Morphological and chemical evolution of corundum (ruby and sapphire): Crystal ontogeny reconstructed by EMPA, LA-ICP-MS, and Cr\(^{3+}\) Raman mapping

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**ABSTRACT**

The term “ontogeny,” which is commonly used in biology, was introduced into the Earth sciences in 1961 to include the genesis and evolution of single crystals and crystal aggregates. The term encompasses nucleation, growth, alteration, and destruction. We present results of studies concerning the ontogeny of natural corundum (rubies and sapphires), and the chemical and morphological evolution of corundum crystals from deposits in Africa (Kenya, Tanzania, Madagascar) and Southeast Asia (Vietnam). Trace-element compositions indicative for different corundum habits were determined by rim-to-rim LA-ICP-MS and electron microprobe analyses. Raman spectroscopy was applied for Cr\(^{3+}\) photoluminescence mapping. Results traced the development of corundum crystals and the evolution of their chemistry and morphology, and helped to clarify the geological processes within particular deposits. These variations of corundum morphology are directly correlated with Cr and Fe contents and varying P-T conditions that prevailed during crystal growth. Dipyramidal habits combined with white color in corundum from two deposits in the Mangari area in Kenya have Cr concentrations of ~200–700 μg/g in crystals that grew under high P-T conditions. Prismatic habit of bright red ruby crystals was linked to Cr concentrations of ≥1500 μg/g in samples from Luc Yen (Vietnam) and Mangari (Kenya), formed under lower P-T. Concentrations of Cr between 700–1500 μg/g are associated with pink color and combinations of different habits (dipyramidal, prismatic, or dipyramidal-prismatic) in these samples. Contents of Fe ~700 μg/g and Cr ~1200 μg/g in sapphire crystals from the Morogoro area of Tanzania caused pink color that correlated with dipyramidal habit and elongation along the c axis. Rhombohedral habit and blue-violet color were observed at Cr ~600 μg/g and Fe ≥2000 μg/g in sapphires from Andranondambo in Madagascar, formed during the final stage of contact metamorphism.

**Keywords:** Corundum, ruby, sapphire, ontogeny, evolution, genesis, geochemistry, crystal morphology, Kenya, Tanzania, Madagascar, Vietnam

**INTRODUCTION**

“...for a given body of certain form, created according to the laws of Nature, there are evidences within the body disclosing the place and method of its creation” - Nicolai Stenonis, De solido... (1669)

Corundum, α-Al\(_2\)O\(_3\), is a common minor component of metamorphic rocks. Yet crystallization of its transparent varieties, ruby and sapphire, only occurs in a few rock types depleted in silica and enriched in alumina (Giuliani et al. 2007) in approximately 20 deposits worldwide (Hughes 1997). Some of the geological factors influencing corundum morphology have been studied by Hartman (1962, 1980), Popov (1984), and Sunagawa (2000). However the prime genetic question—how and why transparent corundum actually grows—is not fully understood.

“Ontogeny” is used in biology to describe the developmental history of an organism within its own lifetime. The term was introduced into the Earth sciences in 1961 by the Soviet mineralogist D.P. Grigor’ev to relate the genesis and evolution of single crystals and crystal aggregates, specifically including their nucleation, growth, alteration, and destruction stages (Grigor’ev 1961, 1965). In the decades after Grigor’ev’s publication, many researchers have worked in the field of mineral ontogeny (e.g., Yushkin 1977, 1985; Zabin 1979; Pirogov 1985; Pavlishin et al. 1988; Prieto et al. 1992; Self and Hill 2003; Sorokina 2011; Alekseev and Marin 2012; Sorokina et al. 2012). Evolutionary direction in the proliferation of mineral species has been proposed and discussed by Hazen et al. (2008) and Krivovichev (2013).

This article presents results on the evolution of morphology and changing geochemistry of opaque and transparent corundum crystals during growth and alteration for samples from five important deposits in Africa (Mangari area in Kenya, Morogoro in Tanzania, and Andranondambo in Madagascar) and Southeast Asia (Luc Yen in Vietnam). Rim-to-rim chemical analyses were combined with Cr\(^{3+}\) photoluminescence mapping for corundum with visual color zoning from Mangari. This allowed in situ observations of the distribution of Cr\(^{3+}\) content within the mineral matrix.

In the past, rim-to-rim ion probe analyses on Scottish sapphires were combined with cathodoluminescence in a study