LITHIUM STABLE ISOTOPE RECORDS OF IRRADIATION IN APOLLO LUNAR ZIRCON. H. Tang[1], Y. Li[2], D. Trail[3], and K.D. McKeegan[1], [1]Department of Earth, Planetary, and Space Sciences, University of California Los Angeles, Los Angeles CA; [2]Lunar and Planetary Science Laboratory, Macau University of Science and Technology, Macau, China; [3]Department of Earth and Environmental Sciences, University of Rochester, Rochester, NY. (haolantang@ucla.edu).

Introduction: The Moon is permanently inhospitable due to the bombardment of high energy particles of galactic and solar cosmic rays. The absence of a radiation-absorbing atmosphere and a global magnetic field at the present day results in the lunar surface being directly exposed to the external particles. These cosmic ray particles, with energies ranging from a few keV to over 100 GeV per nucleon, can generate large cascades of secondary particles (e.g. neutrons, protons, and pions), which, with their primary particles, can induce nuclear reactions in the target minerals, producing both radioactive and stable nuclides. Investigation of these cosmogenic nuclides in lunar samples can improve our understanding of the evolution of lunar regolith, solar activity, and cosmic ray radiation [1].

Previous work done on the noble gas isotopic compositions in the Apollo samples have indicated variable exposure ages. These ages measure the duration of the bulk samples exposed to the cosmic rays without considering the cosmic ray effects on individual minerals. On the other hand, some radioactive geochronometry systems, in particular minerals, (e.g., Lu-Hf in zircon) can be sensitive to the isotopic shifts generated from long-term cosmic ray effects. Thus, in this project we revisited exposure ages of Apollo zircons separated from lunar breccia and lunar soils by evaluating the effects of cosmic ray irradiation on lithium stable isotope compositions. Ultimately, the revisited exposure ages of lunar zircons may help improve our understanding of the history of lunar regolith.

Sample and analytical methods: The lunar samples investigated here were collected at the Apollo 14 landing site located 600-800 km from the rim of the Imbrium basin. Zircons were separated from one lunar breccia (14311) and two lunar soils (14163 and 14259). In situ U-Pb dating of these samples yield crystallization ages ranging from 3.8 to 4.3 Ga. The previous noble gas analyses indicated the bulk exposure ages of ~661 Ma (14311), ~710 Ma (14163), and ~550 Ma (14259) [2-3].

In situ analyses of lithium concentration and isotopic composition in zircons were performed on the CAMECA ims-1270 ion probe at UCLA. Lithium isotopes (^{6}Li and ^{7}Li) and silicon abundance (^{28}Si) in zircon were measured in a peak jumping mode with the mass sequence of 5.7, ^{6}Li, ^{7}Li, and $^{28}Si^{+++}$. Positive Li and Si secondary ions were produced by a 5-12 nA O^{-} primary ion beam focused to a ~20 μm spot. A pre-sputtering time of 180 seconds was used to minimize surface Li contamination. The mass resolving power was ~2,200 and typical count rates of ^{6}Li and ^{7}Li in zircon were 5-12 counts per second. We utilized synthetic zircon ZLi13 grown in the University of Rochester experimental geochemistry laboratory. The standard zircon grain displays homogeneous Li concentration ([Li] = 0.40±0.01 μg/g) and ^{7}Li/^{6}Li ratio (12.1982±0.0118). The NIST NBS 612 glass was used as the isotopic standard (^{7}Li/^{6}Li = 12.4606) to internally correct the instrumental mass fractionation (IMF ~ 0.9853). In addition, Li concentration and isotopic composition of terrestrial samples with varied Li concentrations, including NIST NBS 616 glass, Duluth Complex anorthositic zircon (AS3), and kimberlite megacryst zircon (KIM) were also analyzed to monitor the accuracy of the *in situ* analyses.

Results and discussion: All the terrestrial samples analyzed with the lunar zircons exhibit consistent terrestrial Li isotopic values (^{7}Li/^{6}Li~12.2), indicating the reliability of our measurements. Li concentration varies from 3×10^{-4} to 10 μg/g in the lunar zircons. Significant cosmic ray effects on Li isotopic composition can be observed in most of Li-poor grains, especially for the samples from 14311. ^{7}Li/^{6}Li isotopic ratios range from 3.67 to 12.33 (Fig 1A). In addition, multiple spots on three large grains (> 400 μm) were analyzed to examine homogeneity of their isotope ratios (Fig. 1B). Given the different proportions of the contribution of spallogenic Li, mixing lines could be obtained between the terrestrial composition (^{7}Li/^{6}Li~12.2) and spallogenic ratio (^{7}Li/^{6}Li~1.32) (Fig. 1). Grains experiencing similar exposure histories will therefore follow the same mixing lines. Based on the principle of mass balance, the concentration of spallogenic Li can be estimated with the range of 0.01-0.69 ng/g in the lunar zircons. Additionally, as shown in Fig 1B, two of the large grains display relatively homogeneous contribution of spallogenic Li (14311,58z74 and 14259,511z6), whereas the spots on 14311,58z75 are distributed on different mixing lines, indicating heterogeneous spallogenic Li concentration ($[Li]_{spallation}$ ~ 0.10-0.44 ng/g).

The abundance of spallogenic Li in lunar zircons directly reflects their exposure histories. To calculate the exposure ages of lunar zircons, numerical simulation analyses are required to estimate the production rate of cosmogenic Li. Here given the properties of the bulk samples, we developed four numerical models. We defined a sphere with radius of 1000 m to simulate the

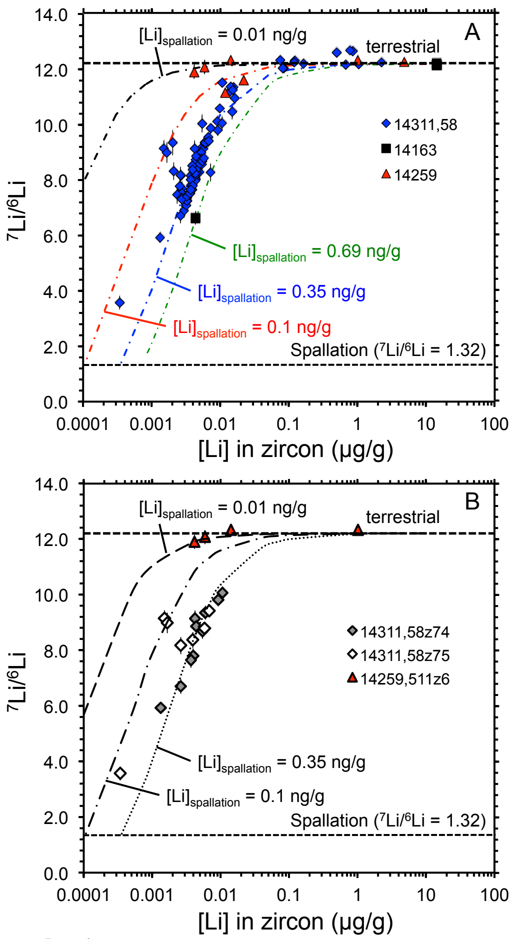

Fig. 1. $^7Li/^6Li$ isotopic ratio varied with lithium concentration in (A) all lunar zircons and (B) >400 μm lunar zircons. Different dash curves imply mixing lines with different contribution of spallogenic Li.

Moon and composition of lunar basalt. In model 1 and 2, the target layers to simulate production of spallogenic Li were composed of lunar basalt with defined depths of 0.9-1.1 cm and 6.9-7.1 cm, corresponding to the depth of zircon in lunar soil and lunar breccia (the diameter of 14311 sample is ~15 cm). In model 3 and 4, the target layers were composed of zircon ($ZrSiO_4$), while other parameters remained the same. We applied the physical model derived from analysis of radioactive nuclides (e.g., ^{26}Al and ^{14}C) in Apollo 15 drilling core based on Geant4 toolkit (GEometry And Tracking) [4]. In this model, galactic cosmic rays (0.01~100 GeV/nucleon) yielded a respective flux of proton and alpha particles of 3.498 $cm^{-2}s^{-1}$ and 0.339 $cm^{-2}s^{-1}$. The contribution from solar cosmic rays, although insignificant compared to galactic cosmic rays, was also counted in the models with the flux of 134 $cm^{-2}s^{-1}$ (10 ~1000 MeV). The production rates of spallogenic ^6Li in lunar basalt at 0.9-1.1 cm and 6.9-7.1 cm were determined to be 3.81×10^{-4} ng/g Ma^{-1} and 2.69×10^{-4} ng/g Ma^{-1}, respectively. With the target of zircon, these rates were reduced to be 2.50×10^{-4} ng/g Ma^{-1} and 1.76×10^{-4} ng/g Ma^{-1}. The latter production rates are favored in our calculation given that the cosmogenic Li nuclides were primarily generated inside of zircon compared to those implanted from surrounding minerals. $^7Li/^6Li$ ratios of spallogenic Li are 1.21 at 0.9-1.1 cm and 1.32 at 6.9-7.1 cm. As the result, ~72% of breccia zircons were exposed to cosmic rays for over 800 Ma with the maximum age of 1600 Ma (Fig. 2), indicating longer exposure histories in zircons compared to the bulk breccia (~661 Ma). Regarding the zircons from lunar soils, indigenous Li is relatively enriched in ~50% of zircons, hindering the quantification of spallogenic Li. A short exposure age of ~30 Ma can be inferred in the large 14259 zircon (14259,511z6), much shorter than that of the bulk soil sample (~550 Ma). In addition, the Li-depleted zircons from 14163 has an exposure age of ~1160 Ma, ~400 Ma longer than that of the bulk sample (~710 Ma).

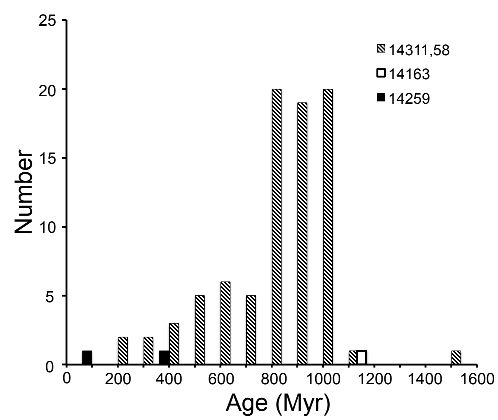

Fig. 2. Exposure ages in lunar zircons determined from spallogenic Li.

The inconsistency of exposure ages between zircons and their host rocks and soils could reflect the evolution of lunar regolith. The long average exposure ages of 14311 zircons compared to the bulk host breccia could suggest that the breccia was formed in 200-300 Ma after the zircons were exposed on the lunar surface. Likewise, the significantly short exposure history in the 14259 zircon relative to the lunar soil indicates a complex history of the lunar surface with multiple disturbing events. Additional analyses of other stable isotopic systems, such as boron, will be done in the near future to confirm the exposure ages in lunar zircons and further constrain the cosmic ray effects on the Moon.

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