



# Metamict fergusonite-(Y) in a spessartine-bearing granitic pegmatite from Adamello, Italy

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## ABSTRACT

A granitic pegmatite associated with the Monte Bruffione granodiorite in the Tertiary Adamello Massif, Italy, consists primarily of albite, potassium feldspar, muscovite, quartz, and spessartine (graphic intergrowths with quartz). As accessory minerals, the pegmatite contains magnetite, pyrophanite, monazite-(Ce), uraninite, xenotime-(Y), zircon, and fergusonite-(Y). This yttrium niobate mineral contains inclusions of Th-rich uraninite, and is itself rich in UO<sub>2</sub> (average 7.1 ± 1.1 wt.%, *n* = 34) and ThO<sub>2</sub> (average 3.5 ± 1.0 wt.%). Many fergusonite crystals display growth zoning, characterized by a general increase towards the rim in the contents of Y and rare earth elements at the expense of U and Th. Irregular or patchy zoning as well as sector zoning are also observed in some of the crystals. Due to the alpha-decay of the U and Th fergusonite is metamict, as documented by transmission electron microscopy (TEM) and micro-Raman spectroscopy. Nevertheless, the mineral could be identified as fergusonite-(Y) on the basis of a canonical discriminant analysis of its chemical composition and on the basis of the close similarity of its Raman spectrum with that of a reference β-fergusonite. The crystalline-to-metamict transformation was associated with macroscopic swelling, as indicated by microfractures that are arranged radially around fergusonite inclusions in pyrophanite. The TEM data revealed that the amorphous fergusonite contains U-rich nanocrystals (5–15 nm across), which in most cases are distributed randomly and which probably nucleated after metamictization. The TEM investigations further revealed the ubiquitous presence of nano-sized (typically 5–25 nm across), nearly circular features, which exhibit a low diffraction contrast and which we interpret as nanopores. We propose that these nanopores represent former bubbles of radiogenic helium. The data presented here allowed us to determine that the critical amorphization dose of fergusonite is  $\leq 0.97 \times 10^{16}$  alphas/mg, i.e., lower than that of other actinide-rich oxide minerals (e.g., pyrochlore). The presence of the U-rich nanocrystals indicates that fergusonite is able to retain actinides even when it is entirely metamict and even if it is surrounded by microfractures, which represent potential fluid pathways. This result suggests that fergusonite could be a phase suitable to be included in ceramics designed for the immobilization of high-level nuclear waste.

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## 1. Introduction

Fergusonite, ideally YNbO<sub>4</sub>, was first reported by Haidinger (1826) as a new mineral species, associated with quartz from Kikertaursak, near Cape Farewell, South Greenland. The report is purely descriptive and based on crystal morphology. A chemical analysis of this material was given by Hartwell (1828), and with the assumption that the ZrO<sub>2</sub> reported is from an included Zr-phase, the analysis yields a stoichiometry of Y<sub>1.06</sub>Nb<sub>0.95</sub>O<sub>4</sub>, i.e., remarkably close to ideal YNbO<sub>4</sub>. Fergusonite occurs as an accessory mineral in granitic

pegmatites, mostly in rare earth element (REE)-enriched pegmatites (Ervanne, 2004; Ercit, 2005), and often in combination with one or more Y, Th, Nb, Ta, Ti oxide accessory minerals (Lumpkin, 1998). Among other reported occurrences, fergusonite has also been described as a hydrothermal mineral from the Pilansberg nepheline syenite (Olivo and Williams-Jones, 1999). Recently, fergusonite has been discovered in a porphyritic biotite granite of the Xihuashan complex, Southern China, where it occurs as discrete crystals and as inclusions in Y-bearing spessartine, in both cases associated with uraninite (Wang et al., 2003).

Two structures have been reported for fergusonite: a scheelite-type tetragonal fergusonite (α-fergusonite, *I*4<sub>1</sub>/*a*); and at high temperatures, a monoclinic polymorph (β-fergusonite, *I*2/*c*) (see recent review by Tomašić et al., 2006). Fergusonite can, however,

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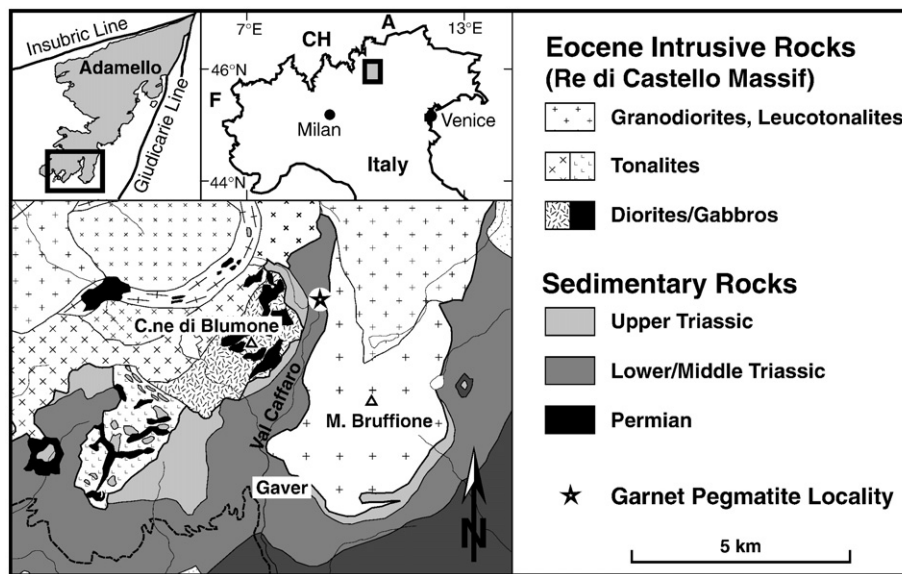


Fig. 1. Tectonic map of the southern Adamello region, Italy. Modified after Zhang et al. (2001).

contain significant quantities of U and Th, and thus is commonly metamict (Ervanne, 2004; Tomašić et al., 2006; Ruschel et al., 2007). Therefore, the structure is usually established only after heating the metamict mineral, provided that sufficient material is available. Structural characterization after annealing metamict minerals, however, can lead to a mis-identification; for example, in a systematic study of samarskite (Warner and Ewing, 1993), one of the 19 samarskite samples annealed recrystallized with a

fergusonite structure. Additionally, for crystals less than 100  $\mu\text{m}$  in size, this procedure is not technically feasible and therefore, identification requires to be founded on chemical composition. A recent study by Ercit (2005), based on statistical analyses of chemical compositions, provides the best opportunity to distinguish fergusonite from a range of (Y,REE,U,Th)-(Nb,Ta,Ti) oxide accessory minerals commonly associated with REE-enriched granitic pegmatites.

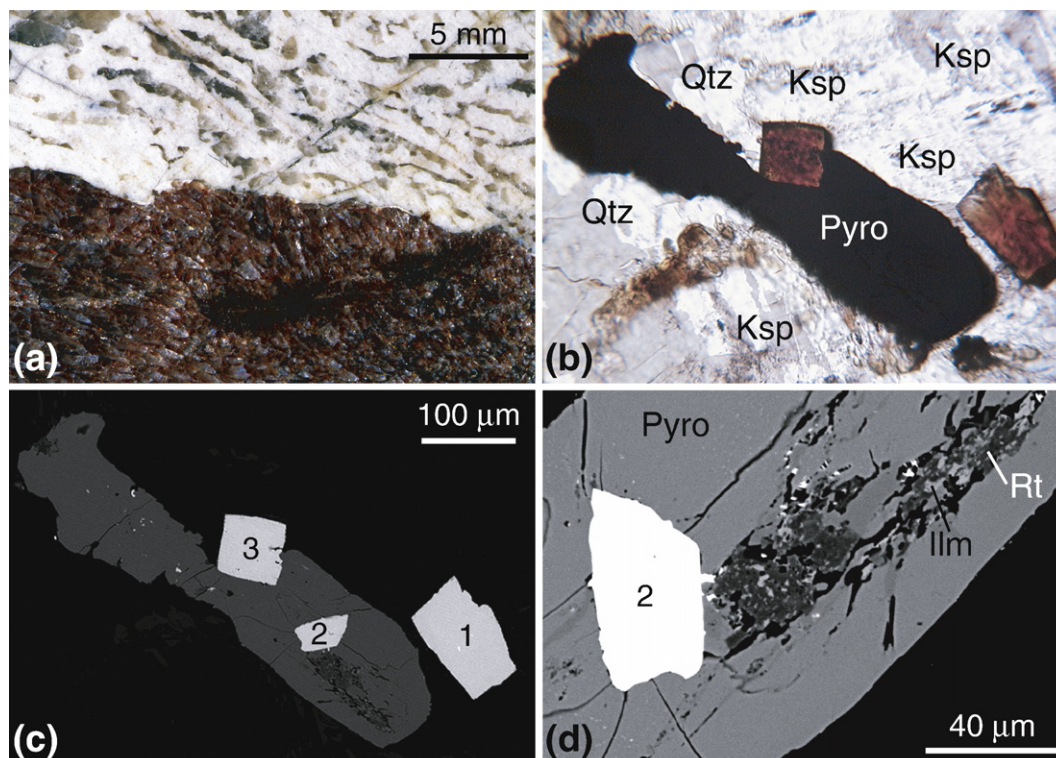


Fig. 2. Hand specimen and thin section images of the garnet-bearing granitic pegmatite from Val Caffaro, Adamello (Italy). (a) Polished slab of sample 269a. (b) Photomicrograph of Adamello thin section AU18B (plane polarized light), showing two brown fergusonite-(Y) grains (crystals #1 and #3) together with pyrophanite (opaque), potassium feldspar, and quartz. (c) BSE image of the area displayed in (b) revealing an additional fergusonite-(Y) grain (crystal #2) as well as many tiny fergusonite crystals (white blebs in left part of the pyrophanite crystal) as inclusions in pyrophanite. (d) Detail of (c) showing radial cracks in pyrophanite around fergusonite crystal #2. The small bright spots are fergusonite-(Y) inclusions in pyrophanite, whereas the fine-grained assemblage in the center of the image consists of rutile (darker than pyrophanite) and ilmenite (brighter than pyrophanite). Numbers refer to crystal numbers. Abbreviations: Ksp = potassium feldspar; Pyro = pyrophanite; Qtz = quartz; Rt = rutile; Ilm = ilmenite.

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