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# **Cesium Deposits**

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## Introduction

Cesium or caesium? Cesium is the accepted spelling of element 55 by the American Ceramic Society (ACS). Caesium is the International Union of Pure and Applied Chemistry's (IUPAC) preferred spelling. Both are used interchangeably. Its chemical symbol is Cs. Cesium was discovered by Bunsen and Kirchhoff in 1860 through emission spectroscopy and in etymology finds its name in the Latin "Caesius" for the sky-blue lines in its emission spectrum.

It is a little known element, the bulk of which is used in oil and gas well servicing in the form of cesium formate. It is also used as catalysts in plastics, petroleum refining to remove sulphur, experimental Magneto Hydro Dynamic (MHD) power generation, in cesium clocks for telecommunications and GPS navigation, specialty glasses, opto-electronics, ion-propulsion motors, high-density alkaline batteries, and solar cells where it extends the electromagnetic spectrum. As a dense medium, cesium formate is also used in metallurgical test work, to separate DNA and, because of its density, it has a potential use in coal cleaning (Tuck 2020; Pickup et al. 2020). Cesium has one stable isotope, Cs<sup>133</sup> whereas <sup>137</sup>/<sub>55</sub> Cs or radiocesium is an artificially produced isotope used in cancer treatment. It has a potential application in the sequestration nuclear waste (Cerný 1978; Yoshinobu et al. 2014). It has no known natural bioavailability and is used in the food preparation industry.

Cesium has chemical and physical properties similar to the other Group I elements and has a melting point of 28.4 °C. It is explosively pyrophoric when added to water. Of the alkali metals, cesium has the lowest crustal abundance at 0.3 ppm. In contrast, rubidium has a crustal abundance of 9 ppm and lithium has a crustal abundance of 2 ppm (Chemical Rubber Company Handbook 2016-2017). Twenty-two minerals are known to host cesium (Webmineral.com 2019), and the present authors propose to add another mineral, Cs-pharmacosiderite.

Primarily, cesium has been recovered from the minerals pollucite and lepidolite hosted in "giant" pegmatites such as Tanco in central Canada, Bikita in Zimbabwe, and the Yichun granite in China (Fig. 1). These sources are now problematic; production at Tanco ceased due to safety issues related to the fall of loose rock and Bikita by exhaustion of stockpiles. The small Sinclair deposit in Australia was mined out in 2019, and several small cesium deposits have been identified in Africa, the U.S. and Canada but none have reached systematic production.

A second type of cesium deposit is formed through greisening. The Yichun rare metal granite in China is an example of Li – Cs greisening formed by hydrothermal alteration of precursor composite intrusives.



Figure 1. World map showing the location of described or mentioned cesium-bearing deposits.

A third type of cesium deposit is found in the form of epithermal geyserites (hot spring environment) and several have been identified including the Targejia deposit in Tibet and the Taron deposit in Argentina, and although of lower cesium grades and widely varying mineral assemblages may be equal or larger in their contained cesium than the large pegmatite sources. In Yellowstone National Park in the northwest USA, Li-Cs greisening is ongoing and Li – Cs assemblages are forming at very low temperatures and pressures in various sedimentary rocks and rhyolites. The authors consider Targejia and Yellowstone as active analogues to the Taron deposit (see description of this deposit in second article in this issue). Cesium brines are reported to occur in Tibet and China by Hu *et al.* (1988), in India in the Puga Valley by Chowdry *et al.* (1974) and at Amarillo (Mina Antuco) in Argentina by Richards (pers. comm. 2020).

The source of Cs-bearing pegmatites is probably from fractionation of S-type granites (Cerný *et al.* 2012). The source of Cs-bearing geyserites is probably similar to that at Yellowstone in which large volumes of rhyolite over the Yellowstone mantle plume are being leached by circulating epithermal fluids.

Cesium is now on the U.S., Canadian and several other government's list of critical elements and there is a global burgeoning of exploration for Cs-bearing pegmatites. At present, Albemarle Corp. and Sinomine Specialty Fluids (formerly Cabot Specialty Fluids) in China control the cesium refining industry and supply chain. At present there is no mine production of cesium. This literature review was undertaken to characterize the importance of geology, geochemistry, mineralogy, and geometallurgy of these types of cesium deposits and offer some insights as to the origins of such novel deposits.

#### The Tanco Pegmatite, Canada

"Nothing on earth is more refined than a pollucite-bearing pegmatite" (London 2019). The Archean Tanco pegmatite was discovered by prospector Jack Nutt when exploring for tin at Bernic Lake in Manitoba, central Canada (Fig. 1), in the late 1920s. The tin was apparently identified from blowpipe testing of mineralization in a separate and smaller pegmatite dyke from the larger Tanco pegmatite. The geology of the deposit is shown in Figure 2.

The Tanco pegmatite was emplaced in an essentially horizontal position (Brisbin 1986) and shows fractional crystallization (Brown *et al.* 2017) culminating in upwardly disposed zones of lithium phosphates, spodumene, lepidolite, pollucite, and quartz. A north – south cross section fence diagram is shown in Figure 2, after Stillings *et al.* (2006) and the mineralogy of the zoning in shown in Table 1 after Cerný (2005). The constituent mineralogy of the Tanco pegmatite is



*Figure 2.* North – south cross section through the Tanco pegmatite showing internal zoning, from Stillings et al. (2006). Note vertical exaggeration.

provided in Table 1 which shows the nine major mineralogical zones that fractionally crystallized in the temporal sequence of (1) to (9). Cesium is found as constituents of two zones, (5) and (8). In zone (8) it is the dominant mineral. The pollucite zone was discovered while driving a decline for access to the tantalum orebody.

A description of the pollucite zone by Cerný and Simpson (1978) is as follows: "Pollucite forms pods and nodules from 1 to 2 m in diameter and several large lenticular bodies.....the largest of these, 180 x 75 x 12 m in size, is located in the eastern flank of the pegmatite, and three smaller bodies are found in the western part......masses of pollucite show simple smooth boundaries against quartz, and are anhedral with respect to all other primary constituents of zone (5) where pollucite may locally cement fractured or brecciated silicate assemblages but it is never observed as a replacement".

The Tanco deposit underwent several stages of low temperature hydrothermal alteration. Three consecutive alteration stages may be distinguished: (i) coarse polygonal veining by microcline, albite, quartz and lepidolite; (ii) fine braided veining which carries spodumene, lithian muscovite and adularia, and (iii) replacement by kaolinite and montmorillonite (+/- quartz, calcite) but it is considered a closed system and the alteration phases derived internally. The authors consider this as endo-greisening and Cerný's "nauchshub" or "resupply" in the Czech language. The preproduction pollucite resource at Tanco was approximately 46,000 tonnes of contained Cs<sub>2</sub>O at 12.8% Cs<sub>2</sub>O (Tetratech 2011).

Zone	Main constituents	Characteristic subordinate (accessory) & rare minerals
Exomorphic unit	Biotite, tourmaline, holmquistite	Arsenopyrite
(1) Border zone	Albite, quartz	Tourmaline, apatite, (biotite), beryl, triphylite
(2) Wall zone	Albite, quartz	Beryl, (tourmaline), muscovite, Li-muscovite, microcline- perthite
(3) Aplitic albite zone	Albite, quartz, (muscovite)	Muscovite, Ta-oxides, beryl, (apatite), tourmaline, cassiterite, ilmenite, zircon, sulphides
(4) Lower intermediate zone	Microcline-perthite, quartz, spodumene, amblygonite	Li-muscovite, lithiophyllite, lepidolite, petalite, Ta-oxides
(5) Upper intermediate zone	Spodumene, quartz, amblygonite	Microcline-perthite, pollucite <sup>(1)</sup> , lithiophyllite, (albite, Li- muscovite), petalite, eucryptite, Ta-oxides
(6) Central intermediate zone	Microcline-perthite, quartz, albite, muscovite	Beryl, (Ta-oxides), zircon, ilmenite, spodumene, sulphides, lithiophyllite, apatite, cassiterite
(7) Quartz zone	Quartz	Spodumene, amblygonite
(8) Pollucite zone	Pollucite	Quartz, spodumene, petalite, muscovite, lepidolite, albite, microcline, apatite
(9) Lepidolite zone	Li-muscovite, lepidolite, microcline-perthite	Albite, quartz, beryl, (Ta-oxides, cassiterite), zircon

Table 1. Mineralogy and zoning of the Tanco Pegmatite after Cerny (2005)

<sup>(1)</sup> Red script denotes zeolites in this and subsequent tables

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#### The Yichun Granite, Jiangxi Province, China

The Mesozoic Yichun granite is located in Jiangxi Province, China (Fig. 1). The granite has been mined for Ta-Nb and Li-Cs and byproduct clay minerals. It is a composite intrusion consisting of 3 units: a protolithionite-muscovite granite, a Li-mica granite, a topaz lepidolite granite and later pegmatites (Yin *et al.* 1995; Huang *et al.* 2002) and was intruded into Neoproterozoic mica schists (Fig. 3).



Figure 3. Bedrock geology map of the Yichun deposit, after Li et al. (2017).

Of interest is the topaz – lepidolite granite unit which was the latest phase in the intrusive sequence and, as described by Wang *et al.* (2004), has an albite-rich facies containing pollucite and a Cs-dominant analogue of polylithionite that assays 1709 ppm Cs. Wang *et al.* (2004) also described lepidolite with Cs-rich zones that contain up to 25.8% Cs<sub>2</sub>O and suggested this lepidolite may be a Cs-dominant analogue of polylithionite. Yin *et al.* (1995) commented that occurrence of a cesian-lepidolite implies the presence of a Cs-rich late magmatic fluid.

Wang commented further that "according to their distribution and compositional characteristics, pollucite and the Cs-dominant analogue of polylithionite seem to have formed at the late-magmatic or magmatic to hydrothermal transition stage of evolution of the leucocratic magma" and interpret the system at Yichun in its final stages to be open and deriving fluids for the greisening from the surrounding mica schist. The authors of this EXPLORE article consider this an example of exo-greisening. The grade of Cs<sub>2</sub>O in the topaz lepidolite granite at Yichun is 0.18% Cs<sub>2</sub>O in an unspecified tonnage (Wang *et al.* 2004).

## Yellowstone Geyserites, Wyoming

The Yellowstone Plateau in northwest Wyoming, USA, consists of Pliocene and Pleistocene rhyolitic pyroclastic rocks and lesser flow rocks. The areal extent of these rocks at Yellowstone is estimated to be some 2900 km<sup>2</sup> (Abedini *et al.* 2007) and with its collapsed caldera is estimated to incorporate some 3700 km<sup>3</sup> (Christiansen 2001). Subjacent and intercalated with these are basaltic rocks and underlying older rocks are Precambrian crystalline basement, Phanerozoic carbonates, sandstones, redbeds, mudstone, coal, and shales including oil bearing traps.

Between 1967 and 1969, the United States Geological Survey undertook the drilling of 13 holes in Yellowstone Park

(White *et al.* 1975). The location of drill holes Y-1,-2,-3,-5 in the Lower and Upper Geyser Basins are shown in Figure 4. These holes were selected for their mineral occurrences listed in Table 2. Of interest is lepidolite and analcime forming in Drill Hole Y-3 at depths of 23.8 to 28.5 m at a temperature of 130° to 140° C, and a presumed pressure of 245 kPa uncorrected for density or site elevation.

The rocks hosting the analcime are described by Keith *et al.* (1983) as "Cs is selectively concentrated in analcime" and "the Cs content of analcime-bearing altered sedimentary rocks is as high as 3000 ppm and altered rhyolite lava flows....now contain up to 250 ppm Cs". Keith *et al.* (1983) also describes concentrates, presumably handpicked, of Yellowstone analcime as containing as much as 4700 ppm Cs. The present authors consider the lepidolite an exo-greisen mica and as having been derived from the rhyolites.

The Yellowstone area does not have a quantified Cs resource. It could have a significant value given the volumes of the underlying rhyolites being leached. There are no Cs brine resources known.



Table 2. Minerals present in Drill Holes Y-1,-2,-3,-5 at Yellowstone,Wyoming, after Bargar et al. (1973) and Bargar et al. (1987)

Lepidolite	K(Li,Al,Rb) <sub>2</sub> (Al,Si) <sub>4</sub> O <sub>10</sub> (F,OH) <sub>2</sub>
Quartz	SiO <sub>2</sub>
Analcime	(Na,Cs)AlSi <sub>2</sub> O <sub>6</sub> •H <sub>2</sub> O
Pyrite	FeS <sub>2</sub>
Calcite	CaCO₃
Fluorite	CaF <sub>2</sub>
Chalcedony	SiO <sub>2</sub>
Clinoptilolite	(Na,K,Ca) <sub>2-3</sub> Al <sub>3</sub> (Al,Si) <sub>2</sub> Si <sub>13</sub> O <sub>36</sub> .12H <sub>2</sub> O
Darchiardite	K <sub>4</sub> (Si <sub>20</sub> Al <sub>4</sub> O <sub>48</sub> )•13H <sub>2</sub> O
Pectolite	NaCa₂(Si₃Oଃ(OH)
Montmorillonite	(Na,Ca) <sub>0.33</sub> (Al,Mg) <sub>2</sub> (Si <sub>4</sub> O <sub>10</sub> )(OH) <sub>2</sub> • <i>n</i> H <sub>20</sub>
Yugawaralite	$CaAl_2Si_6O_{16}.4H_2O$
Mordenite	(Ca,Na <sub>2</sub> K <sub>2</sub> )Al <sub>2</sub> Si <sub>10</sub> O <sub>24</sub> 7H <sub>2</sub> O
Epistilbite	$CaAl_2Si_6O_{16}\bullet 5H_2O$
Albite?	NaAlSi₃O <sub>8</sub>
Truscottite	(Ca,Mn) <sub>14</sub> Si <sub>24</sub> O <sub>58</sub> (OH) <sub>8</sub> •2H <sub>2</sub> O
β-cristobalite	SiO <sub>2</sub>
α cristobalite	SiO <sub>2</sub>
Opal	SiO <sub>2</sub> • <sub>n</sub> H <sub>2</sub> O
Hyalite	SiO <sub>2</sub> • <sub>n</sub> H <sub>2</sub> O
Erionite	SiO <sub>2</sub> • <sub>n</sub> H <sub>2</sub> O
Kaolin	$AI_2(Si_2O_5)(OH)_4$
Illite	$K_{0.65}AI_{2.0}(AI_{0.65}Si_{3.35}O_{10})(OH)_2$
Celadonite	K(MgFe <sup>2+</sup> )Fe <sup>3+</sup> (Si <sub>4</sub> O <sub>10</sub> )(OH) <sub>2</sub>
Manganite	MnO(OH)
Cryptomelane	K(Mn <sup>4+</sup> 7Mn <sup>3+</sup> )O <sub>16</sub>
Pyrolusite	MnO <sub>2</sub>

Figure 4. Drill hole locations at Yellowstone Park, Wyoming, holes Y-1-8 and Y-13 located at the yellow dot, holes Y-9 and Y-12 at the red dot, and hole Y-11 at the blue dot (after Christiansen 2001).

# **Exploration for cesium**

Trueman (1978), in collaboration with Dr. Alexi Beus of the National Academy of Sciences, Moscow, developed lithogeochemical and gravitational methods for the exploration of granitic pegmatites. Subsequently Trueman & Cerný (1982) published more definitive work utilizing mineral chemistry and practical guidelines to map distribution within pegmatite fields and narrow down targets for rare metal exploration. These methods have been improved upon and now utilize soil, botanical, sediment, till and water geochemistry (Bradley et al. 2010; Steiner 2019). Galeschuk & Vanstone (2007) show examples of down hole Cs geochemical data for core samples. It should be cautioned that diamond drill greases are waterproof and accordingly may contain considerable lithium, a pathfinder element in exploration geochemistry, and used at the Tanco Mine.

Lithium, rubidium, and cesium usually are detected in geochemical surveys for granitic rocks; especially those that are highly fractionated. Surveys in active geyser or paleogeyser terrain responded significantly to cesium analysis, and also arsenic, manganese, and thallium. Arsenic and manganese are routinely detected in the percent ranges and thallium up to 0.74%; the highest thallium values known to the authors or colleagues.

In the course of writing this paper, it became apparent that the widely held cesium resource at the Tanco Mine (Cerný *et al.* 1996) of 317,515 tonnes @ 21.98% Cs in production turned out to be 12.82% Cs (Tetratech 2011). Also, according to reports by Pioneer Resources Ltd. (2020), the Sinclair Mine in Australia produced 10,000 tonnes of run of mine pollucite ore in 2018, and in 2019 were delivered with a shortfall of contained cesium.

# Geometallurgy of Cesium

Three principle methods of processing pollucite are by acid digestion, alkaline decomposition, or direct reduction (Ferguson & Gorrie 2011). At Tanco, the pollucite is digested in hot 35 - 45% sulphuric acid and a cesium alum is precipitated (Butterman *et al.* 2005). The alum is further digested to form cesium sulphate that is in turn reacted with formic acid to produce cesium formate and is the preferred commercial process for producing cesium (Tetratech 2011). This process is not dissimilar to the metallurgy developed for the Taron deposit in that it employs a hot sulphuric acid digestion, precipitation, and redigestion to yield cesium hydroxide; a saleable commodity and a starting point for other cesium compounds (Mokmeli *et al.* 2017, 2018). A second stage of testing led to SO<sub>2</sub> sparging with a significant reduction in acid requirements. An alkaline leach process has also been developed at the University of British Columbia, Canada. According to Butterman *et al.* (2005) in an alkaline decomposition process, pollucite is roasted with a CaCO<sub>3</sub>– CaCl<sub>2</sub> mixture or a Na<sub>2</sub>CO<sub>3</sub>–NaCl mixture, then leached with water or dilute ammonia to extract a dilute CsCl solution and converting the chloride to cesium alum.

Direct reduction to produce cesium metal involves heating pollucite with calcium, potassium or sodium metal in a vacuum or an inert atmosphere (Butterman *et al.* 2005). Hu *et al.* (1988) reported a 98% extraction of cesium from the Yichun deposit using Solvent Extraction (SX). Arnold *et al.* (1965) reported a 99% extraction of cesium from pollucite of an unspecified origin with SX, and the Cs brines in Tibet and China are reported as targets for SX by Hu *et al.* (1988).

#### **Discussion and Conclusions**

According to Cerný & Ercit (2005), pegmatitic granites and cesium-bearing pegmatitic granites were probably derived from underlying S-type granites. Rare metal granites such as the Yichun are probably products of continuing fractionation (Huang *et al.* 2002) of earlier granitic phases. According to Keith *et al.* (1983), the cesium source rocks at Yellowstone are the underlying rhyolites which are being leached by epithermal fluids and at Taron, the source rocks were probably Miocene rhyolites (Richards 2005).

In the described deposits, crystallization has proceeded through hydrothermal phases with zeolite mineralization and then deuteric alteration. Pollucite and lepidolite formed at Tanco and Yichun, lepidolite and analcime at Yellowstone, and those ultimately broke down to clays as shown in Figure 5.

The presence of microbes have been well documented at Targejia in active geysers in cesian opal in Tibet (Zhao *et al.* 2008; Wang *et al.* 2012; Zhang 2014). Microbial activity is abundant at Yellowstone at shallow depths and Bargar *et al.* (1985) described what they interpreted as micrometer sized rod-like, threadlike and irregular microorganisms in liquid rich fluid inclusions in quartz from drill holes. However, they had difficulty reconciling a PT regime of 190°C and 1.24 kPa in which the microbes were present.

Cesium is a little known rare metal and there is little cesium research being conducted, other than for the



Figure 5. Pressure temperature data from Teertstra & Cerny (1995) and online at <u>http://www.iza-online.org/natural/Datasheets/Pollucite/</u> Pollucite.html radioactive Cs isotopes and their sequestration. The authors welcome any new advances in the understanding of the geology and geochemistry of cesium together with research and development in, for example, a cesium battery which could be expected to have a higher power density (amount of power per unit volume) than lithium contemporaries. As with many elements,

uses will be found if they are available.

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