A SODIAN SULFATIAN FLUORAPATITE FROM AN EPITHERMAL CALCITE–QUARTZ VEIN OF THE KUSHIKINO MINE, KAGOSHIMA PREFECTURE, JAPAN

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ABSTRACT

A sodian-sulfatian fluorapatite, a new variety of apatite, was found in sulfide concentrates of gold–silver–tellurium ores from the Shinpi No. 1 epithermal-type calcite–quartz vein of the Kushikino mine, Kagoshima Prefecture, Japan. This paper is the first detailed report on the occurrence of this mineral in sulfide concentrates of gold–silver–tellurium ores from the Shinpi No. 1 epithermal-type calcite–quartz vein of the Kushikino mine, Kagoshima Prefecture, Japan. The naturally occurring sulfate and sulfatian apatite from this type of ore deposit. The mineral is closely associated with quartz, interstratified chlorite-smectite, and ore minerals. The chemical composition of the mineral was determined by electron microprobe: CaO 51.51–54.30, Na2O 0.13–2.26, K2O 0.00–0.02, MgO 0.00–0.02, MnO 0.03–0.10, FeO 0.01–0.10, P2O5 35.09–40.82, SO3 0.40–7.39, SiO2 0.05–0.66, Cl 0.00–0.01, F 3.03–3.62 wt.%. The (Na + Si) : S atomic ratio is close to one. The charge increase involved by the substitution of (PO)3− by (SO)3− is considered to be electrostatically compensated for by the concomitant substitutions of Na+ for Ca2+ and, although minor, of (SiO/3− for (PO)3−. The general formula, disregarding trace to minor constituents, is expressed as follows: Ca5−xNa2(PO3−xS)xO12(PO4x−yFy)(OH1−y), where x ranges from 0.02 to 0.41, and y, from 0.80 to 0.96. The naturally occurring sulfate and sulfatian apatites should be divided into two series, apatite–ellestadite and apatite–cesanite, on the basis of mode of substitution. These minerals form under high late(O), and these species of sulfate and sulfatian apatite are known to occur in nature; these are ellestadite, hydroxyllesladite, "wilkeite" (now discredited by the IMA), and cesanite, in whose structures (SO/3− substitutes for (PO)3−. Ellesladite, a sulfate silicate apatite (OH > Cl > F in atomic percent), occurs in a contact zone between crystalline limestone and granodiorite in Crestmore, California (McConnell 1937, Rouse & Dunn 1982). Hydroxyllesladite, an OH analogue of ellesladite, was reported from a skarn zone of the Chichibu pyrometasomatic iron ore deposit, Saitama Prefecture, Japan (Harada et al. 1971). "Wilkeite", a sulfate- and silicate-bearing fluorapatite, occurs in Crestmore (Eakle & Rogers 1914, McConnell 1937, Rouse & Dunn 1982). "Wilkeite" having Cl > OH > F (in atomic percent) was reported from the Chichibu mine (Harada et al. 1971), and "wilkeite" having OH >> F + Cl (in atomic percent) occurs in a calcite vein in skarn in crystalline limestone, Hiroshima Prefecture, Japan (Kusachi et al. 1982). Cesanite, a sodium sulfate hydroxylapa-
tite, occurs in the Cesano geothermal field, Italy (Cavarreta et al. 1981).

During the course of a mineralogical study of gold–silver ores from epithermal-type calcite–quartz veins of the Kushikino mine, Kagoshima Prefecture, Japan, the present authors found sodian-sulfatian fluorapatite, a new variety of apatite (Shiga et al. 1985). This is the first detailed report on the occurrence of sulfate and sulfatian apatite from this type of ore deposit. In this paper, the modes of occurrence and chemical compositions of the Kushikino apatite are described, and the nomenclature of the sulfate and sulfatian apatites and the physicochemical conditions of formation of the minerals are discussed.

MODE OF OCCURRENCE AND MINERAL ASSOCIATIONS

The Kushikino gold–silver mining area lies in the northern part of Kushikino city (Fig. 1). Regional exploration for gold and silver deposits has been carried out around the mining area by the Metal Mining Agency of Japan since 1978. The Kushikino gold–silver ore deposits are representative of epithermal-type calcite–quartz veins developed in andesite of Miocene age. K–Ar ages measured on adularia from the veins are 4.0±0.3 Ma (MITI 1979). The ore mineralogy and mineralization of the deposits have been studied by many investigators (e.g., Fukuyama 1954, Mukaiyama & Izawa 1964,

![Fig. 1. Map showing location of the Kushikino mine in Kagoshima Prefecture, Japan.](image-url)

Apatite occurs in the sulfide concentrates of the gold–silver–tellurium ores only from the 6th level of the Shinpi No. 1 vein, in spite of extensive examination of both types of ores. The vein, with a thickness of 2 or 3 metres, lies on the hanging-wall side of the Kunkikino No. 1 champion vein, and is composed mainly of quartz, calcite, adularia and interstratified chloride–smectite, as determined by X-ray powder-diffraction studies on untreated material. Two kinds of ores, of different texture and mineralogy, occur in the vein. One is a so-called “black band” ore (Fig. 2), in which sphalerite–galena–chalcopyrite intergrowths occur with minor to trace amounts of pyrite, tetrahedrite and tellurium-bearing minerals as black bands and films (less than 1 cm in thickness) running parallel to the boundaries between the vein and wall rock. The other is ore (Fig. 3) in which minerals such as sphalerite, chalcopyrite, clausthalite–galena solid solution, pyrite, and minor to trace amounts of tellurium-bearing minerals and electrum are disseminated in clay-rich parts composed chiefly of quartz and interstratified chloride–smectite. Apatite occurs abundantly as euhedral to subhedral crystals in close association with both
kinds of ores (Figs. 4, 5). In the black-band ores, fine- to medium-grained apatite crystals accumulate along the boundary between the quartz (adularia-sericite) zone and the black-band zone (Fig. 4–1), where the crystals are rarely in contact with adularia and sericite. The crystals are concentrated also within the black-band zone (Fig. 4–2). In other parts of the ores, the mineral often occurs as thin filmy aggregates parallel to the black bands, with or without accompanying ore minerals. On the other hand, in the quartz-clay ores, rather coarse grains of apatite, 200 × 200 × 400 µm, intimately associated with interstratified chlorite-smectite and ore minerals, are dispersed in a quartz matrix (Fig. 5). Homogenization temperatures measured on fluid inclusions in quartz and calcite associated with apatite range from approximately 200 to 250°C (unpubl. data).

Megascopically, aggregates of apatite are whiter in color and more turbid than quartz. In thin section, the apatite grains are short tabular prisms. The index of refraction n measured by the immersion method is 1.620(2), and the birefringence is very low. The X-ray powder-diffraction pattern, indexed on a hexagonal cell, gives a 9.375(2), c 6.884(2) Å (Shiga et al. 1985).

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CHEMICAL COMPOSITION OF THE APATITE

Chemical analyses of the Kushikino apatite were performed with an ARL-Shimadzu X-ray microanalyzer, model EMX-SM. Line-scan analyses on more than ten single crystals from different modes of occurrence revealed that most of them are markedly heterogeneous in distribution of constituents (Fig. 6), similar to apatite from other localities (e.g., Nedachi & Ueno 1981, Tsusue et al. 1981).

Instrumental conditions for microprobe analyses were: 15 kV excitation voltage, 2 × 10⁻⁸ A sample current on a natural apatite standard, 5 μm beam diameter, 20 s count times on samples and standards. The following materials were utilized as standards: apatite for Ca, P, F and Cl, anhydrite for S, albite for Na, biotite for Si, Fe, Mg and K, pyrosmalite for Mn. Data were reduced using the Bence & Albee method (1968). The results thus obtained are as follows: CaO 51.51-54.30, Na₂O 0.13-2.26, K₂O 0.00-0.02, MgO 0.00-0.02, MnO 0.03-0.10, FeO 0.01-0.10, P₂O₅ 35.09-40.82, SO₃ 0.40-7.39, SiO₂ 0.05-0.66, Cl 0.00-0.01, F 3.03-3.62 wt.% (Table 1). Structural formulae calculated from the analytical data are presented in Table 2.

In sulfur-bearing apatite, the S, Na and Si contents are of particular importance, as discussed later. The S and Na contents of this apatite show apparent negative correlations with the P and Ca contents, respectively (see the line-scan profiles, Fig. 6). The SiO₂ content, although generally low (less than 0.7 wt.%), does not show any distinct correlation with other constituents. The material is also characterized by a high but variable F-content and by a low Cl-content. No relationship between the F and Cl contents is apparent.

### Table 1. Chemical Composition of Apatite from the Shinpi No. 1 Vein of the Kushikino Mine

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<td>36.64</td>
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<td>37.49</td>
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<tr>
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<td>0.66</td>
<td>0.09</td>
<td>0.05</td>
<td>0.20</td>
<td>0.23</td>
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<td>3.56</td>
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<td>3.11</td>
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<td>H₂O**</td>
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<td>0.25</td>
<td>0.20</td>
<td>0.27</td>
<td>0.17</td>
<td>0.25</td>
<td>0.07</td>
<td>0.09</td>
<td>0.36</td>
<td>0.29</td>
<td>0.25</td>
<td>0.18</td>
<td>0.17</td>
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</table>

* Determined by electron microprobe (in wt. percent). * Total Fe as FeO. ** Calculated on the basis of Cl+F+OH=1.000 in the structural formula.
the charge increase is made up by the substitution material, disregarding trace and minor constituents, similar to that for cesanite. The general formula of the substitution of $\text{Na}^+$ for $\text{Ca}^{2+}$ (Cavarretta et al. 1981, Tazzoli 1983), also considered this mineral to be "one member of the apatite–ellestadite series". After that, Tazzoli (1983) also considered this mineral to be "one member of the apatite–ellestadite series".

Since the charge increase resulting from the substitution of $(\text{SO}_4)^{2-}$ for $(\text{PO}_4)^{3-}$ in the sulfate and sulfapatite is compensated by two clearly different schemes of substitution, $(\text{SiO}_4)^{4-}$ for $(\text{PO}_4)^{3-}$ and $\text{Na}^+$ for $\text{Ca}^{2+}$, it seems more appropriate to divide them into two series: 1) apatite–ellestadite $\text{Ca}_5\text{P}_2\text{O}_{12}(\text{F},\text{OH},\text{Cl})$ – $\text{Ca}_3\text{S}_3\text{Si}_3\text{O}_{12}(\text{F},\text{OH},\text{Cl})$ and 2) apatite – cesanite $\text{Ca}_5\text{P}_2\text{O}_{12}(\text{F},\text{OH},\text{Cl})$ – $\text{Ca}_3\text{Na}_2\text{S}_3\text{O}_{12}(\text{F},\text{OH},\text{Cl})$ series, not considering anions. The general formula of the first series is expressed as follows: $\text{Ca}_x\text{Na}_y(\text{P}_{3-x}\text{S}_x)\text{O}_{12}(\text{F},\text{OH},\text{Cl})$, where $x$ ranges from 0.02 to 0.41, and $y$ ranges from 0.80 to 0.96. Cavarretta et al. (1981) regarded cesanite as "the end member of the apatite–ellestadite series". After that, Tazzoli (1983) also considered this mineral to be "one member of the apatite–ellestadite series".

The general formula of the material, disregarding trace and minor constituents, is: $\text{Ca}_x\text{Na}_y(\text{P}_{3-x}\text{S}_x)\text{O}_{12}(\text{F},\text{OH},\text{Cl})$, where $x$ ranges from 0.02 to 0.41, and $y$ ranges from 0.80 to 0.96. Cavarretta et al. (1981) regarded cesanite as "the end member of the apatite–ellestadite series". After that, Tazzoli (1983) also considered this mineral to be "one member of the apatite–ellestadite series".

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**TABLE 2. STRUCTURAL FORMULAE OF APATITE OBTAINED FROM THE CHEMICAL COMPOSITIONS GIVEN IN TABLE 1**

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<td>Na</td>
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<td>0.247</td>
<td>0.163</td>
<td>0.359</td>
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<td>0.394</td>
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<tr>
<td>Mg</td>
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<td>0.001</td>
<td>0.001</td>
<td>0.001</td>
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<td>0.002</td>
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**TABLE 3. REVIEW OF CHEMICAL COMPOSITION OF NATURAL APATITE (IN WT. PERCENT)**

<table>
<thead>
<tr>
<th>Locality</th>
<th>Na$_2$O</th>
<th>SO$_3$</th>
<th>SiO$_2$</th>
<th>Cl</th>
<th>F</th>
<th>Reference</th>
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<tbody>
<tr>
<td>1. igneous rock syenite</td>
<td>Shonkin Sag, Montana</td>
<td>0.16-0.35</td>
<td>0.15-0.30</td>
<td>0.45-0.65</td>
<td>0.12-0.20</td>
<td>2.1-2.7</td>
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<tr>
<td>Skagerrak intrusion iron deposit</td>
<td>Shonkin Sag, Montana</td>
<td>0.14-0.34</td>
<td>0.15-0.22</td>
<td>0.56-0.61</td>
<td>0.11-0.23</td>
<td>3.1-3.3</td>
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<td>2. carbonate marble</td>
<td>Avnik, Turkey</td>
<td>0.00-0.21</td>
<td>0.10-0.22</td>
<td>0.13-0.42</td>
<td>0.00-0.10</td>
<td>2.00-4.22</td>
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<td>3. metamorphosed marble</td>
<td>Ontario, Canada</td>
<td>0.18</td>
<td>&lt;0.03</td>
<td>0.15</td>
<td>&lt;0.02</td>
<td>4.0</td>
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<td>4. hydrothermal ore deposit and hydrothermally altered skarned limestone</td>
<td>north-central Ceylon</td>
<td>-</td>
<td>-</td>
<td>2.31</td>
<td>1.05</td>
<td>Coorey 1970</td>
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<td>5. sedimentary rock</td>
<td>Kushikino mine, Japan</td>
<td>0.13-2.26</td>
<td>0.40-7.39</td>
<td>0.05-0.66</td>
<td>0.00-0.01</td>
<td>3.03-3.62</td>
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<td>Montana, Idaho etc.</td>
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<td>0.3-3.1</td>
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<td>-</td>
<td>3.8-4.2</td>
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* No data. ** Analyst: M. Nedachi.
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