2.1		82.9	–	modified with anion (sulfo group)

[0056] [Table 2]

TABLE 2

Mark	Chemical name	Structural formula	Molecular weight	Minimum area of coating (m^2/g)
SA	N-(2-aminoethyl)-3-aminopropyltrimethoxysilane		222.4	351.9
SB	N-(2-aminoethyl)-3-aminopropylmethyldimethoxysilane		206.4	379.2
SC	3-aminopropyltrimethoxysilane		179.3	436.5
SD	3-aminopropyltriethoxysilane		221.4	353.5
SE	-		172.2	454.5
SF	-		258.4	302.9
SG	-		329.3	237.7
SH	N-(2-aminoethyl)-3-aminopropyltriethoxysilane		264.5	295.9
SI	3-triethoxysilyl-(1,3-dimethyl-butylidene)propylamine		303.5	257.9
SJ	N-phenyl-3-aminopropyltrimethoxysilane		255.4	306.0

[0057] [Aggregation Stability Test]

The various polishing compositions (undiluted) were left undisturbed in a 50 °C atmosphere for 30 days, and were evaluated based on the difference between the initial average particle size and the average particle size after the 30-day period at 50 °C. Average particle size was measured using dynamic light scattering (secondary particle size), and was measured by a particle-size measurement system “ELS-Z2” from Otsuka Elctronics Co.

TABLE 3

Test No.			1	2	3	4
Undiluted solution	Abrasives (silica)	Type	A			
		Concentration (wt%)	9.0			
	Chelating agent	Type	DTPA			
		Concentration (wt%)	0.06			
	Basic compound	Type	KOH			
		Concentration (wt%)	0.40			
		Ratio to abrasives	4.4			
	Organic silicon compound	Type	–	SA		
		Concentration (wt%)	–	0.9	0.6	0.3
		Ratio to abrasives	–	10.0	6.7	3.3
	pH		10.26	10.69	10.70	10.63
	Aggregation stability		good	unsat.	good	good
Total surface area of abrasives (m^2)			709.2			
Total minimum area of coating (m^2)			–	316.7	211.1	105.6
Percentage of coating (%)			–	44.7	29.8	14.9
Dilution factor			61			
POU abrasive concentration (wt%)			0.15			
Oxide removal time (sec)			48	1	3	13
Polishing rate (μm/min)			0.08	0.26	0.22	0.12
Ra (nm)			0.24	0.35	0.30	0.23
Difference GBIR (μm)			0.15	0.21	0.19	0.17
Additional info			comp. ex.	inv. ex.	inv. ex.	inv. ex.

[0068] The rows “Ratio to abrasives” for “Basic compound” and “Organic silicon compound” in Table 3 indicate the ratio of the weight of abrasives to the weight of silica, rather than to the total weight, where the weight of the silica is represented as 100. Further, the row “Total surface area of abrasives” indicates the total surface area of the silica for 100 g of the polishing composition (undiluted). “Total minimum area of coating” indicates the total minimum area of coating of the organic silicon compound for 100 g of the polishing composition (undiluted). The row “Percentage of coating” indicates the total minimum area of coating divided by the total surface area of the abrasives and multiplied by 100. The row “POU abrasive concentration” indicates the silica concentration at a point of use, i.e., after dilution. All this applies to Tables 4 to 14 shown below.

[0069] A comparison between Test No. 1 and Test Nos. 2 to 4 shows that adding the organic silicon compound significantly reduced the oxide removal time. A comparison among Test Nos. 2 to 4 shows that the higher the

concentration of the organic silicon compound, the shorter the oxide removal time. It also shows that the higher the concentration of the organic silicon compound, the higher the polishing rate.

[0070] Next, the polishing compositions labeled Test Nos. 3 and 5 to 7, shown in Table 4, were used to investigate the relationship between the dilution factor and oxide removal performance.

[0071] [Table 4]

TABLE 4

Test No.			3	5	6	7
Undiluted solution	Abrasives (silica)	Type	A			
		Concentration (wt%)	9.0			
	Chelating agent	Type	DTPA			
		Concentration (wt%)	0.06			
	Basic compound	Type	KOH			
		Concentration (wt%)	0.40			
		Ratio to abrasives	4.4			
	Organic silicon compound	Type	SA			
		Concentration (wt%)	0.6			
		Ratio to abrasives	6.7			
Total surface area of abrasives (m^2)			709.2			
Total minimum area of coating (m^2)			211.1			
Percentage of coating (%)			29.8			
Dilution factor			61	91	121	151
POU abrasive concentration (wt%)			0.15	0.10	0.07	0.06
Oxide removal time (sec)			3	3	3	2
Polishing rate (μm/min)			0.22	0.20	0.16	0.14
Ra (nm)			0.30	0.25	0.24	0.23
Difference GBIR (μm)			0.19	0.16	0.13	0.15
Additional info			inv. ex.	inv. ex.	inv. ex.	inv. ex.

[0072] As shown in Table 4, the oxide removal performance was maintained even for higher dilution factors (i.e., lower concentrations of the silica and organic silicon compound).

[0073] Next, the polishing compositions labeled Test Nos. 8 to 18, shown in Table 5, were used to investigate the relationship between the type of the organic silicon compound and the oxide removal performance.

[0074] [Table 5]

TABLE 5

Test No.			8	9	10	11	12	13	14	15	16	17	18
Undiluted solution	Abrasives (silica)	Type	A										
		Concentration (wt%)	9.0										
	Chelating agent	Type	DTPA										
		Concentration (wt%)	0.06										
	Basic compound	Type	KOH										
		Concentration (wt%)	0.50										
		Ratio to abrasives	5.6										
	Organic silicon compound	Type	–	SA	SB	SC	SD	SE	SF	SG	SH	SI	SJ
		Concentration (wt%)	–	0.6									
		Ratio to abrasives	–	6.7									
	pH		10.56	10.93	11.00	11.02	11.03	11.00	10.91	10.64	10.93	10.90	10.55
	Aggregation stability		good	good	good	unsat.	unsat.	unsat.	good	unsat.	good	good	unsat.
Total surface area of abrasives (m^2)			709.2										
Total minimum area of coating (m^2)			–	211.1	227.5	261.9	212.1	272.7	181.74	142.62	177.5	154.74	183.6
Percentage of coating (%)			–	29.8	32.1	36.9	29.9	38.5	25.6	20.1	25.0	21.8	25.9
Dilution factor			61										
POU abrasive concentration (wt%)			0.15										
Oxide removal time (sec)			163	6	40	1	5	1	3	20	13	122	92
Polishing rate (μm/min)			0.05	0.20	0.21	0.15	0.13	0.16	0.18	0.17	0.18	0.05	0.05
Ra (nm)			0.25	0.25	0.24	0.25	0.23	0.22	0.25	1.51	0.24	0.20	0.19
Difference GBIR (μm)			0.16	0.22	0.17	0.18	0.17	0.23	0.35	0.67	0.30	0.19	0.09
Additional info			comp. ex.	inv. ex.	inv. ex.	inv. ex.	inv. ex.	inv. ex.	inv. ex.	inv. ex.	inv. ex.	comp. ex.	comp. ex.

TABLE 6

Test No.			19	20	21	22	23	24
Undiluted solution	Abrasives (silica)	Type	A					
		Concentration (wt%)	9.0					
	Chelating agent	Type	DTPA					
		Concentration (wt%)	0.18					
	Basic compound	Type	KOH					
		Concentration (wt%)	0.50	0.65	0.80	0.95	1.10	0.65
		Ratio to abrasives	5.6	7.2	8.9	10.6	12.2	7.2
	Organic silicon compound	Type	SA					–
		Concentration (wt%)	1.8					–
		Ratio to abrasives	20.0					–
	pH		10.57	10.84	11.03	11.20	11.32	10.57
	Aggregation stability		unsat.	good	good	good	good	good
Total surface area of abrasives (m^2)			709.2					
Total minimum area of coating (m^2)			633.4					–
Percentage of coating (%)			89.3					–
Dilution factor			181					
POU abrasive concentration (wt%)			0.05					
Oxide removal time (sec)			1	2	1	3	2	142
Polishing rate (μm/min)			0.22	0.22	0.21	0.21	0.20	0.03
Ra (nm)			0.26	0.27	0.24	0.24	0.25	0.27
Difference GBIR (μm)			0.20	0.22	0.24	0.23	0.25	0.09
Additional info			inv. ex.	inv. ex.	inv. ex.	inv. ex.	inv. ex.	comp. ex.

[0081] As shown in Table 6, changes in the concentration of the basic compound did not affect the oxide removal performance. A tendency was observed that the lower the pH, the lower the aggregation stability.

[0082] Next, the polishing compositions labeled Test Nos. 20 and 24 to 29, shown in Table 7, were used to investigate the time required to remove oxide film for yet larger changes in dilution factor.

[0083] [Table 7]

TABLE 7

Test No.			24	25	26	27	20	28	29
Undiluted solution	Abrasives (silica)	Type	A						
		Concentration (wt%)	9.0						
	Chelating agent	Type	DTPA						
		Concentration (wt%)	0.18						
	Basic compound	Type	KOH						
		Concentration (wt%)	0.65						
		Ratio to abrasives	7.2						
	Organic silicon compound	Type	–	SA					
		Concentration (wt%)	–	1.8					
		Ratio to abrasives	–	20.0					
Total surface area of abrasives (m^2)			709.2						
Total minimum area of coating (m^2)			–	633.4					
Percentage of coating (%)			–	89.3					
Dilution factor			181	31	61	121	181	361	901
POU abrasive concentration (wt%)			0.05	0.29	0.15	0.07	0.05	0.02	0.01
Oxide removal time (sec)			142	12	4	1	2	5	14
Polishing rate (μm/min)			0.03	0.35	0.31	0.27	0.22	0.19	0.14
Ra (nm)			0.27	0.31	0.30	0.30	0.27	0.27	0.23
Difference GBIR (μm)			0.09	0.20	0.23	0.28	0.22	0.23	0.19
Additional info			comp. ex.	inv. ex.	inv. ex.	inv. ex.	inv. ex.	inv. ex.	inv. ex.

[0084] As shown in Table 7, the oxide removal performance was maintained at certain levels, even for a dilution factor of 901. Further, although the reason is not clear, a tendency was observed that an excessively low dilution factor lowered the oxide removal performance. Particularly good oxide removal performance was achieved for a dilution factor in the range of 121 to 181 (i.e., POU abrasive concentration in the range of 0.05 to 0.07 weight %).

[0085] Next, the polishing compositions labeled Test Nos. 20 and 30 to 36, shown in Table 8, were used to investigate the relationship between the type of silica and the oxide removal performance.

[0086] [Table 8]

TABLE 8

Test No.			20	30	31	32	33	34	35	36
Undiluted solution	Abrasives (silica)	Type	A	B	C	D	E	F	G	H
		Concentration (wt%)	9.0							
	Chelating agent	Type	DTPA							
		Concentration (wt%)	0.18							
	Basic compound	Type	KOH							
		Concentration (wt%)	0.65							
		Ratio to abrasives	7.2							
	Organic silicon compound	Type	SA							
		Concentration (wt%)	1.8							
		Ratio to abrasives	20.0							
Total surface area of abrasives (m^2)			709.2	1006.2	420.3	699.3	826.2	1487.7	801.9	1120.5
Total minimum area of coating (m^2)			633.42							
Percentage of coating (%)			89.3	63.0	150.7	90.6	76.7	42.6	79.0	56.5
Dilution factor			181							
POU abrasive concentration (wt%)			0.05							
Oxide removal time (sec)			2	2	2	5	4	1	21	16
Polishing rate (μm/min)			0.22	0.24	0.24	0.22	0.24	0.22	0.22	0.21
Ra (nm)			0.27	0.30	0.31	0.29	0.29	0.29	0.28	0.28
Difference GBIR (μm)			0.22	0.26	0.22	0.27	0.26	0.27	0.24	0.23
Additional info			inv. ex.	inv. ex.	inv. ex.	inv. ex.	inv. ex.	inv. ex.	comp. ex.	comp. ex.

[0087] The polishing compositions labeled Test Nos. 35 and 36 had poor oxide removal performances compared with the polishing compositions labeled Test Nos. 20 and 30 to 34. This is presumably because the silanol group density on the silica surfaces in these polishing compositions was too low.

[0088] Next, the polishing compositions labeled Test Nos. 20 and 37 to 39, shown in Table 9, were used to investigate the effects of the addition of the water-soluble polymer on the oxide removal performance. The row "Ratio relative to abrasives" for "Water-soluble polymer" in Table 9 indicates the ratio of the weight of abrasives to the weight of silica, rather than to the total weight, where the weight of the silica is represented as 100.

[0089] [Table 9]

TABLE 9

Test No.			20	37	38	39
Undiluted solution	Abrasives (silica)	Type	A			
		Concentration (wt%)	9.0			
	Chelating agent	Type	DTPA			
		Concentration (wt%)	0.18			
	Basic compound	Type	KOH			
		Concentration (wt%)	0.65			
		Ratio to abrasives	7.2			
	Organic silicon compound	Type	SA			
		Concentration (wt%)	1.8			
		Ratio to abrasives	20.0			
	Water-soluble polymer	Type	–	HEC	PVA	PGL
		Concentration (wt%)	–	0.36		
		Ratio to abrasives	–	4.0		
Total surface area of abrasives (m^2)			709.2			
Total minimum area of coating (m^2)			633.42			
Percentage of coating (%)			89.3			
Dilution factor			181			
POU abrasive concentration (wt%)			0.05			
Oxide removal time (sec)			2	3	3	8
Polishing rate (μm/min)			0.22	0.10	0.18	0.09
Ra (nm)			0.27	0.16	0.25	0.15
Difference GBIR (μm)			0.22	0.14	0.33	0.12
Additional info			inv. ex.	inv. ex.	inv. ex.	inv. ex.

[0090] As shown in Table 9, the oxide removal performance was not impaired by the addition of the water-soluble polymer.

[0091] Next, the polishing compositions labeled Test Nos. 27, 40 and 41, shown in Table 10, were used to investigate the relationship between the type of basic compound and the oxide removal performance.

[0092] [Table 10]

TABLE 10

Test No.			40	27	41
Undiluted solution	Abrasives (silica)	Type	A		
		Concentration (wt%)	4.5	9.0	6.0
	Chelating agent	Type	DTPA		
		Concentration (wt%)	0.03	0.18	0.02
	Basic compound	Type	KOH	KOH	DETA
		Concentration (wt%)	0.20	0.65	1.00
		Ratio to abrasives	4.4	7.2	16.7
	Organic silicon compound	Type	SA		
		Concentration (wt%)	0.3	1.8	0.3
		Ratio to abrasives	6.7	20.0	5.0
Total surface area of abrasives (m^2)			354.6	709.2	472.8
Total minimum area of coating (m^2)			105.6	633.4	105.6
Percentage of coating (%)			29.8	89.3	22.3
Dilution factor			31	121	121
POU abrasive concentration (wt%)			0.15	0.07	0.05
Oxide removal time (sec)			9	1	1
Polishing rate (μm/min)			0.23	0.27	0.27
Ra (nm)			0.29	0.30	0.31
Difference GBIR (μm)			0.13	0.28	0.21
Additional info			inv. ex.	inv. ex.	inv. ex.

[0093] As shown in Table 10, the oxide removal performance was not affected by a change of the basic compound from an inorganic alkali compound (KOH) to an amine compound (DETA).

[0094] Next, the polishing compositions labeled Test Nos. 20, 24, 42 and 43, shown in Table 11, were used to investigate whether similar levels of oxide removal performance can be achieved if the addition of the organic silicon compound is replaced by the use of silica that has been surface-modified with an amino group or the like in advance.

[0095] [Table 11]

TABLE 11

Test No.			24	42	43	20
Undiluted solution	Abrasives (silica)	Type	A	I	J	A
		Concentration (wt%)	9.0			
	Chelating agent	Type	DTPA			
		Concentration (wt%)	0.18			
	Basic compound	Type	KOH			
		Concentration (wt%)	0.65			
		Ratio to abrasives	7.2			
	Organic silicon compound	Type	–			SA
		Concentration (wt%)	–			1.8
		Ratio to abrasives	–			20.0
	pH		10.62	10.84	10.44	10.84
	Aggregation stability		good	good	good	good
Total surface area of abrasives (m^2)			709.2	689.4	746.1	709.2
Total minimum area of coating (m^2)			–	–	–	633.4
Percentage of coating (%)			–	–	–	89.3
Dilution factor			181			
POU abrasive concentration (wt%)			0.05			
Oxide removal time (sec)			142	94	80	2
Polishing rate (μm/min)			0.03	0.05	0.06	0.22
Ra (nm)			0.27	0.23	0.23	0.27
Difference GBIR (μm)			0.09	0.11	0.16	0.22
Additional info			comp. ex.	comp. ex.	comp. ex.	inv. ex.

[0096] The times required to remove oxide film for the polishing compositions each using silica that had been surface-modified with an amino group or sulfo group in advance (Test Nos. 42 and 43) were shorter than that for Test No. 24, but significantly longer than that for Test No. 20. This shows that levels of oxide removal performance comparable to compositions having the organic silicon compound cannot be achieved by using silica that has been surface-modified with an amino group or the like in advance.

[0097] Next, the polishing compositions labeled Test Nos. 20 and 44 to 49, shown in Table 12, were used to inverstigate the relationship between the concentration of the organic silicon compound and the oxide removal performance for larger changes in the compound concentration. "–" in the row for aggregation stability indicates that aggregation stability was not measured. The same applies to Tables 13 and 14, shown further below.

[0098] [Table 12]

TABLE 12

Test No.			44	45	20	46	47	48	49
Undiluted solution	Abrasives (silica)	Type	A						
		Concentration (wt%)	9.0						1.8
	Chelating agent	Type	DTPA						
		Concentration (wt%)	0.18						
	Basic compound	Type	KOH						NH_4OH
		Concentration (wt%)	0.65						0.50
		Ratio to abrasives	7.2						27.8
	Organic silicon compound	Type	SA						
		Concentration (wt%)	0.3	0.6	1.8	3.6	7.5	15.0	0.12
		Ratio to abrasives	3.3	6.7	20.0	40.0	83.3	166.7	6.7
	Water-soluble polymer	Type	-						HEC
		Concentration (wt%)	-						0.36
		Ratio to abrasives	-						20
	pH		11.05	11.09	10.84	10.96	10.98	11.05	-
	Aggregation stability		good	good	good	good	good	good	-
Total surface area of abrasives (m^2)			709.2						141.8
Total minimum area of coating (m^2)			105.6	211.1	633.4	1266.8	2639.3	5278.5	42.2
Percentage of coating (%)			14.9	29.8	89.3	178.6	372.1	744.3	29.8
Dilution factor			181						
POU abrasive concentration (wt%)			0.05						0.01
Oxide removal time (sec)			14	1	2	1	1	1	11
Polishing rate (μm/min)			0.09	0.15	0.22	0.28	0.31	0.38	0.04
Ra (nm)			0.22	0.26	0.27	0.39	0.34	0.39	0.14
Difference GBIR (μm)			0.09	0.11	0.22	0.15	0.19	0.44	0.09
Additional info			inv. ex.	inv. ex.	inv. ex.	inv. ex.	inv. ex.	inv. ex.	inv. ex.

[0099] Table 12 shows that good oxide removal performance was maintained even when the concentration of the organic silicon compound was increased or decreased.

[0100] Further, Test No. 49 shows that good oxide removal performance can be achieved if the concentrations of the abrasives and the organic silicon compound are reduced and a water-soluble polymer is added.

[0101] Next, the polishing compositions labeled Test Nos. 20, 48, 50 and 51, shown in Table 13, were used to investigate oxide removal performance for even lower POU abrasive concentrations.

[0102] [Table 13]

TABLE 13

Test No.			20	48	50	51
Undiluted solution	Abrasives (silica)	Type	A			
		Concentration (wt%)	9.0		3.0	1.0
	Chelating agent	Type	DTPA			
		Concentration (wt%)	0.18			
	Basic compound	Type	KOH			
		Concentration (wt%)	0.65			
		Ratio to abrasives	7.2		21.7	65.0
	Organic silicon compound	Type	SA			
		Concentration (wt%)	1.8	15.0	15.0	15.0
		Ratio to abrasives	20.0	166.7	500.0	1500.0
	pH		10.84	11.07	11.29	11.39
	Aggregation stability		good	good	–	–
Total surface area of abrasives (m^2)			709.2		236.4	78.8
Total minimum area of coating (m^2)			633.4	5278.5		
Percentage of coating (%)			89.3	744.3	2232.9	6698.6
Dilution factor			181			
POU abrasive concentration (wt%)			0.05	0.05	0.02	0.01
Oxide removal time (sec)			2	1	1	1
Polishing rate (μm/min)			0.22	0.40	0.38	0.36
Ra (nm)			0.27	0.37	0.42	0.53
Difference GBIR (μm)			0.22	0.34	0.34	0.33
Additional info			inv. ex.	inv. ex.	inv. ex.	inv. ex.

[0103] Table 13 shows that the oxide removal performance was maintained even for low POU abrasive concentrations as long as a sufficient amount of the organic silicon compound was present relative to the silica. On the other hand, excessively large amounts of the organic silicon compound relative to the silica tended to increase Ra. Further, dissolution of the silica during aggregation stability testing was observed in Test Nos. 50 and 51. This shows that the concentration of the organic silicon compound is preferably not higher than 300 parts by weight, where the amount of silica is represented as 100 parts by weight.

[0104] Lastly, the polishing compositions labeled Test Nos. 21, 52 and 53 shown in Table 14 were used to investigate the amount of the organic silicon compound relative to the silica and the oxide removal performance.

[0105] [Table 14]

TABLE 14

Test No.			21	52	53
Undiluted solution	Abrasives (silica)	Type	A		
		Concentration (wt%)	9.0	9.0	15.0
	Chelating agent	Type	DTPA		
		Concentration (wt%)	0.18		
	Basic compound	Type	KOH		
		Concentration (wt%)	0.80	0.50	0.40
		Ratio to abrasives	8.9	5.6	2.7
	Organic silicon compound	Type	SA		
		Concentration (wt%)	1.8	0.3	0.3
		Ratio to abrasives	20.0	3.3	2.0
	pH		11.03	10.67	9.45
	Aggregation stability		good	–	–
Total surface area of abrasives (m^2)			709.2		1182.0
Total minimum area of coating (m^2)			633.4	105.6	
Percentage of coating (%)			89.3	14.9	8.9
Dilution factor			181		
POU abrasive concentration (wt%)			0.05	0.05	0.08
Oxide removal time (sec)			1	8	15
Polishing rate (μm/min)			0.21	0.08	0.05
Ra (nm)			0.24	0.25	0.23
Difference GBIR (μm)			0.24	0.07	0.09
Additional info			inv. ex.	inv. ex.	inv. ex.

[0106] Table 14 demonstrates that the oxide removal performance was maintained even when the concentration of the organic silicon compound was as low as 2.0 parts by weight, where the amount of silica is represented as 100 parts by weight.

[0107] Embodiments of the present invention have been described. The above-described embodiments are exemplary only, intended to allow the present invention to be carried out. Accordingly, the present invention is not limited to the above-described embodiments, and the above-described embodiments, when carried out, may be modified as appropriate without departing from the spirit of the invention.