

Article

Aurihydrargyrumite, a Natural Au_6Hg_5 Phase from Japan

Daisuke Nishio-Hamane ^{1,*}, Takahiro Tanaka ² and Tetsuo Minakawa ³¹ Institute for Solid State Physics, the University of Tokyo, Kashiwa, Chiba 277-8581, Japan² Sunagawa cho, Tachikawa, Tokyo 190-0031, Japan; penta@kuh.biglobe.ne.jp³ Ehime University Museum, Ehime University, Matsuyama, Ehime 790-8577, Japan; minagawa@sci.ehime-u.ac.jp

* Correspondence: hamane@issp.u-tokyo.ac.jp; Tel.: +81-4-7136-3462

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Abstract: Aurihydrargyrumite, a natural Au_6Hg_5 phase, was found in Iyoki, Uchiko, Ehime Prefecture, Shikoku Island, Japan. Aurihydrargyrumite with a metallic silver luster occurs as a submicron- to 2 μm -thick layer on the outermost surface of the placer gold. A prismatic face may be formed by {001} and {100} or {110}. The streak is also silver white and its Mohs hardness value is ca. 2.5. Its tenacity is ductile and malleable, and its density, as calculated based on the empirical formula and powder unit-cell data, is $16.86 \text{ g}\cdot\text{cm}^{-3}$. The empirical formula of aurihydrargyrumite, on the basis of 11 Au + Hg, is $\text{Au}_{5.95}\text{Hg}_{5.05}$. Aurihydrargyrumite is hexagonal, $P6_3/mcm$, with the lattice parameters $a = 6.9960(10) \text{ \AA}$, $c = 10.154(2) \text{ \AA}$ and $V = 430.40(15) \text{ \AA}^3$, which is identical with the synthetic Au_6Hg_5 phase. The seven strongest lines in the powder X-ray diffraction (XRD) pattern [d in $\text{Å}(I/I_0)(hkl)$] were 2.877(29)(112), 2.434(42)(113), 2.337(100)(104), 2.234(87)(211), 1.401(39)(314), 1.301(41)(404), and 1.225(65)(217). Aurihydrargyrumite forms through the weathering of mercury-bearing placer gold by involvement of self-electrorefining. This new mineral has been approved by the IMA-CNMNC (2017-003) and it is named for its composition, being a natural amalgam of gold (Latin: aurum) and mercury (Latin: hydrargyrum).

Keywords: aurihydrargyrumite; Au_6Hg_5 phase; gold; placer; self-electrorefining; Ehime; Japan

1. Introduction

The investigation of the phase relations in the Au–Hg system has a long history. In the latest phase diagram [1], three amalgam solid phases appear in the system: Au_4Hg , Au_3Hg , and Au_2Hg . Additionally, Lindahl [2] reported a metastable Au_6Hg_5 phase synthesized by reducing gold and mercury ions. Au_6Hg_5 phase is stable around room temperature, while it alters by leaching the mercury component at temperatures of above 70 °C [2].

In nature, several amalgam phases without structural identification have been reported from the placer deposit from the Palakharya River, Bulgaria [3,4]. They are polymineral aggregates consisting of stoichiometric compounds among gold, silver, and mercury: (Au,Ag,Hg) phases with mercury contents of up to 3 wt.%, $(\text{Au,Ag})_3\text{Hg}$, $(\text{Au,Ag})_2\text{Hg}$, $(\text{Au,Ag})_3\text{Hg}_2$, and AuAgHg phases. A mineral with $(\text{Au,Ag})_{1.2}\text{Hg}_{0.8}$ ($=\text{Au}_3\text{Hg}_2$) composition was described as the new mineral weishanite from the Weishancheng ore field, China [5]. Although the stoichiometric compound $(\text{Au,Ag})_3\text{Hg}_2$ does not exist on the phase diagram, the symmetry and unit-cell parameters for weishanite correspond to those of a synthetic Au_3Hg_2 phase [5]. Amalgam grains with $\text{Au}_{94-88}\text{Hg}_{6-12}$ composition were also found in Pleistocene alluvial deposits along the Snake River near Blackfoot, Idaho [6]. The X-ray diffraction (XRD) pattern of this amalgam was indexed as monoclinic symmetry, however this phase also does not appear in the phase diagram.

The anthropogenic mercury that was used to collect gold in the past affects the composition and texture of present gold grains through weathering processes. Barkov et al. [7] reported zoned amalgam of composition $(\text{Au}_{1.5-1.9}\text{Ag}_{1.1-1.4})\Sigma_{2.8-3.0}\text{Hg}_{1.0-1.2}$ from a placer deposit in the Tulameen–Similkameen river system, British Columbia, Canada. They discussed the concentration of the mercury component at the grain rim by some electrochemical factors that are related to the process of self-electrorefining, although they considered the amalgam grain to be anthropogenic in origin [7]. Recently, Svetlitskaya et al. [8] described native gold from Inagli Pt–Au placer deposit, in the Aldan Shield, Russia. Most gold placer was determined to be natural in origin, while some gold grains with a brain-like appearance were rich in a mercury component brought by humans to recover gold in this region [8].

Many gold deposits, including placer deposits, have been reported in the Japanese Islands [9,10]. For example, the gold deposits of Shikoku Island, Ehime Prefecture, were summarized by Miyahisa and Higaki [11]. Although gold was recovered as a by-product of the Kieslager-type copper deposit, there are no deposits that are mined primarily for gold in Ehime [11]. Additionally, a small amount of placer gold was reported in the basin of the Dozan River in eastern Ehime. In the present study, we conducted a survey in an area where gold has not been known to occur to date, and discovered a new placer locality in southwest Ehime. There, the surface of some gold particles is partly or completely coated by metal with a silver luster. We subsequently studied this unidentified silver metal and found that it corresponds to a new mineral, a natural Au_6Hg_5 phase.

The new mineral, which we name aurihydrargyrumite, is named after its chemical composition, being a natural amalgam of gold (Latin: aurum) and mercury (Latin: hydrargyrum). Both the new mineral and its name have been approved by the International Mineralogical Association, Commission on New Minerals, Nomenclature and Classification (no. 2017-003). The type specimen has been deposited in the collections of the National Museum of Nature and Science, Japan (specimen number NSM-M45047). We herein describe this new mineral.

2. Occurrence

Aurihydrargyrumite, which is a natural Au_6Hg_5 phase, was found in a placer from Iyoki, Uchiko, Ehime Prefecture, Shikoku Island, Japan. The discovery site is located in the middle of the Oda river; Figure 1 shows the geological map for the investigated area. The placer was carried by the Oda and Tamatani rivers, which flow through areas where Sanbagawa metamorphic rocks and Mikabu greenstone are distributed. Sanbagawa metamorphic rocks mainly consist of mafic, pelitic, and psammitic schists. Mikabu greenstone consists of metamorphosed basaltic tuff and lava, metagabbro, and metadolerite.

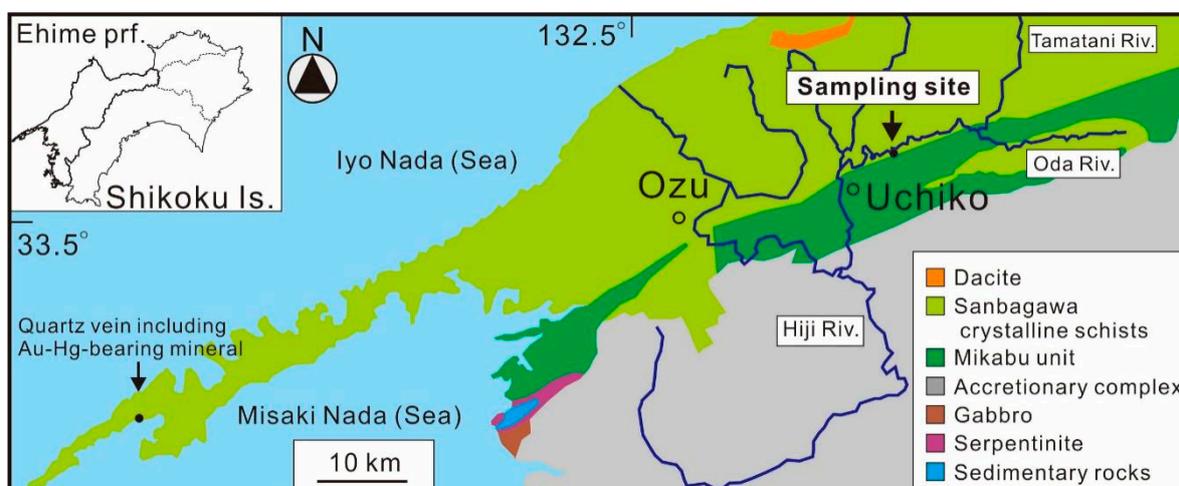


Figure 1. Geological map and water system of the investigated area, southwest Ehime Prefecture, Shikoku Island, Japan.

These units belong to the Sanbagawa metamorphic rocks. Mikabu greenstone contains significant amounts of Kieslager-type copper deposits and manganese deposits. Although no gold deposit has yet been reported in the investigated area [11], we found a small quartz vein that included the gold- and mercury-bearing mineral from the Sanbagawa metamorphic rocks (Figure 1). Subsequently, we investigated the placer of rivers and found a small amount of gold placer from Iyoki.

The placer collected from Iyoki consists of ilmenite, magnetite, chromite, zircon, scheelite, gold, and the platinum-group minerals (PGMs) iridium, osmium, and irarsite. Aurihydrargyrumite is closely associated with gold and is formed through the weathering of mercury-bearing gold, as discussed later.

3. Appearance and Physical Properties

The surface of some gold particles is partly or completely coated by aurihydrargyrumite with a metallic silver luster (Figure 2a). Such particles show a zoned internal texture (Figure 2b). Aurihydrargyrumite occurs as a thin layer on the outermost surface of the gold particles. Beneath the aurihydrargyrumite layer, a gold-rich zone containing mercury is distributed. The core of the particle consists of silver- and mercury-bearing gold.

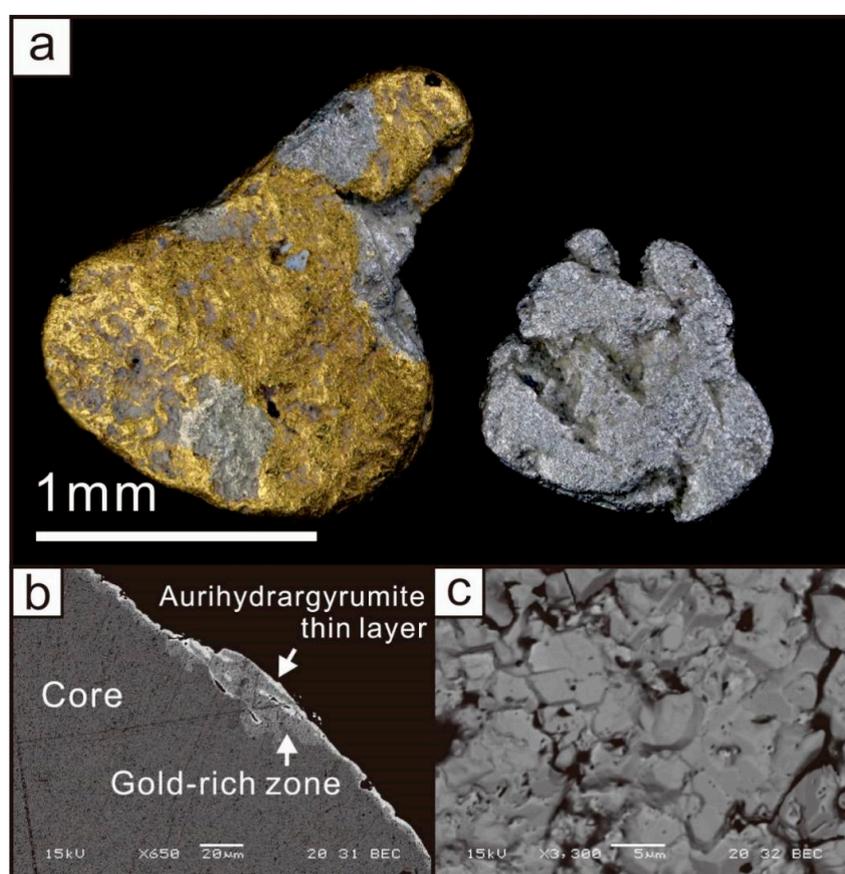


Figure 2. (a) Photographic image of aurihydrargyrumite (silver material); (b) backscattered electron image of a grain cross section; and (c) backscattered electron image of the surface of the grain.

Aurihydrargyrumite with a metallic silver luster occurs as a thin, submicron- to 2 µm-thick layer on the outermost surface of the gold particles. The mineral is commonly anhedral, and occasionally shows subhedral hexagonal-like crystals up to 2 µm in width (Figure 2c). Prismatic crystal may be formed by {001} and {100} or {110}. Its streak is silver white, its Mohs hardness value is ca. 2.5, and its tenacity is ductile and malleable. The density, as calculated based on the empirical formula and powder unit-cell data, is 16.86 g·cm⁻³. Other physical and optical data were not determined due to the small size of the crystals.

4. Chemical Composition

Table 1 summarizes the chemical analysis of aurihydrargyrumite. The data were obtained from the natural surface, because the aurihydrargyrumite layer on the thin section is too small to analyze. Attempts to use WDS could not be used reliably for analysis due to the non-horizontal surface. Therefore, chemical analyses were carried out while using a JEOL JSM-5600 scanning electron microprobe (EDS mode, 15 kV, 0.4 nA, 1 μ m beam diameter), and the ZAF method was used for data correction. The slightly high analytical total obtained is probably due to the irregular surface topography. The empirical formula of aurihydrargyrumite, on the basis of 11 Au + Hg, is Au_{5.95}Hg_{5.05}. The ideal formula is Au₆Hg₅, which requires Au 54.09 and Hg 45.91 for a total of 100 wt.%.

Table 1 also shows the chemical compositions inside the particles on which the aurihydrargyrumite layer was observed. The gold-rich zone consists of a gold–mercury alloy, while the silver components are generally absent. The core of the particle consists mainly of a gold–silver–mercury alloy.

Table 1. Analytical data for aurihydrargyrumite and the inside of the particles.

| | Aurihydrargyrumite | | Gold-Rich Zone | Core |
|------------|---------------------|-------|---------------------|---------------------|
| | Mean 5 (Range) wt.% | Ideal | Mean 5 (Range) wt.% | Mean 5 (Range) wt.% |
| Au | 54.92 (54.26–55.76) | 54.09 | 96.82 (95.47–98.73) | 88.20 (88.15–88.87) |
| Ag | - | - | - | 9.90 (9.83–10.04) |
| Hg | 47.50 (46.54–48.91) | 45.91 | 2.96 (1.41–4.60) | 1.69 (1.28–2.17) |
| Total wt.% | 102.42 | 100 | 99.78 | 99.79 |
| | pfu | | pfu | pfu |
| Au | 5.95 | 6 | 97.09 | 81.72 |
| Ag | - | - | - | 16.75 |
| Hg | 5.05 | 5 | 2.91 | 1.53 |
| Σ | 11 | 11 | 100 | 100 |

5. X-ray Crystallography

The micro-XRD measurement technique was applied for the powder XRD, which was carried out using a Rigaku ultrax18 (CrK α radiation, 40 kV, 200 mA, 100 μ m collimator) instrument equipped with a curved position sensitive proportional counter and an oscillation sample stage. The powder XRD spectrum of aurihydrargyrumite was successfully obtained from the surface of the particle while using this method. The XRD spectrum is shown in Figure 3, and the resulting data are summarized in Table 2. The seven strongest lines in the powder XRD spectrum [d in \AA (I/I_0)(hkl)] are 2.877(29)(112), 2.434(42)(113), 2.337(100)(104), 2.234(87)(211), 1.401(39)(314), 1.301(41)(404) and 1.225(65)(217). Based on these data, aurihydrargyrumite can be indexed to the hexagonal $P6_3/mcm$ space group. The unit-cell parameters as refined from the powder data are $a = 6.9960(10)$ \AA , $c = 10.154(2)$ \AA and $V = 430.40(15)$ \AA^3 . The c/a ratio calculated from the unit-cell parameters is 1.451.

Aurihydrargyrumite contains one Au site and two Hg sites. Each distinct site forms a sheet in the ab plane (Figure 4). Gold atoms form triangular trimers, which are arranged in a triangular net in the Au sheets. The Hg atoms form a ditrigonally distorted Kagome net in the Hg1 sheets, but a honeycomb net in the Hg2 sheets. Two Au sheets and one Hg1 sheet form a compound Au–Hg1–Au layer and the next such layer is rotated 60° around the c -axis. The Hg2 sheets occur between the layers.

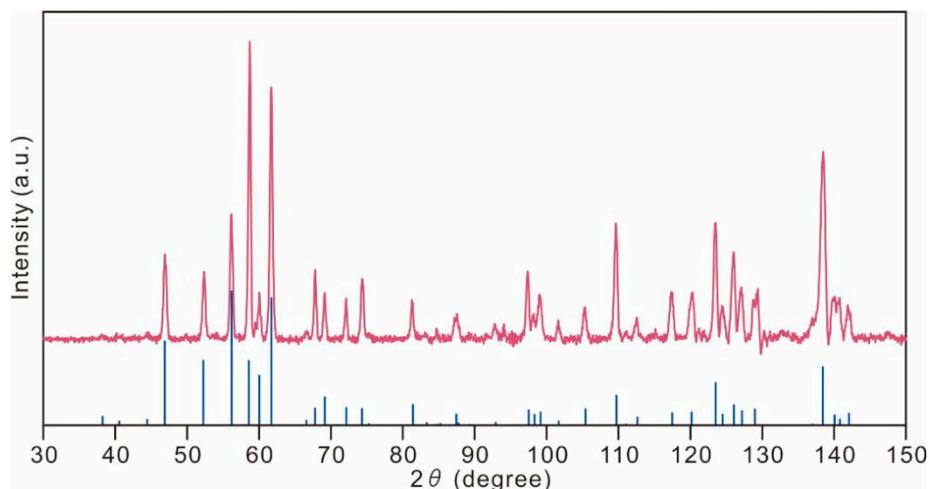


Figure 3. X-ray diffraction spectrum (CrK α radiation) of aurihydrargyrumite. The bars below the diffraction profile are the simulation calculated using the atomic position by Lindahl [2]. All of the diffraction peaks can be indexed as aurihydrargyrumite.

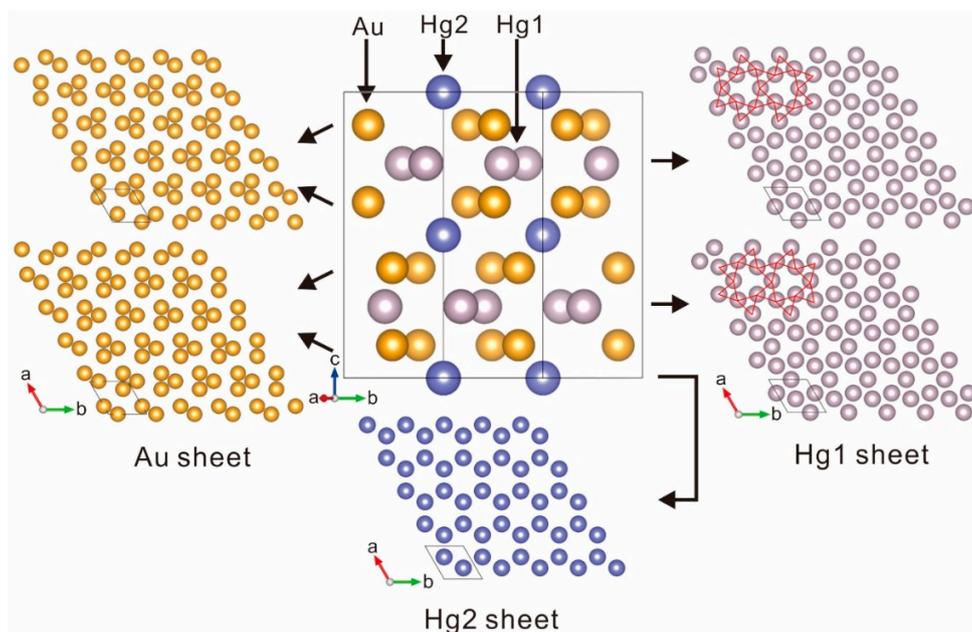


Figure 4. Crystal structure of aurihydrargyrumite.

Table 2. Powder X-ray diffraction data (CrK α radiation) for aurihydrargyrumite.

| Aurihydrargyrumite | | | Synthetic Au ₆ Hg ₅ [2] | | |
|--------------------|------------------------------|-------------------------------|---|-------------------------------|---------------------------|
| <i>hkl</i> | <i>d</i> _{obs.} (Å) | <i>d</i> _{calc.} (Å) | <i>I</i> / <i>I</i> ₀ | <i>d</i> _{calc.} (Å) | <i>I</i> _{calc.} |
| 100 | | 6.059 | | 6.057 | 4 |
| 002 | | 5.077 | | 5.074 | <1 |
| 102 | | 3.891 | | 3.889 | <1 |
| 110 | 3.502 | 3.498 | 1 | 3.497 | 5 |
| 111 | 3.309 | 3.307 | 1 | 3.306 | 2 |
| 200 | 3.025 | 3.029 | 2 | 3.028 | 4 |
| 112 | 2.877 | 2.881 | 29 | 2.879 | 52 |

Table 2. Cont.

| <i>hkl</i> | Aurihydrargyrumite | | <i>I/I</i> ₀ | Synthetic Au ₆ Hg ₅ [2] | |
|------------|------------------------------|-------------------------------|-------------------------|---|---------------------------|
| | <i>d</i> _{obs.} (Å) | <i>d</i> _{calc.} (Å) | | <i>d</i> _{calc.} (Å) | <i>I</i> _{calc.} |
| 202 | 2.597 | 2.602 | 23 | 2.600 | 49 |
| 004 | | 2.539 | | 2.537 | <1 |
| 113 | 2.434 | 2.432 | 42 | 2.431 | 100 |
| 104 | 2.337 | 2.341 | 100 | 2.340 | 46 |
| 210 | 2.290 | 2.290 | 13 | 2.289 | 38 |
| 211 | 2.234 | 2.234 | 87 | 2.233 | 91 |
| 212 | 2.087 | 2.088 | 3 | 2.087 | 5 |
| 114 | 2.053 | 2.055 | 22 | 2.053 | 12 |
| 300 | 2.019 | 2.020 | 15 | 2.019 | 21 |
| 204 | 1.947 | 1.946 | 13 | 1.945 | 14 |
| 213 | 1.895 | 1.897 | 21 | 1.896 | 18 |
| 302 | 1.876 | 1.877 | 2 | 1.876 | 2 |
| 115 | 1.758 | 1.756 | 13 | 1.755 | 13 |
| 220 | | 1.749 | | 1.748 | <1 |
| 221 | | 1.724 | | 1.723 | 3 |
| 214 | | 1.700 | | 1.700 | 1 |
| 006 | | 1.692 | | 1.691 | 4 |
| 310 | | 1.680 | | 1.680 | <1 |
| 311 | 1.658 | 1.658 | 8 | 1.657 | 10 |
| 222 | | 1.654 | | 1.653 | 2 |
| 106 | | 1.630 | | 1.629 | 3 |
| 312 | | 1.595 | | 1.595 | 1 |
| 304 | 1.581 | 1.580 | 5 | 1.580 | 4 |
| 223 | | 1.554 | | 1.553 | <1 |
| 116 | 1.525 | 1.523 | 23 | 1.523 | 18 |
| 215 | | 1.519 | | 1.519 | <1 |
| 400 | 1.516 | 1.515 | 8 | 1.514 | 12 |
| 313 | 1.506 | 1.505 | 15 | 1.505 | 15 |
| 206 | 1.478 | 1.477 | 5 | 1.477 | 3 |
| 402 | | 1.452 | | 1.451 | <1 |
| 224 | 1.441 | 1.440 | 11 | 1.440 | 18 |
| 314 | 1.401 | 1.401 | 39 | 1.401 | 31 |
| 320 | | 1.390 | | 1.390 | 2 |
| 321 | 1.378 | 1.377 | 7 | 1.377 | 8 |
| 216 | | 1.361 | | 1.360 | 1 |
| 322 | 1.341 | 1.341 | 16 | 1.340 | 16 |
| 117 | | 1.340 | | 1.339 | 4 |
| 225 | | 1.325 | | 1.325 | 1 |
| 410 | 1.321 | 1.322 | 16 | 1.322 | 18 |
| 411 | | 0.131 | | 1.311 | 1 |
| 404 | 1.301 | 1.301 | 41 | 1.300 | 49 |
| 306 | | 1.297 | | 1.296 | <1 |
| 315 | 1.294 | 1.295 | 11 | 1.294 | 14 |
| 323 | 1.286 | 1.286 | 29 | 1.285 | 29 |
| 412 | 1.280 | 1.280 | 18 | 1.279 | 21 |
| 008 | 1.269 | 1.269 | 15 | 1.269 | 18 |
| 108 | | 1.242 | | 1.242 | <1 |
| 413 | | 1.232 | | 1.231 | 3 |
| 217 | 1.225 | 1.225 | 65 | 1.225 | 89 |
| 324 | 1.219 | 1.219 | 15 | 1.219 | 13 |
| 226 | 1.216 | 1.216 | 14 | 1.216 | 11 |
| 500 | 1.211 | 1.212 | 11 | 1.211 | 19 |
| 118 | | 1.193 | | 1.192 | 1 |

Figure 4 shows a crystal structure of aurihydrargyrumite based on the result of Lindahl [2]. Aurihydrargyrumite is identical to the synthetic Au₆Hg₅ phase.

6. Relation to Other Species

Minerals and synthetic amalgam phases are summarized in Table 3. In the Au–Hg system [1], three amalgam phases were observed: Au₄Hg, Au₃Hg, and Au₂Hg. Above the amalgam phases, the structures have been solved for Au₃Hg [12]. Hexagonal symmetry was reported in Au₄Hg and Au₂Hg phases, while their structures are unsolved [12]. Additionally, Lindahl [2] reported a metastable phase with an Au₆Hg₅ composition that was synthesized by reducing ions in a solution using hydrazine sulfate and electrolytic refining while using a gold anode and a mercury cathode. The Au₆Hg₅ phase is stable at around room temperature, while it alters by leaching the mercury component at above 70 °C [2].

Among natural amalgams, the unit-cell parameters are known for aurihydrargyrumite, weishanite, and UM192-08-EAuHg. In this study, Aurihydrargyrumite is equivalent to the synthetic Au₆Hg₅ phase. Although the composition of weishanite is (Au,Ag)₃Hg₂, the symmetry and unit-cell parameters can be indexed as the synthetic Au₃Hg phase [5]. There are no synthetic phases corresponding to UM192-08-EAuHg [6].

Table 3. Comparable data for minerals and synthetic phases in Au–Hg system.

| Minerals | | | | | | | | | | |
|--------------------|--|--------------------------------------|--------------|--------------|--------------|-------|-------|-------|----------------------------|------------|
| Name | Composition | Symmetry | <i>a</i> (Å) | <i>b</i> (Å) | <i>c</i> (Å) | α (°) | β (°) | γ (°) | <i>V</i> (Å ³) | Reference |
| Aurihydrargyrumite | Au ₆ Hg ₅ | <i>P</i> 6 ₃ / <i>mcm</i> | 6.996 | = <i>a</i> | 10.154 | 90 | 90 | 120 | 430.4 | This study |
| Weishanite | (Au,Ag) ₃ Hg ₂ | <i>P</i> 6 ₃ / <i>mmc</i> | 2.9265 | = <i>a</i> | 4.8176 | 90 | 90 | 120 | 35.7 | [5] |
| UM1992-08-E:AuHg | Au _{94.88} Hg ₆₋₁₂ | Monoclinic | 4.729 | 5.243 | 4.546 | 90 | 90.9 | 90 | 112.7 | [6] |
| Synthetic Phases | | | | | | | | | | |
| Name | Composition | Symmetry | <i>a</i> (Å) | <i>b</i> (Å) | <i>c</i> (Å) | α (°) | β (°) | γ (°) | <i>V</i> (Å ³) | Reference |
| - | Au ₆ Hg ₅ | <i>P</i> 6 ₃ / <i>mcm</i> | 6.9937 | = <i>a</i> | 10.148 | 90 | 90 | 120 | 430.4 | [2] |
| - | Au ₄ Hg | Hexagonal | 8.736 | = <i>a</i> | 9.577 | 90 | 90 | 120 | 633 | [12] |
| - | Au ₃ Hg | <i>P</i> 6 ₃ / <i>mmc</i> | 2.918 | = <i>a</i> | 4.8113 | 90 | 90 | 120 | 35.7 | [12] |
| - | Au ₂ Hg | Hexagonal | 13.98 | = <i>a</i> | 17.2 | 90 | 90 | 120 | 2911.2 | [12] |

7. Discussion

The Au₆Hg₅ phase (=aurihydrargyrumite) could not be formed by the direct reaction between gold and mercury [1], and the special synthetic methods [2] cannot be applied directly to the natural environment. However, aurihydrargyrumite certainly occurs in nature, as demonstrated in this study. The formation of this mineral is discussed in the following.

Aurihydrargyrumite occurs as a thin layer on the outermost surface of the placer gold. This texture is clearly of secondary origin, since the thin surface layer would probably peel off if the placer gold were transported. Therefore, aurihydrargyrumite likely occurred after the placer gold was stably deposited. The accretion of a mercury component from the river water is unlikely, since no mercury was detected in the Oda River in the official water quality survey that was conducted by the Ehime prefectural office. Aurihydrargyrumite is probably formed from an Hg component already present in the placer gold; indeed, the gold grain includes a mercury component in the core. In the synthetic experiment, the Au₆Hg₅ phase (aurihydrargyrumite) was formed by reduction from ions and by electrolytic synthesis. Therefore, in nature, ionization and precipitation may interact in a complex manner on the surface of the placer gold to form aurihydrargyrumite. When considering the situation, self-electrorefining is probably involved in the migration of the gold and mercury components to the surface layer, which is analogous to the process of forming a gold-rich rim in the placer gold [13]. We suggest that the placer gold dissolves at the interface, and that subsequently the gold and mercury components immediately precipitate back onto the surface. The gold and mercury component that was concentrated through this process first formed the gold-rich zone, and aurihydrargyrumite was subsequently crystallized by further self-electrorefining based on the interface of the gold-rich zone. In this way, aurihydrargyrumite probably occurs in the process of weathering of the mercury-bearing placer gold.

These formation processes were also suggested to explain the zoned texture of anthropogenic amalgam grains from the Tulameen–Similkameen river system, Canada [7]. The zoning texture in amalgam phases was also reported in placer gold from the Palakharya River, Bulgaria [3,4], and Inagli Pt–Au placer deposit, Russia [8]. In natural weathering environments, the gold and mercury components in placer gold seem to concentrate on the surface by repeated leaching and deposition. However, the mechanism of leaching and deposition is still controversial, and various mechanisms might be closely involved. Although self-electrorefining is one of the most probable mechanisms, there is no evidence to discount other mechanisms, such as the involvement of microbially assisted reactions in nugget and layer alloy formations. Ionization and re-metallization of gold and mercury components must be a contributory factor in the formation of aurihydrargyrumite.

8. Conclusions

Aurihydrargyrumite is found in placer gold from Iyoki, Uchiko, Ehime Prefecture, Japan. The chemical composition and X-ray diffraction profile confirmed that aurihydrargyrumite and the Au₆Hg₅ phase were identical. Aurihydrargyrumite occurs as thin layers on the outermost surface of the placer gold and its formation relates to the natural weathering environment, which involves ionization and re-metallization of gold and mercury components in the placer gold.

Author Contributions: D.N.-H. wrote the paper. D.N.-H., T.T. and T.M. took part in the field works. T.M. found the first specimen, and subsequently T.T. and D.N.-H. collected the placer samples. D.N.-H. performed the observation, chemical analysis, and XRD study.

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Conflicts of Interest: The authors declare no conflict of interest.

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