The U-Pb systematics and cooling rate of plutonic angrite NWA 4590. Y. Amelin[1], A. Kaltenbach[2] and C.H. Stirling[2], [1]Research School of Earth Sciences, Building 61, Mills Road, The Australian National University, Canberra ACT 0200 Australia (yuri.amelin@anu.edu.au), [2]Centre for Trace Element Analysis and Department of Chemistry, University of Otago, PO Box 56, Union Place, Dunedin, New Zealand.

Introduction: Due to their rich and unique mineralogy and good preservation, angrites offer an excellent opportunity for studying chronology of one of the oldest known differentiated bodies in the Solar System. Since the earliest chronological studies [1, 2], it has been known that angrites contain at least two U-bearing minerals suitable for precise U-Pb dating: Al, Ti-rich clinopyroxene, and Ca-phosphate merrillite. Combining the ages of these minerals from the same meteorite with the rate of Pb diffusion known from experimental studies makes it possible to calculate cooling rates of meteorite parent bodies and precisely link these cooling rates to the absolute time scale. However, recent studies of angrite chronology focused on dating rock-forming minerals and whole rocks that approximate the timing of crystallization, whereas the opportunity of direct chronological determination of the cooling rate of the angrite parent body remains to be explored.

Angrite NWA 4590, a very fresh coarse-grained igneous cumulate rock composed of Al-Ti-rich clinopyroxene, anorthite, Ca-rich olivine with kirschsteinite exsolution, ulvöspinel, and accessory minerals [3, 4] is one of the most suitable meteorites for such determination. Among other accessory minerals, NWA 4590 contains merrillite [5], a mineral that was successfully used in U-Pb dating of other angrites. The crystallization age of NWA 4590 of 4558.86 ± 0.30 Ma (assuming $^{238}U/^{235}U$=137.88) was determined from a Pb-Pb isochron for five pyroxene fractions [6]. Here we report the uranium isotopic composition of NWA 4590, and results of a detailed U-Pb study of rock-forming and accessory minerals, including silico-phosphate – the U-rich mineral that appears to be the major host of uranium in this meteorite.

Methods: Uranium isotope composition was analyzed at the University of Otago using techniques of [7]. First five U-Pb analyses of pyroxene, and three U-Pb analyses of whole rock were carried out at the Geological Survey of Canada using techniques of [8]. All other U-Pb analyses were carried out at the Australian National University using techniques of [7].

Since we report an exceptionally precise Pb-Pb isochron age for the silico-phosphate, additional details of standardization of Pb isotope analysis, justifying this precision, are described here. As in our previous studies, reproducibility and accuracy of Pb-isotope analyses were checked using 300 pg aliquots of SRM-981 standard, spiked with a mixed tracer containing ca. 30 pg of each of ^{202}Pb and ^{205}Pb. Sixteen such analyses on a MAT-261 mass spectrometer at the ANU over the course of this study yielded average values 0.05903 (±0.23%, 2 s.d.) for $^{204}Pb/^{206}Pb$, 0.91476 (±0.024%, 2s.d.) for $^{207}Pb/^{206}Pb$, and 2.16809 (±0.035%, 2 s.d.) for $^{208}Pb/^{206}Pb$. These uncertainties were quadratically added to within-run errors of analyses of most meteorite fractions that contained similar amounts of Pb, and were spiked with the same amount of the mixed tracer. Larger (1.5 ng) loads of the SRM-981 standard, spiked with a mixed tracer containing 150 pg of each of ^{202}Pb and ^{205}Pb, were analyzed along with the larger silico-phosphate fractions. These standard analyses yielded average values 0.05903 (±0.021%, 2 s.d.) for $^{204}Pb/^{206}Pb$, 0.914717 (±0.006%, 2s.d.) for $^{207}Pb/^{206}Pb$, and 2.16787 (±0.008%, 2 s.d.) for $^{208}Pb/^{206}Pb$. These smaller uncertainties were used in error propagation for analyses of the larger silico-phosphate fractions containing ca. 2 ng of Pb.

Results: Two whole rock U isotope analyses yielded consistent values with the weighted average $^{238}U/^{235}U$ ratio of 137.789±0.021, reported relative to the value of 137.836 in the CRM-145 standard. The $^{238}U/^{235}U$ ratio in NWA 4590 is lower than the "terrestrial" value of 137.821, but is higher than the Allende meteorite whole rock and chondrule value of 137.747±0.017 [7]. The $^{238}U/^{235}U$ ratio in NWA 4590 is also almost identical (after re-normalization to the same standard ratio) to the value reported for the angrite D'Orbigny by [9], suggesting that U isotopic composition in the angrite parent body is homogeneous.

A Pb-Pb isochron for six pyroxene fractions (Fig.1; five analyses at the GSC, and one at the ANU) yielded the age of 4557.93±0.28 Ma, calculated using the measured $^{238}U/^{235}U$ ratio of 137.789. Including the error of $^{238}U/^{235}U$ ratio introduces additional age uncertainty of 0.22 Ma, and increases the total age error to ±0.36 Ma.

All whole rock data points plot below the pyroxene isochron (Fig. 2), suggesting the presence of "young" radiogenic Pb. The likely carriers of this Pb components are fragments of quartz grains (probably a terrestrial contamination), which were found in the rock. Low $^{207}Pb/^{206}Pb$ at relatively high $^{206}Pb/^{204}Pb$, measured in the acid washed plagioclase fraction, suggests that some quartz fragments were accidentally included

during hand-picking. The Pb isotopic composition of the whole rock fractions can be interpreted as a mixture of ca. 4558 Ma radiogenic Pb, non-radiogenic Pb of asteroidal and/or terrestrial origin, and young radiogenic Pb contained in quartz.

A notable feature of Pb-isotopic systematics of plagioclase and whole rock fractions put through acid washing is that the first (ultrasonic treatment in 0.5 M HNO_3) leachates contain substantially more radiogenic Pb than second (hot concentrated HNO_3 and HCl) leachates. The source of soluble radiogenic Pb was identified with silico-phosphate – a mineral that easily dissolves in 0.5 M HNO_3 at room temperature, and contains 11-22 ppm U. Detailed information on the composition and structure of this mineral are presented in the companion abstracts by Amelin, Iizuka and Huyskens, and Mikouchi et al., respectively. A Pb-Pb isochron for nine silico-phosphate fractions (Fig.1) yielded the age of 4557.381±0.066 Ma (or ±0.23 Ma, including uncertainty in the $^{238}U/^{235}U$ ratio), calculated using the measured $^{238}U/^{235}U$ ratio of 137.789. The U-Pb systems in all pyroxene and silico-phosphate fractions are concordant or less than 2% discordant, thus confirming the closed-system behavior and reliability of the ages.

Discussion: The age difference of 0.55±0.29 Ma between pyroxene and silico-phosphate can be interpreted as a result of later closure of Pb diffusion in silico-phosphate a in slowly cooling rock. Assuming the difference in closure temperature of 296±50 Ma [10] for Pb diffusion in pyroxene and silico-phosphate, this age difference corresponds to the cooling rate of 540±290 K/Ma. This cooling rate is 20-100 times faster than the cooling rate of the H-chondrite parent body, determined from equilibrated chondrites Richardton [10] and Estacado [11] using similar approach. Fast cooling could imply a small size of the angrite parent body, but may also be a result of a rapid cooling event [4, 8, 12] that probably affected the source rocks of plutonic angrites at 4557-4558 Ma.

Alternatively, the apparent age difference between pyroxene and silico-phosphate could be unrelated to cooling, but caused by fractionation between ^{235}U and ^{238}U during mineral growth. The difference in the $^{238}U/^{235}U$ ratio of 0.052, or 3.8 ε-units, is required to explain the measured age difference (assuming simultaneous closure of Pb diffusion in both minerals). The latter possibility can be tested by separate high-precision U isotopic analysis of silico-phosphate and acid-washed pyroxene.

References: [1] Wasserburg G. J. et al. (1977) *EPSL* 35, 294–316. [2] Chen J. H. & Wasserburg G. J. (1981) *EPSL 52*, 1-15. [3] Irving A. J. et al. (2006) EOS, Trans. AGU 87, Fall Mtg. Suppl., Abstract #P51E-1245. [4] Kuehner S. M. and Irving A. J., (2007) 38th *LPS*, Abstract # 1522. [5] Sanborn M. E. and Wadhwa M. (2010) 40th *LPS*, Abstract # 1345. [6] Amelin Y. and Irving A. J. (2007) *Workshop on Chronology of Meteorites*, Abstract # 4061. [7] Amelin Y. et al. (2010) *EPSL* 300, 343-350. [8] Amelin Y. (2008) *GCA 72*, 221–232. [9] Brennecka et al. (2010) 41st *LPS*, Abstract # 2117. [10] Amelin Y. et al. (2005) *GCA* 69, 505–518. [11] Blinova A. et al. (2007) *MPS* 42, 1337–1350. [12] McKay et al. (1998) *MPS* 33, 977–983.

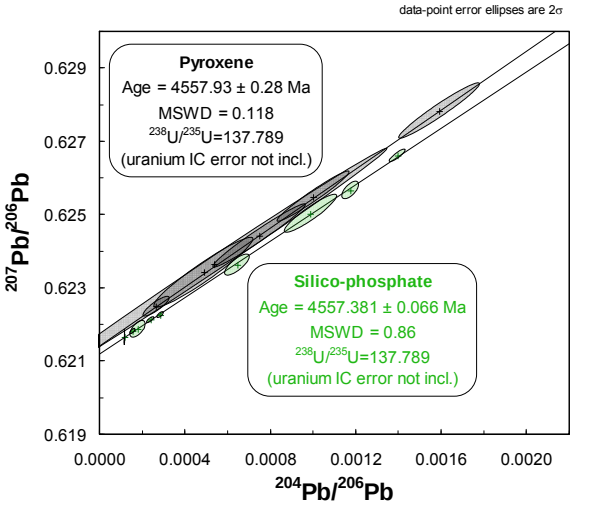

Fig. 1. Pb-Pb isochrons for pyroxene (6 fractions) and silico-phosphate (9 fractions) from angrite NWA 4590. The age errors do not include uncertainty of the $^{238}U/^{235}U$ ratio.

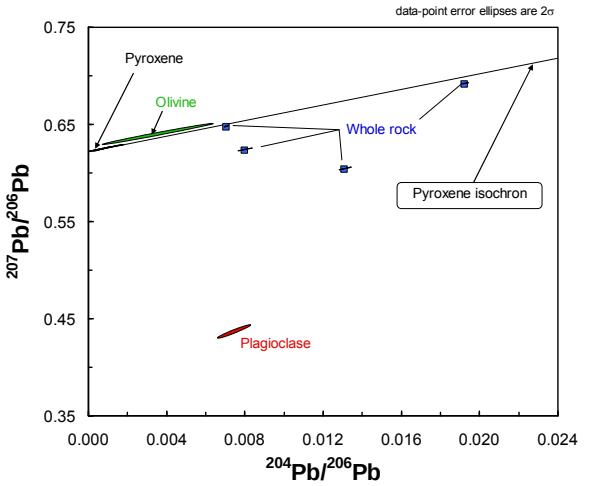

Fig. 2. Pb-isotopic systematics of whole rock samples and acid-washed silicate minerals in the angrite NWA 4590.