



Understanding phengite argon closure using single grain fusion age distributions in the Cycladic Blueschist Unit on Syros, Greece



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ABSTRACT

The preservation of $^{40}\text{Ar}/^{39}\text{Ar}$ ages of high pressure (HP) metamorphic white mica reflects an interplay of processes that mobilise ^{40}Ar , either through mica recrystallisation or by diffusive ^{40}Ar loss. The applicability of resulting ages for dating tectonic processes is critically dependent on whether either of these processes can be proven to be efficient and exclusively active in removing ^{40}Ar from mica. If not, preservation of an inherited or mixed age signal in a sample must be considered for interpretation. The Cycladic Blueschist Unit on Syros has become a new focal area in the discussion of the geological significance of argon age results from multi-grain step heating experiments. While some argue that age results can directly be linked to deformation or metamorphic growth events, others interpret age results to reflect the interplay of protracted recrystallisation and partial resetting, preserving a mixed age signal. Here, we demonstrate the potential of a new approach of multiple single grain fusion dating. Using the distribution of ages at the sample, section and regional scale, we show that in Northern Syros mica ages display systematic trends that can be understood as the result of three competing processes: 1) crystallisation along the prograde to peak metamorphic path, 2) a southward trend of increasing ^{40}Ar loss by diffusion and 3) localised and rock type dependent deformation or metamorphic reactions leading to an observed age spread typically limited to ~ 10 Myr at the section scale. None of the sections yielded the anomalously old age results that would be diagnostic for significant excess ^{40}Ar . The recorded trends in ages for each of the studied sections reflect a range of P–T conditions and duration of metamorphism. Diffusion modelling shows that in a typical subduction metamorphic loop, subtle variations in P–T–t history can explain that age contrasts occur on a regional scale but are limited on the outcrop scale. Our new approach provides a comprehensive inventory of the range of ages present in different rocks and at different scales, which results in a more refined understanding of argon retention and isotopic closure of phengite and the geological significance of the ages. We verify the added value of our new approach by comparison with multi-grain step heating experiments on selected samples from the same sections.

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1. Introduction

1.1. Preservation of white mica $^{40}\text{Ar}/^{39}\text{Ar}$ ages

The abundance of potassium-bearing white mica in HP metamorphic rocks and their importance in forming rock fabrics has made their $^{40}\text{Ar}/^{39}\text{Ar}$ age an attractive potential recorder of the timing and rate of metamorphism and deformation. However, the complex interplay of factors controlling argon mobility under HP conditions limits the possibility to attribute white mica $^{40}\text{Ar}/^{39}\text{Ar}$ ages directly to a single geological mechanism and thus to (re)crystallisation, cooling, tectonic or fluid infiltration events

(e.g., Beltrando et al., 2009; Bröcker et al., 2013; Laurent et al., 2017; Lips et al., 1998; Lister and Forster, 2016; McDonald et al., 2016; Putlitz et al., 2005; Scaillet et al., 1990; Warren et al., 2012b; Wijbrans and McDougall, 1986).

White mica $^{40}\text{Ar}/^{39}\text{Ar}$ ages are interpreted in the context of processes that govern the mobility of radiogenic ^{40}Ar following initial mica crystallisation. In-situ produced radiogenic ^{40}Ar can be removed from a crystal through two mechanisms: 1) thermally activated volume diffusion, potentially allowing the determination of a cooling history (Dodson, 1973; Wijbrans and McDougall, 1986), or 2) recrystallisation, potentially dating a reaction or a deformation driven recrystallisation event (Villa, 1998). In some HP terranes, externally produced radiogenic ^{40}Ar is added to mica through infiltration of 'parentless' or excess ^{40}Ar . Infiltration of externally sourced ^{40}Ar in minerals may occur by incorporation in fluid inclusions during mineral growth or deformation

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(Qiu and Wijbrans, 2006), by partitioning during crystal growth or by volume diffusion in existing crystals from a high concentration grain boundary reservoir (Kelley, 2002; Menold et al., 2016).

For measured ages to unambiguously date cooling or (re)crystallisation events, ^{40}Ar loss must be efficient so that all locally produced radiogenic argon is removed by the process targeted for dating and no inherited argon age signal remains. Inherited argon may remain when the grain boundary system is not an open system and argon cannot be transported away to a local sink. In addition, the interpreted age forming process must be the only process removing ^{40}Ar from the crystal lattice, so (re)crystallisation ages are not altered by subsequent diffusive ^{40}Ar loss and *vice versa*. The broad age ranges commonly found for rocks from subduction zones indicate that assumptions regarding the full efficiency and exclusive activity of a resetting process are often not valid (e.g., Bröcker et al., 2013). Previous modelling studies suggest that mica crystallisation ages can be preserved in the greenschist and blueschist domain (Warren et al., 2012a) and rock permeabilities are potentially too low to allow effective removal of argon (Baxter et al., 2002; Smye et al., 2013; Warren et al., 2012b). The resultant apparent unpredictability of argon mobility during the metamorphic cycle remains a critical issue to be resolved before the method can be unambiguously applied to dating tectonic events.

The Cycladic Blueschist Unit (CBU) in Greece is emerging as a key area for testing new interpretations of subduction related white mica $^{40}\text{Ar}/^{39}\text{Ar}$ ages. Multi-grain step heating experiments on phengite from blueschists and eclogites from Sifnos and Syros produce undulating age spectra ranging between 20 and 60 Ma. While these experiments are classically regarded as producing geologically meaningless mixed ages (Baldwin, 1996; Bröcker et al., 2013), others assume a higher closure temperature for argon diffusion in mica and argue that steps in age spectra reflect the timing of recrystallisation (Laurent et al., 2017; Villa, 1998) or metamorphic growth events (Lister and Forster, 2016). Lacking clear evidence of the isotopic closure mechanism and the degree of age heterogeneity in a sample, interpretation of multi-grain step heating experiments remains ambiguous.

Here, in an alternative approach, multiple single grain fusion $^{40}\text{Ar}/^{39}\text{Ar}$ dating is used as a quick and efficient means to assess the range of bulk grain ages in a sample. The key advantage of this method is that it constrains age heterogeneity within a sample and at larger scales by taking multiple samples from an outcrop, section or region. The method is applied to four >100 m metasedimentary sections in the CBU of Northern Syros. Our new results are contrasted with constraints from previous studies (Bröcker et al., 2013; Cliff et al., 2017; Lagos et al., 2007; Laurent et al., 2017; Lister and Forster, 2016; Maluski et al., 1987; Putlitz et al., 2005; Rogowitz et al., 2015; Tomaschek et al., 2003), and interpreted in the context of diffusion modelling to obtain a better understanding of age preservation in HP metamorphic systems.

1.2. Multiple single grain fusion approach

In low to moderate temperature (<500 °C) metamorphic terranes, limited argon diffusion (Harrison et al., 2009; Warren et al., 2012a) and protracted recrystallisation during overprinting will result in contrasting bulk ages for individual mica crystals at the scale of a hand specimen. In conventional multi-grain step heating methods, apparent age spectra based on mixtures of such different age signals need to be interpreted within the method's limitations (e.g., Bröcker et al., 2013; Lister and Forster, 2016). Different mica generations inevitably degas simultaneously, as demonstrated by experiments on artificial mixtures of micas with contrasting ages (Kula et al., 2010; Wijbrans and McDougall, 1986) and by selective dating in micro-structurally well-characterised samples (Beltrando et al., 2009; Lips et al., 1998). The resulting age spectrum is geo-

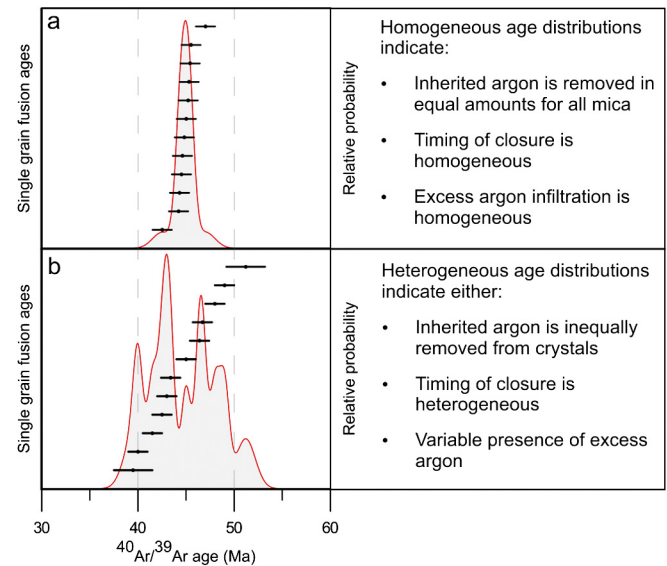


Fig. 1. Theoretical single grain fusion age distributions (black bars) and their relative probability density plots (red curve). a) Uniform Gaussian distributions result from efficiently open or closed system conditions and a uniform closure mechanism. b) Heterogeneous age populations indicate perturbed argon systematics by locally varying processes. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

logically meaningless as no precise end-member ages can be extracted from this mixture.

Flat plateau shapes commonly obtained for step heating experiments performed on single crystals in the greenschist–blueschist domain (Huet et al., 2015; Laurent et al., 2017; Wijbrans et al., 1990) indicate that single grain incremental heating and single fusion results often yield equivalent information per crystal. We contend that different information is obtained by multiple single grain laser fusion experiments, which efficiently constrain the size and shape of the age distribution within a sample. The information obtained provides quantitative constraints on the homogeneity of the age distribution and hence the mechanism controlling ^{40}Ar loss.

For a given grain size, a single Gaussian population of single grain fusion ages (Fig. 1a) suggests that ages were influenced by processes acting uniformly at the sample scale. This implies: 1) if present, any inherited age signals have been removed in equal amounts from all crystals before closure, 2) the timing of closure for that grain size is uniform at the sample scale and 3) any excess argon has infiltrated equally in each crystal. Heterogeneous distributions (Fig. 1b) suggests that one or more of the following processes affected the mica age population: 1) inherited ^{40}Ar is variably removed from the crystals due to inefficient diffusion or locally closed conditions limiting argon mobility; 2) the timing of closure is heterogeneous due to, e.g., localised continued (re)crystallisation below conditions for diffusion-controlled resetting or 3) locally varying amounts of excess argon have infiltrated the mica following closure. Homogeneous distributions indicate that argon mobility is systematically open or closed on the sample scale, where fully open-system behaviour is the simplest case. Heterogeneous distributions demonstrate that the assumptions for complete resetting in open system conditions are not met. Instead, the obtained age information is the result of processes that did not go to completion or acted heterogeneously on the sample scale.

The size and shape of a multiple single grain fusion age population show whether age heterogeneity occurs within a sample and reflects the degree of heterogeneity, e.g., whether the age population spans the entire metamorphic cycle or only covers certain segments of a P–T–t loop. When compared for different rocks in

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