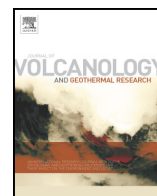




Contents lists available at ScienceDirect

Journal of Volcanology and Geothermal Research

journal homepage: www.elsevier.com/locate/jvolgeores

Carbon isotope composition of CO₂-rich inclusions in cumulate-forming mantle minerals from Stromboli volcano (Italy)

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ARTICLE INFO

Article history:

Received 14 December 2016

Received in revised form 14 March 2017

Accepted 2 April 2017

Available online xxxxx

Keywords:

Stromboli volcano

CO₂

Ultramafic cumulates

δ¹³C

Fluid inclusions

Mantle

ABSTRACT

We report on measurements of concentration and carbon isotope composition (δ¹³C_{CO2}) of CO₂ trapped in fluid inclusions of olivine and clinopyroxene crystals separated from San Bartolo ultramafic cumulate Xenoliths (SBX) formed at mantle depth (i.e., beneath a shallow Moho supposed to be at 14.8 km). These cumulates, erupted about 2 ka ago at Stromboli volcano (Italy), have been already investigated by Martelli et al. (2014) mainly for Sr-Nd isotopes and for their noble gases geochemistry. The concentration of CO₂ varies of one order of magnitude from 3.8 · 10⁻⁸ mol g⁻¹ to 4.8 · 10⁻⁷ mol g⁻¹, with δ¹³C values between -2.8‰ and -1.5‰ vs V-PDB. These values overlap the range of measurements performed in the crater gases emitted at Stromboli (-2.5‰ < δ¹³C_{CO2} < -1.0‰). Since SBX formed from relatively primitive mantle-derived basic magmas, we argue that the isotope composition displayed by fluid inclusions and surface gases can be considered representative of the magma volatile imprinting released by partial melting of the mantle source beneath Stromboli (-2.8‰ < δ¹³C < -1.0‰). In addition, the δ¹³C signature of CO₂ is not significantly modified by fractionation due to magmatic degassing or intracrustal contamination processes owing to magma ascent and residence within the volcano plumbing system. Such δ¹³C values are higher than those commonly reported for MORB-like upper mantle (-8 ‰ ÷ -4‰) and likely reflect the source contamination of the local mantle wedge by CO₂ coming from the decarbonation of the sediments carried by the subducting Ionian slab with a contribution of organic carbon up to 7%.

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1. Introduction

Fluid inclusions are small gas bubbles (order of microns) trapped as “defects” in the crystals during their growth, and represent a very useful tool for understanding the formation process of the rock containing the minerals. They can be found in terrestrial igneous, metamorphic, sedimentary rocks, but also in extraterrestrial (Roedder et al., 1963; Roedder, 1984). In magmatic environments, the most investigated minerals are olivine and pyroxene, that are the first to growth in the silicate melts and whose crystallographic characteristics allow preservation of the fluids, amenable to noble gas studies (e.g., Hilton et al., 1993; Trull and Kurz, 1993; Nuccio et al., 2008). The most common fluid inclusions are CO₂-dominated and the study of its concentration and isotope composition is generally coupled with the inferences obtained by noble gases to assess source characteristics, the extent of degassing or phenomena of crustal or atmospheric contamination of the magmas during their ascent towards the surface (e.g., Macpherson et al., 1999; Deines, 2002; Burnard et al., 2004; Aubaud et al., 2005; Aubaud et al., 2006; Shaw et al., 2006; Nuccio et al., 2008; Martelli et al., 2011; Correale

et al., 2012, 2014, 2015; Paonita et al., 2012). The carbon isotope composition of CO₂ is also a powerful tracer to determine the composition and evolution of the fluids present in the mantle (e.g., Pineau and Mathez, 1990; Sano and Marty, 1995; Yang et al., 2001), or to recognize the contribution of carbonate sediments from subducting slabs acting to metasomatize the mantle wedge (e.g., Sano and Marty, 1995; Hilton et al., 2010; Vaselli et al., 2010).

The fluid geochemistry of Stromboli, active volcano in the Aeolian islands, has attracted the interest of many authors that have investigated the fluid output, the chemical and isotope characteristics of water and gases either during eruptive or quiescence phases (Carapezza and Federico, 2000; Capasso et al., 2005; Allard et al., 2008; Federico et al., 2008; Grassa et al., 2008; Aiuppa et al., 2010; Inguaggiato et al., 2011; Rizzo et al., 2009, 2015). Such studies identified the presence of a hydrothermal aquifer (T ~ 40 °C) at a few meters below sea level that is routinely sampled to detect eventual temporal variations of geochemical parameters connected with volcanic activity. Fumaroles present at the top of the volcano have been also sampled, but the air contamination and the high-risk area limit the possibility to acquire reliable data and robust continue time series. Finally, soil CO₂ fluxes in the summit area, CO₂/S ratio in the plume gases and the SO₂ total flux are measured during volcanic surveillance activities.

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An issue that commonly occurs in fluid geochemistry studies is how different are the elemental ratios and isotope values of surface fluids with respect to the magmatic signatures, that in turn are inherited from the mantle source. Many processes may indeed hide or obliterate the pristine magmatic signature of light stable isotopes, through their fractionation, during magma/gas rising and residence in the crust, mixing of different magma batches, magma degassing, crustal assimilation, gas-water interaction and atmospheric contamination (Hilton et al., 1993; Trull et al., 1993; Deines, 2002; Paonita et al., 2012; Correale et al., 2014, 2015; Di Piazza et al., 2015; Robidoux et al., 2016). It is therefore very helpful to compare surface fluids to fluid inclusions entrapped at depth in magmatic minerals.

Despite the Stromboli plumbing system being characterized by subvolcanic cumulate roots from mantle depth (Cigolini et al., 2008), to lower-middle crust (undeformed and mylonitic gabbros; Mattioli et al., 2003; Spiess et al., 2017) only one distinct suite of ultramafic xenoliths has been recognised (S. Bartolo Xenoliths, SBX; Laiolo and Cigolini, 2006). Such a suite is constituted by wehrlite and dunite cumulates, through the crystallization of basic magmas formed by partial melting, driven by the slab fluids, of the mantle-wedge beneath Stromboli. Therefore, these cumulates offer the opportunity to study the fluids entrapped therein these formed at mantle depth. SBX did not interact with crustal rocks (Martelli et al., 2014) and are thus witnesses to deep (>15 km, namely the Moho crust-mantle transition, Morelli et al., 1975; Panza et al., 2007; Martinez-Arevalo et al., 2009) magmatic processes of Stromboli before any eventual process that may occur during rising and storage of magma in the volcano plumbing system. Olivine and clinopyroxene minerals of SBX are gas-rich and this allowed their $^3\text{He}/^4\text{He}$ ratios to be measured and compared to values reported for Stromboli over its subaerial history (the last 100 ka) and in the aqueous and gaseous emissions of surface fluids (Martelli et al., 2014).

In this work, we investigate the elemental and isotope composition of CO_2 trapped in SBX minerals to compare the results to those of fluids emitted at the surface and gain more information about the origin of fluids in the mantle beneath Stromboli and within its magmatic system.

2. Study area

The island of Stromboli (Italy) belongs to the Aeolian volcanic arc, which is the result of the subduction of African plate beneath that European (e.g., Gasparini et al., 1982). It represents one of seven islands emerging in the south-eastern Tyrrhenian Sea and has dimensions comparable to Mt. Etna, which is the largest strato-volcano in Europe. Indeed, Stromboli has an edifice with approximately 2 km b.s.l. and 1 km a.s.l., which developed on a relatively thin continental crust (e.g., Morelli et al., 1975; Pontevivo and Panza, 2006). In particular, according to the teleseismic studies of Martinez-Arevalo et al. (2009) the thinned crust beneath Stromboli is now defined by a shallow Moho at a depth of 14.8 km, where S-wave mantle velocities reach 4.0 km/s. The S-waves increase with depth, reaching a velocity of 4.58 km/s at 32 km, then with a strong V_S reduction (as low as ca. 3.0 km/s) in the underlying layer down to about 60 km. This low velocity layer in the uppermost mantle just below 32 km thus suggests a possible, partially melted, region.

Stromboli volcano is famous worldwide for the persistent “Strombolian” activity, characterized by periodic explosions, sporadic violent explosions (“major explosions” or “paroxysms”) and lava effusions mainly flowing in the NW flank of the cone named “Sciara del Fuoco” (e.g., Rosi et al., 2000). Magma feeding the eruption of juvenile products, associated with the present-day Strombolian activity and/or with lava effusion, produces high porphyritic (HP), volatile-poor, basaltic scoriae. Conversely, major explosion or paroxysms are fed by a deep-provenance magma erupting pumices with low phenocrysts (LP) content and volatile-rich (e.g., Francalanci et al., 2004). HP and LP magmas have the same bulk composition but differ mostly for the degrees of crystallization and for some geochemical tracers such as

$^{87}\text{Sr}/^{86}\text{Sr}$ ratio (e.g., Pompilio et al., 2012). During the periods of intense eruptive activity a mingling process between the two magmas can be observed, either at macroscopic or at microscopic scale (Bertagnini et al., 1999, 2008; Francalanci et al., 1999, 2004).

Over the subaerial eruptive history of Stromboli, four different magmatic series have been recognised: calc-alkaline (CA), high potassium calc-alkaline (HKCA), shoshonitic (SHO), and potassic (KS), with a progressive increase in potassium, in different trace elements contents and in Sr- and Nd-isotope ratios (Francalanci et al., 1988, 1989; Hornig-Kjarsgaard et al., 1993). Mafic terms ($\text{SiO}_2 < 55\%$, $\text{Mg} > 4\%$) are present in all the magmatic series, while no clear temporal progression between one series and another exists. The wide variation of elemental and isotope characteristics among the series has been investigated by many studies, with the widely accepted conclusion that rocks of CA, HKCA and SHO series originate from mantle source heterogeneity rather than wall-rock crustal assimilation (Ellam et al., 1989; De Astis et al., 2000; Francalanci et al., 2007; Tommasini et al., 2007). Some extent of crustal assimilation may have occurred for certain samples of the KS series (e.g., Tommasini et al., 2007).

The most accepted explanation for this peculiar geochemical variability considers the partial melting of a heterogeneous mantle wedge affected by various extents and types of metasomatism due to fluids from the Ionian subducting slab (Ellam et al., 1989; Francalanci et al., 1993, 2004; Tommasini et al., 2007; Schiavi et al., 2012). The wedge beneath Stromboli would be therefore enriched by fluids released by the slab, both from its basaltic crust and the relative sedimentary cover (Ellam et al., 1989; Peccerillo, 2001; Francalanci et al., 2007; Tommasini et al., 2007).

HP and LP products erupted at present day belong to SHO magmatic series, and prior to eruption are stored at respectively 2–4 km and 7–10 km in the crust (e.g., Métrich et al., 2010). The steady state of persistent Strombolian activity erupting HP magma seems to be ensured by the continuous refilling of the shallow magma body by deep volatile-rich LP magmas and by the contemporarily magma emission from the craters (e.g., Landi et al., 2009). Among the Stromboli extrusives, the only evidence of products coming directly from the mantle depth (>15 km) are represented by the ultramafic cumulate xenoliths brought to the surface by the San Bartolo magma (HKCA series; Francalanci et al., 1989) that erupted ca. 2 ka ago (Laiolo and Cigolini, 2006; Cigolini et al., 2008; Martelli et al., 2014). These products (San Bartolo Xenoliths, SBX), formed at subcrustal levels through cumulate fractionation of mafic minerals from mantle-derived basic magmas, are the subject of this study.

3. Samples and methods

3.1. Samples description and preparation

Ten aliquots of olivine and clinopyroxene crystals have been separated from four different ultramafic nodules (Fig. 1). Part of these SBX nodules has been studied and well characterized by Martelli et al. (2014) for major and trace elements, for Sr-Nd and noble gases isotope composition trapped in fluid inclusions (see Section 6 for further details).

The samples were prepared with great care. After grinding and sieving the rocks, crystals without impurities and >0.5 mm were hand-picked under a binocular microscope following the laboratory protocol developed at the Istituto Nazionale di Geofisica e Vulcanologia (INGV), Sezione di Palermo (Italy) (e.g., Nuccio et al., 2008; Correale et al., 2012, 2014, 2015). Before loading minerals in the sampler, these were ultrasonically cleaned in 6.5% HNO_3 and then rinsed with deionized water. After that, they were accurately weighted and loaded in a stainless steel sampler (1 cm internal diameter, 6–10 cm length) (Fig. 2a, on the right). The amount of crushed sample varied from 0.3 to 1.2 g, depending on the dimensions and availability of nodules (Table 1).

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