ABSTRACT

Some 100 μg of fine black porous powders were deposited at the bottom of the electrolytic cell during the electrolysis on Au electrodes for 20-30 days at current densities above 200 mA/cm². The main constituting elements of the deposits were Hg, Pt, Os, Fe, Si and F other than Au. The isotopic distributions of Hg, Fe and Si containing in the deposits were evidently different from their natural ones. In addition, a lot of micro craters which are allowed to imagine the occurrence of some micro explosion were created on Au electrode surface during the electrolysis.

INTRODUCTION

In the previous study, we found that some 10 μg of Fe, whose isotopic distribution was clearly different from its natural isotopic abundance, was formed on/in Au electrode during the electrolysis for 7 days at a current density of 100 mA/cm². In addition, there was a linear relationship between the amount of Fe and excess heat observed simultaneously [1-2]. This suggests strongly that Fe was produced by some nuclear transmutation. In the present study, we investigated whether there exist some elements other than Fe produced as a result of nuclear transmutation by noting the deposits obtained in the present electrolysis conducted at high current densities.

EXPERIMENTAL

The electrolytic cell was made of fused quartz. The working electrodes were cold worked Au plates (2.0 cm² app. area; 0.1 mm thick; 99.99 % purity; Ag: 21 ppm, Pd: 3 ppm, Cu: 1 ppm, Fe, Si, Rh, Pt: < 1 ppm, no detectable for Hg, Os and Zn), whose surfaces were scraped with a cleaned glass fragment. The roughness factor of the electrode was around 2.0. The counter electrode was a 1 x 7 cm, 80 mesh Pt gauze (99.98 % purity; Rh: 18 ppm, Pd, Cr, Si: 2 ppm, no detectable for Hg, Os and Zn). The electrolytes used were Na₂SO₄ and Na₂CO₃ from Merck (sprapur grade, Fe, Zn: < 0.01 ppm, no detectable for Si, Hg, Os and F for Na₂SO₄; Si: < 0.5 ppm, Fe, Hg: < 0.05 ppm, Zn: < 0.01 ppm, no detectable for Os and F for Na₂CO₃). The volume of the electrolyte...
solution was 100 ml and the concentration was adjusted to 0.5 M. The electrolysis was carried out at a constant current ranging from 1 to 3 A. During the electrolysis, Milli-Q water was supplied every 24 hours to maintain the total solution amount constant. The deposits obtained were washed with Milli-Q water and placed on Ni or Zr plates and the constituent elements of the deposits were analyzed by means of SIMS, AES, EPMA and EDX.

RESULTS AND DISCUSSION

Analysis of the deposits

The deposits were very porous with a lot of fine pores of nm order, look like pumices. Maximal constituent element of the deposits was Au, besides, considerable amounts of Hg, Pt, Os, Fe, Si, and F were contained. The yield of Si was increased significantly when electrolysized in Na2SO4 at a high current density. The contents of Fe and F were increased in Na2CO3. Os was detected mainly in Na2CO3 at a high current density.

As a typical example, we show EPMA images of the deposits obtained after the electrolysis in Na2SO4 for 30 days at a current density of 800 mA/cm² in Fig. 1. As seen, in this case, fairly clear Hg, Pt and Os images were observed together with the strongest Au image. Fe image was also observed although it was a little weaker in this case than those of above elements. The fact that the major component of the deposits was Au suggests strongly that the deposits were produced at the Au electrode.

Figure 1. EPMA images of elements containing in the deposits. SEM image of the deposits (x 6,700) (a), EPMA images of Au (b), Hg (c), Pt (d), Os (e) and Fe (f). Wavelength of the specific X rays are 1.276, 1.241, 1.313, 3.163 and 1.937 Å for Au, Hg, Pt, Os and Fe, respectively.
Isotopic abundance

The isotopic distributions of Hg, Fe and Si containing in the deposits obtained by SIMS analysis are shown in Figs 2 - 4, respectively, over the scan range up to 15. The data of Hg and Fe were obtained in Na₂CO₃ and the datum of Si was obtained in Na₂SO₄ at a current density of 800 mA. The isotopic distributions of these elements reveal significant deviation from their natural isotopic abundance. Thus for Hg, Fig. 2, the isotopic content of ²⁰⁹Hg is 35-50 %, evidently higher than its natural isotopic abundance of 23.13 %, while the content of ²⁰⁸Hg, which being the major component in the nature, is reduced down to 15-20 % from its natural value of 29.8 %. The components of other isotopes change to some extent.

For Fe, Fig. 3, the content of ⁵⁷Fe drastically increases as in the case of Fe produced on/in Au electrode during the electrolysis at a current density of 100 mA/cm² [1-2], however, in this case, its ratio becomes much higher, exceeding 50 % particularly in the vicinity of the surface of the deposits. The ratio of ⁵⁷Fe has a tendency to decrease gradually with increasing the depth.

As mentioned already, the content of Si in the deposits markedly increased when electrolyzed in Na₂SO₄ at a large current density (eg. 800 mA/cm²), and, in this case, the color of the deposits turns white gray. From EDX measurement the major component was found to be Si, besides Au, Pt, Hg were also included. The isotopic content of ²⁸Si decreased down to ca. 58 % from its natural value of 92.2 % in the vicinity of the surface of the deposits.

On the contrary, the content of ²⁹Si and ³⁰Si increased. The extent of the decrease in the content of ²⁸Si of the deposits obtained by the electrolysis in Na₂CO₃ at a current density of 500 mA/cm² is somewhat smaller, the content is 70-75 % in the vicinity of the surface. These results show that ²⁹Si and ³⁰Si are produced preferentially by the electrolysis in Na₂SO₄ at a large current density. As have reported already, the isotopic distribution of Si present on/in Au electrode after the electrolysis in Na₂SO₄ at a current density of 800 mA/cm² is shown in Fig. 4.
density of 100 mA/cm² agreed closely with natural isotopic abundance [2], and, in the present experiment, the isotopic distribution of Si present on/in the Au electrode after the electrolysis in Na₂CO₃ at a current density of 500 mA/cm² was also in accord with its natural isotopic abundance. Such Si is considered to be the one of glass powder remained in the Au electrode after scraping. Hence, there would be no doubt that ²⁹Si and ³⁰Si were produced at Au electrode by some nuclear reaction occurring when the deposits were produced.

Surface structure of Au electrode

The electrode surface after the electrolysis showed very anomalous structure. Fig. 5 shows the SEM images of the Au electrode surface after the electrolysis in Na₂CO₃ for 30 days at a current density of 500 mA/cm². A lot of craters with various sizes are created and developed along the scraped edge lines of the electrode surface (c,d). Each crater has a deep hole, looks like volcano. The size of the largest crater reaches ca. 20 μm diam. and 30 μm height (a,b). From the appearance of the craters, one can imagine that some micro explosion took place at that place. The outside walls of the craters consist of very fine porous substances, which are very alike to the structure of the deposits. In addition, the major element of the deposits is Au. Hence, there would be no doubt that the deposits were spewed out from these craters as the ashes as a result of some micro explosion, probably caused by some nuclear reactions.

The walls inside the craters are constituted by fine hexagonal crystallites which are assignale Au(111) (e,f). These crystallites may be considered to be produced by the reconstruction of Au polycrystals of the electrode material owing to the intense heat evolved locally resulting from the micro explosion. The amounts of Hg and Pt present on/in Au electrode after the electrolysis determined by SIMS were much smaller than those present in the deposits in every cases. Perhaps, O⁺ irradiation of SIMS was not pin-pointed at the wall of the craters precisely.

Possible reactions

The amount of deposits obtained during the electrolysis was 0.5 - 1 mg. The content of Hg, Pt and Os in the deposit can be estimated at least at several percents from the results of EPMA measurement, from which the total amounts of Hg, Pt and Os produced yield at least some 10 μg. On the other hand, even in 100 ml of the electrolyte solution of 0.5 M Na₂CO₃ containing 0.05 ppm of Hg as impurity, the amount of Hg which must be contained in the deposits as impurity would be at most 0.35 μg. In addition, Os and F are completely free in the electrolysis system used. The deviation of the isotopic distribution of Hg, Fe and Si suggests that these elements were not mixed in the deposits as impurities but produced as a result of some nuclear transmutations. Hence, at least, Hg, Os, Fe, Si and F can be considered to be the products of the nuclear transmutation reaction

Although the reaction scheme of the nuclear transmutations is still ambiguous, we consider at the present stage that the reactions start by the following fusion reaction, ⁹⁷Au + n⁺H → ¹²⁸⁻²⁰⁴Hg. The Hg atoms thus produced would have very high nuclear energies, being unstable, splitting partly to produce Pt, Os and Fe. Si and F would be produced as a result of the split of Fe occurring successively.

It is very mysterious why such transmutations take place in Au electrode by the light water electrolysis at a room temperature. It is known that there are small amount of H
active sites on Au where the hydrogen evolution reaction occurs exclusively by slow recombination reaction [3-5]. Therefore, at that place where such sites are concentrated (perhaps, it corresponds to the lattice defect), the concentration of H would remarkably increase with increasing hydrogen overpotential, and, in addition, a considerable amount of micro cracks would be created with high loading of H. As a result of this, extremely strong electric field would be formed. Under such a condition, some H atoms would have a chance to acquire extremely high kinetic energy and become accessible to Au nuclei enough to cause the nuclear transmutation reaction even instantaneously.

Of course, our concepts concerning the reaction scheme are far from the completion. Nevertheless there is hardly any doubt that the nuclear transmutation reaction occurs in the electrolysis systems to produce Hg, Os, Fe, Si, F, etc. from the experimental results obtained here. To elucidate this problem, more extensive research would be necessary.

REFERENCES