Fig. 5

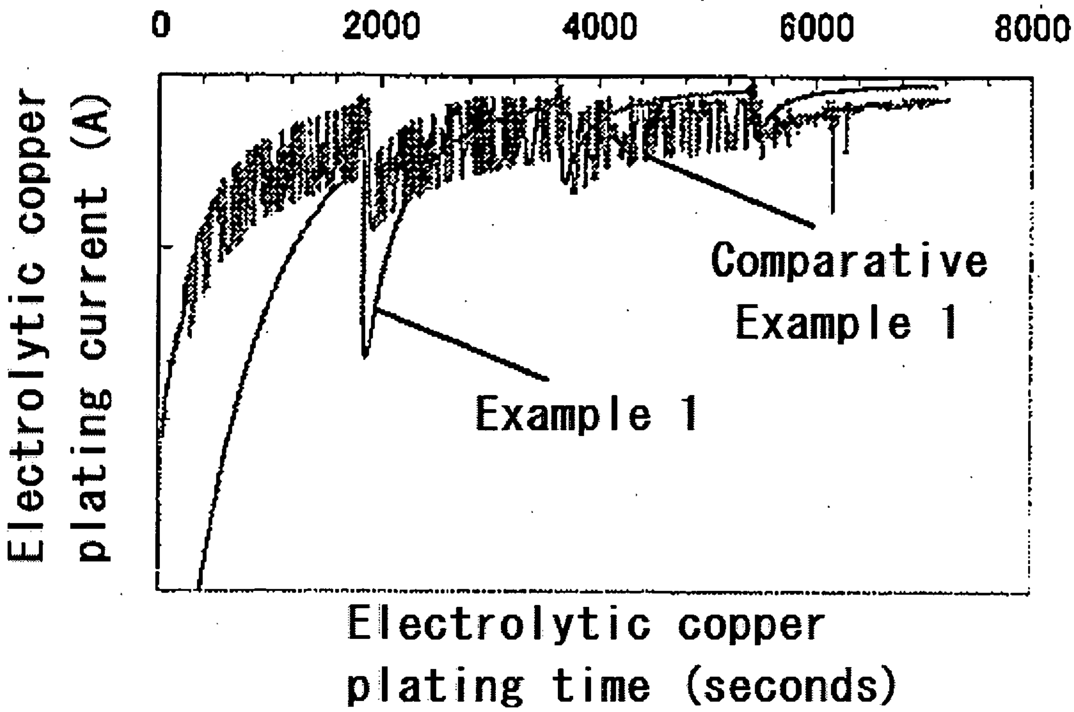

Fig. 6

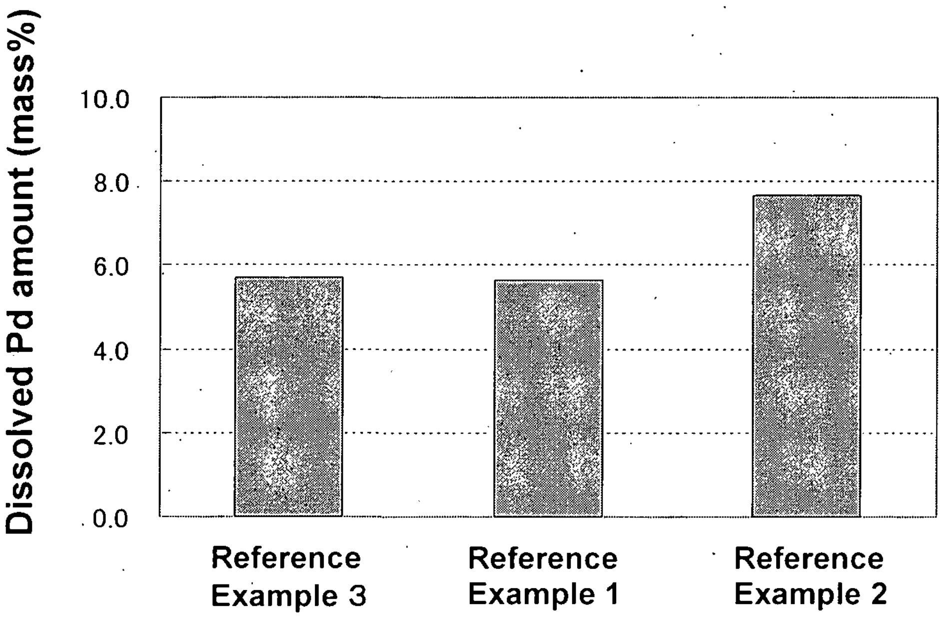

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Table 2

	Example 2	Comparative Example 3
Type of palladium-supported carbon	Production Example 1	Production Example 3
MV (μm)	1.586	7.190
MN (μm)	0.121	0.220
MA (μm)	0.242	0.591
CS	24.762	10.161
SD (μm)	0.493	5.829
x_{50} (μm)	0.367	0.730
x_{90} (μm)	2.105	27.700

[0091]

Fig. 10 is a graph showing the particle size distribution and cumulative curve of a palladium-supported carbon of Production Example 3, which are overlapped on each other. Fig. 10 is a graph with particle diameter (μm) on the horizontal axis, frequency (%) on the left vertical axis, and cumulative index (%) on the right horizontal axis. In Fig. 10, a thick line graph shows particle size distribution, and a thin line graph shows a cumulative curve.

Table 1 shows that the palladium-supported carbon of Production Example 3 has an MV of 35.468 μm, an MN of 5.010 μm, an MA of 16.732 μm, a CS of 0.359, and an SD of 25.752 μm. Accordingly, the MN is within a range of 1 to 10 μm. On the other hand, the MV is a value which is more than 20 μm; the MA is a value which is more than 10 μm; and the SD is a value which is more than 5 μm.

Table 1 shows that the palladium-supported carbon of Production Example 3 has an x_{50} of 29.720μm and an x_{90} of 70.140

μm. Accordingly, the x_{50} is a value which is more than 20 μm, and the x_{90} is a value which is more than 30 μm.

From Fig. 10, it is clear that in the palladium-supported carbon of Production Example 3, most of the particles have a particle diameter of 10 to 100 μm.

[0092]

Fig. 11 is a graph showing the particle size distribution and cumulative curve of a catalyst for fuel cells used in Comparative Example 3, which are overlapped on each other. Fig. 11 is a graph with particle diameter (μm) on the horizontal axis, frequency (%) on the left vertical axis, and cumulative index (%) on the right horizontal axis. In Fig. 11, a thick line graph shows particle size distribution, and a thin line graph shows a cumulative curve.

Table 2 shows that the catalyst for fuel cells used in Comparative Example 3 has an MV of 7.190 μm, an MN of 0.220 μm, an MA of 0.591 μm, a CS of 10.161, and an SD of 5.829 μm. Table 2 also shows that the catalyst for fuel cells used in Comparative Example 3 has an x_{50} of 0.730 μm and an x_{90} of 27.700 μm.

From Fig. 11, it is clear that particles having a particle diameter of 10 μm or more remained in the catalyst for fuel cells used in Comparative Example 3. From this result, it is clear that the catalyst for fuel cells comprising, as a raw material, the palladium-supported carbon having an MV of more than 20 μm, an MA of more than 10 μm, an SD of more than 5 μm, an x_{50} of more than 20 μm, and an x_{90} of more than 30 μm, cannot obtain a good particle size distribution when it is fined by a weaker dispersion method than conventional dispersion methods.

[0093]

Fig. 2 is a graph showing the particle size distribution and cumulative curve of a palladium-supported carbon of Production Example 1, which are overlapped on each other. Fig. 2 is a graph with particle diameter (μm) on the horizontal axis, frequency (%) on the left vertical axis, and cumulative index (%) on the right horizontal axis. In Fig. 2, a thick line graph shows particle size distribution, and a thin line graph shows a cumulative curve.

Table 1 shows that the palladium-supported carbon of Production Example 1 has an MV of 10.323 μm, an MN of 5.527 μm, an MA of 8.669 μm, a CS of 0.692, and an SD of 3.733 μm. Accordingly, the MV is within a range of 1 to 20 μm; the MN is within a range of 1 to 10 μm; the MA is within a range of 1 to 10 μm; and the SD is within a range of 1 to 5 μm.

Table 1 shows that the palladium-supported carbon of Production Example 1 has an x_{50} of 9.777 μm and an x_{90} of 15.490 μm. Accordingly, the x_{50} is within a range of 1 to 20 μm, and the x_{90} is within a range of 10 to 30 μm.

From Fig. 2, it is clear that the particle size distribution of the palladium-supported carbon of Production Example 1 is an almost normal distribution, and the peak of the particle size distribution is about 10 μm.

[0094]

Fig. 3 is a graph showing the particle size distribution and cumulative curve of a catalyst for fuel cells used in Example 2, which are overlapped on each other. Fig. 3 is a graph with particle diameter (μm) on the horizontal axis, frequency (%) on the left vertical axis, and cumulative index (%) on the right horizontal axis. In Fig. 3, a thick line graph shows particle size distribution, and a thin line graph shows a cumulative curve.

Table 2 shows that the catalyst for fuel cells used in Example 2 has an MV of 1.586 μm, an MN of 0.121 μm, and MA of 0.242 μm, a CS of 24.762, and an SD of 0.493 μm. Also Table 2 shows that the catalyst for fuel cells used in Example 2 has an x_{50} of 0.367 μm and an x_{90} of 2.105 μm.

According to Fig. 3, almost no particles having a particle diameter of 10 μm or more remain in the catalyst for fuel cells used in Example 2. From this result, it is clear that the catalyst for fuel cells comprising, as a raw material, the palladium-supported carbon having an MV within a range of 1 to 20 μm, an MN within a range of 1 to 10 μm, an MA within a range of 1 to 10 μm, an SD within a range of 1 to 5 μm, an x_{50} within a range of 1 to 20 μm, and an x_{90} within a range of 10 to 30 μm, can obtain a good particle size distribution, with maintaining the particle structure, even when the it is fined by a weaker dispersion method than conventional dispersion methods.

[0095]

Fig. 9 is a graph showing the particle size distribution and cumulative curve of a palladium-supported carbon of Production Example 2, which are overlapped on each other. Fig. 9 is a graph with particle diameter (μm) on the horizontal axis, frequency (%) on the left vertical axis, and cumulative index (%) on the right horizontal axis. In Fig. 9, a thick line graph shows particle size distribution, and a thin line graph shows a cumulative curve.

Table 1 shows that the palladium-supported carbon of Production Example 2 has an MV of 0.642 μm, an MN of 0.229 μm, an MA of 0.409 μm, a CS of 14.689, and an SD of 0.358 μm. Accordingly, the MV is less than 1 μm; the MN is less than 1 μm; the MA is less than 1 μm; and the SD is less than 1 μm.

Table 1 shows that the palladium-supported carbon of Production Example 2 has an x_{50} of 0.506 μm and an x_{90} of 1.452 μm. Accordingly, the x_{50} is less than 1 μm, and the x_{90} is less than 10 μm.

From Fig. 9, it is clear that in the palladium-supported carbon of Production Example 2, most of the particles have a particle diameter of 0.1 to 1 μm.

However, as described above, it is clear that poor electricity conduction is caused between the palladium-supported carbon of Production Example 2 and the electrode at the time of electrolytic copper plating.

[0096]

6. Palladium dissolution test (durability test)

in the same manner as Example 1. That is, in Reference Example 3, no dispersion was carried out after the covering with Pt.

[0099]

6-2. Palladium dissolution test

A palladium dissolution test was carried out on the catalysts for fuel cells of Reference Examples 1 to 3.

First, each catalyst for fuel cells was immersed in 0.1 mol/L sulfuric acid at 80°C. One hour later, the sulfuric acid was filtered. For the thus-obtained filtrate, the palladium element dissolved in the sulfuric acid was quantitated by inductively coupled plasma mass spectroscopy (ICP-MS).

Fig. 6 is a bar graph showing dissolved palladium amounts eluted from palladium-supported carbons of Reference Examples 1 to 3. The graph of Fig. 6 is a graph with dissolved palladium amount (mass%) on the vertical axis. Fig. 6 shows the following: in the catalyst for fuel cells of Reference Example 1, the dissolved palladium amount is 5.65 mass%; in the catalyst for fuel cells of Reference Example 2, the dissolved palladium amount is 7.67 mass%; and in the catalyst for fuel cells of Reference Example 3, the dissolved palladium amount is 5.70 mass%.

Accordingly, it is clear that the dissolved palladium amount of Reference Example 2, in which the catalyst for fuel cells were fined under the severer condition, is about 1.3 times higher than Reference Example 1, in which the catalyst for fuel cells were fined under the milder condition, and Reference

Fig. 5

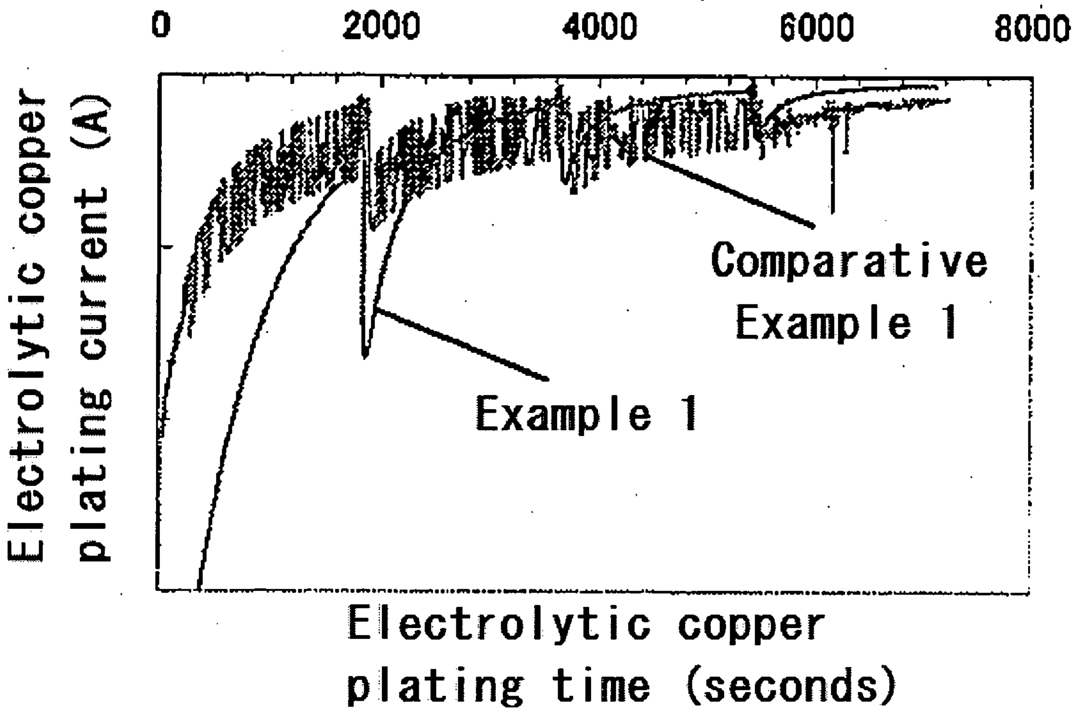
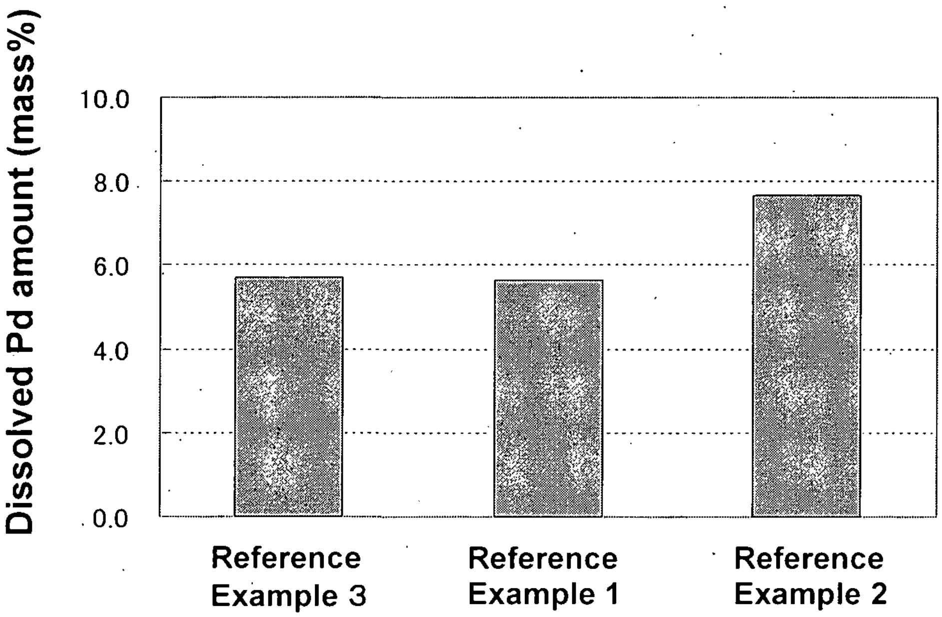


Fig. 6



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