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**An accessory mineral and experimental perspective on the evolution of the early crust**

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**ABSTRACT**

As the only known mineral with confirmed ages >4 Ga, zircon is unmatched in the field of early Earth research. In the past two decades, researchers have continued to establish connections between zircon chemistry and the physical/chemical processes that shaped the early crust. This connection has benefited greatly from the application of high-temperature and high-pressure laboratory experiments. This study presents: (1) new zircon U-Pb geochronology and strategies for characterizing and identifying ancient terrestrial material from the Inukjuak Domain in northern Québec, and the Jack Hills, Western Australia; and (2) a blend of new laboratory experiments and measurements of isotope ratios and trace impurities of natural zircon. Research directions in need of future exploration, with emphasis on early Earth studies, are also explored. Topics include Hadean hydrous magmatism and the structural accommodation of “water” into the zircon lattice, Hadean subaerial crust and the identification of peraluminous or metaluminous source melts, methods to characterize the oxidation state of magmas and fluids, and the complementarity of the Si- and O-isotopic systems as proxies for crustal weathering. Finally, the implications of this work are discussed in the context of a possible transition from prebiotic to biotic chemistry on the early Earth.

**Keywords:** Zircon, oxygen fugacity, hydrous magmatism, Inukjuak, Jack Hills, Si isotopes, origin of life, FTIR

**INTRODUCTION**

Zircon (ZrSiO$_4$) is an excellent carrier of geochemical information through time. In many ways, it is unsurpassed in its physical and chemical durability, and it is readily dated because it incorporates U and Th that decay to isotopes of Pb. It is because of this robustness that zircons are often found in sediments. The mere presence of this mineral is not diagnostic or unique to any specific rock type, tectonic setting, or environment; zircons occur in the continental and oceanic crust, in kimberlites, and in some meteorites and lunar rocks (Ireland and Wlotzka 1992; Valley et al. 1998; Watson et al. 2006; de Hoog et al. 2014; Barboni et al. 2017). It is because zircons do crystallize in diverse settings that researchers have turned to the investigation of trace element chemistry and key isotopic ratios as proxies for their formation environment.

The importance is amplified because detrital zircons yield ages as old as ~4.4 Ga, which exceeds the age of the oldest known, widely agreed upon rock by about 400 million years (Maas et al. 1992; Wilde et al. 2001; Mojzsis et al. 2001; Holden et al. 2009; Thern and Nelson 2012; Mojzsis et al. 2014). Over the past two decades, numerous geochemical investigations of Hadean zircon (>4.0 Ga) have been conducted (e.g., Wilde et al. 2001; Mojzsis et al. 2001; Cavosie et al. 2005, 2006; Trail et al. 2007; Harrison et al. 2008, 2017; Hopkins et al. 2008; Bell et al. 2011, 2017). These studies, which are mainly based on measurements of: (1) oxygen and hafnium isotope ratios; (2) rare earth element (REE) contents; and (3) the composition of other minerals (inclusions) found within the zircons, have led researchers to conclude that the early Earth contained an evolved rock cycle including water-rock interaction, formation of granitic crust and probable sediment cycling. The above studies typically utilized experimental zircon diffusion data (see Cherniak 2010) to argue for primary retention of trace element contents and isotope ratios of ancient grains.

In a generally different strategy, researchers sought to develop zircon-based calibrations in the controlled setting of an experimental geochemistry laboratory. Such experiments have played a fundamental role in the quest to link chemical signatures preserved in ancient zircons with Hadean processes. For instance, the Ti content of zircon was calibrated as an indicator of a zircon’s crystallization temperature and zircon Ce anomalies were investigated as a proxy for redox conditions of early Earth magmas (Watson and Harrison 2005; Watson et al. 2006; Ferry and Watson 2007; Trail et al. 2011a, 2012). This work led to new discoveries about the early Earth, including evidence for water-saturated or near water-saturated Hadean magmas and suggestions that Hadean volcanic emanations that were broadly neutral (e.g., CO$_2$) rather than uniformly reduced (e.g., CO).

This paper presents new data and some speculations, and it additionally highlights a handful of research directions that are presently of interest to the author. With little deviation, this contribution explores the chemistry of the terrestrial zircon age end-members. A progress report is presented for zircon U-Pb geochronology studies of the Inukjuak Domain and the Jack Hills aimed at identifying the oldest terrestrial zircons and new