THE ^{232}Th/^{238}U RATIO IN METEORITES FROM Pb ISOTOPES: A VALUABLE BY-PRODUCT OF U-Pb DATING. Y. Amelin[1], R.E. Zartman[2], T.R. Ireland[1], S. Dunn[3], A.N. Krot[4], Q.Z. Yin[5]. [1]Planetary Science Institute and Research School of Earth Sciences, The Australian National University, Canberra ACT 0200, Australia (yuri.amelin@anu.edu.au, trevor.ireland@anu.edu.au), [2]EAPS, Massachusetts Institute of Technology, Cambridge, MA 02139, USA (rzartman@mit.edu), [3]Curtin University of Technology, Perth, WA, Australia (S.Dunn@exchange.curtin.edu.au), [4]HIGP, University of Hawai‘i at Manoa, Honolulu, HI 96822, USA (sasha@higp.hawaii.edu), [5]Dept. of Geology, University of California Davis, Davis, CA 95616, USA (yin@geology.ucdavis.edu).

Introduction: The ^{232}Th/^{238}U ratio, traditionally termed "κ" (kappa), has many uses in cosmochemistry and geochemistry: from determination of the age of the Galaxy and constraining production rates of actinides in r-process [1, 2], to tracing the evolution of terrestrial crust and mantle [3-5]. Most commonly, the ^{232}Th/^{238}U ratio in meteorites is measured directly (κ_m), but the model ^{232}Th/^{238}U (κ_{Pb}) can also be calculated from the ^{208}Pb*/^{206}Pb* ratio and a known (or assumed) age. The κ_{Pb} is a geochemical tracer, which comes as a free bonus with every U-Pb age determination.

Here we discuss the uncertainties involved in the calculation of κ_{Pb}, compare the accuracy of κ_{Pb} and κ_m, and review some of the uses of κ based on examples from CAIs, bulk chondrites, and differentiated meteorites.

Uncertainties in κ_{Pb}: Uncertainties from three sources are involved in the determination of κ_{Pb}: the error in the ^{208}Pb*/^{206}Pb* ratio, the error in the age, and the deviation from the assumed single-stage evolution of the U-Th-Pb system.

(1) The error of the measured ^{208}Pb/^{206}Pb ratio propagates directly to the error of the ^{208}Pb*/^{206}Pb* ratio. For Pb with ^{206}Pb/^{204}Pb>50 and modern techniques of Pb-isotopic analysis, this source of uncertainty is smaller than 0.2-0.3%. The error of the measured ^{208}Pb/^{206}Pb ratio is magnified for less radiogenic Pb, as much as four times for a 1:1 mixture of a 4567 Ma radiogenic Pb and primordial Pb. The contribution from the measured ^{206}Pb/^{204}Pb varies with the value of this ratio: a 1% error in the measured ^{206}Pb/^{204}Pb adds to the ^{208}Pb*/^{206}Pb* ratio errors of 0.02%, 0.23%, and 1.9%, for ^{206}Pb/^{204}Pb values of 1000, 100, and 20, respectively. For highly radiogenic Pb, this source of uncertainty is trivial even if the ^{206}Pb/^{204}Pb error is rather large. The Pb isotopic composition assumed in subtraction of the "common Pb" component has little effect on the calculation of the ^{208}Pb*/^{206}Pb* ratio. For example, using a primordial composition for a sample that contains modern "common Pb" (or vice versa) adds an uncertainty of 0.1% for ^{206}Pb/^{204}Pb=100, and 0.8% for ^{206}Pb/^{204}Pb=20. The total error of the ^{208}Pb*/^{206}Pb* ratio is, therefore, usually around 0.5%, and possibly much smaller, for the samples with ^{206}Pb/^{204}Pb ≥100.

(2) The contribution from the uncertainty of the age to the calculation of κ_{Pb} is typically very small: 0.06% for a 10 Ma error in the age, and negligible for more precise modern age determinations.

(3) The errors arising from making a single-stage approximation of a multi-stage U-Th-Pb evolution vary depending on the magnitude and the timing of fractionation, and the nature of the element that migrates. Multi-stage evolution can be revealed by monitoring concordance of the U-Pb system, preferably by plotting multiple analyses in a concordia diagram. Ancient loss or gain of U would affect κ_{Pb}, κ_m as well as ^{206}Pb*/^{238}U and ^{207}Pb*/^{206}Pb* dates, and could create patterns difficult to interpret. Fortunately, this process, revealed by variability of ^{207}Pb*/^{206}Pb* and a non-zero lower concordia intercept, is rarely observed in modern studies of meteorites. Ancient loss of Pb seriously disturbs the U-Pb dates, but has only a small effect on κ_{Pb}. A loss of half of the Pb around 2000-3000 Ma ago would cause ca. 2.5% shift in κ_{Pb}, while for more ancient or more recent Pb loss the shift is even smaller. Recent migration of U changes κ_m and ^{206}Pb*/^{238}U, but does not affect κ_{Pb} and ^{207}Pb*/^{206}Pb*. Recent loss of Pb changes ^{206}Pb*/^{238}U, but not the other three parameters. A discordance of the U-Pb dates, therefore, does not necessarily mean that κ_{Pb} is disturbed. Ancient migration of U appears to be the only process that seriously undermines the accuracy of κ_{Pb}.

Two other possible processes are not considered above. First is the gain of Pb (released from other parts of the system) of unknown isotopic composition. It cannot be described by the classical U-Th-Pb multistage formalism [6] that requires continuity of the Pb isotopic composition between the stages. Since Pb is likely to be more mobile than U in processes such as thermal metamorphism, shock metamorphism, and aqueous alteration in reducing conditions, the multistage model is inadequate for describing these processes, and should be replaced by a more general model that considers migration of Pb caused by secondary processes. The second process is migration of Th without migration of U and Pb, but this is unlikely since Th is considered the least mobile among these elements.

κ_{Pb} vs. κ_m: In a mineral or rock that remained closed for migration of U, Th and Pb, the κ_{Pb} and κ_m

values are identical. However, U can be mobile in terrestrial environments in the presence of free oxygen, as well as during the lab treatment by acids and oxidizing agents such as HNO_3. Recent mobility of U compromises κ_m but not κ_{Pb}. The consistency between κ_m and κ_{Pb} confirms that the mineral was closed to migration of U and Th. If the U-Pb dates in this mineral are discordant, then the discordance is caused by recent migration of Pb.

Applications of κ_{Pb} to interpretation of the age data: The κ_{Pb} helps to distinguish mineral phases that we date. This is especially important for interpretation of the analyses of extensively leached rocks and minerals. Acid leaching removes large portion of U, Th and Pb, but it is not obvious whether these elements are extracted selectively, or brought into solution by congruent dissolution of minerals. Variations in κ_{Pb} and the degree of discordance among leachates and residues help distinguishing between these cases. For example, the first leachates (in cold 0.5M HNO_3) from multiple fractions of the angrite Sahara 99555 yielded κ_{Pb} between 3.5-3.8 and nearly concordant U-Pb dates, whereas acid-leached whole rocks yielded κ_{Pb} between 7.3-7.7, and slightly discordant U-Pb dates [7]. These results suggest that the first leaching caused congruent dissolution of a U–bearing mineral, such as Ca-phosphate or a silico-phosphate. In this case, the first leachate Pb-Pb isochron date of 4560.9 ± 1.7 Ma is probably meaningful, reflects closure of the U-Pb system in these minerals, and can potentially be used to study the thermal history of the angrite parent body.

κ_{Pb} in CAIs: The κ_{Pb} values determined in CV CAIs are summarized in Fig. 1:

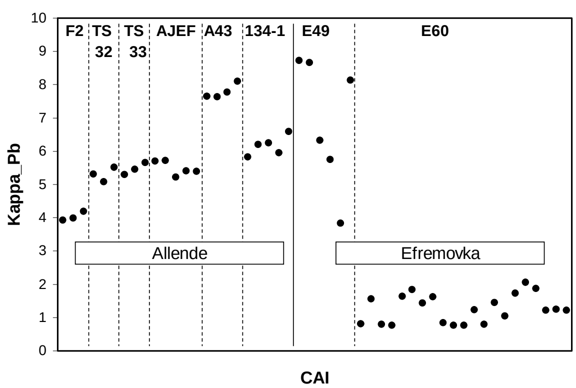

Fig.1: $^{232}Th/^{238}U$ ratios in CV CAIs, calculated from radiogenic Pb isotopic compositions (unpublished data of the authors). Types of CAIs and analyzed minerals are: F2 – type B2, Pl, Mel, Px; TS32 – compact type A, Pl, Mel, Px; TS33 – type B1, Pl, Mel, Px; AJEF – type B, Px, Mel, Px washes; A43 – type B or CTA, Px and bulk; 134-1 – compact type A, bulk and Mel; E49 – compact type A, bulk, Mel ± Sp; E60 – forsterite-bearing type B, bulk and various minerals.

These data show that the variations of κ among the minerals in a single CAI (with exception of E49, and perovskites from CAI 134-1 (not shown), which yielded κ_{Pb} values between 1.6-5.7), are much smaller than the variations between different CAIs. This fact suggests that the most significant fractionation between Th and U occurred before formation of the CAIs.

κ_{Pb} and κ_m in regolith chondrite breccias: A comparison of κ_{Pb} and κ_m in regolith chondrite breccias may help in understanding this uncommon meteorite type. Composed of chondrite fragments, lithic fragments, feldspar and sulfide grains, rare CAIs, and, in several cases, at one time containing water, these breccias appear to be assemblages of debris sampling the early stages of planetary accretion. Thirty fragments of chondrites, feldspars, and sulfides from six meteorites have been analyzed for U and Pb concentration and Pb isotopic composition, and about half of the fragments were also analyzed for Th concentration ([8], and unpublished data of the authors). When Th concentrations were measured κ_{Pb} and κ_m values can be compared, and constraint placed on the timing and extent of open system gain and loss of U, Th, and Pb. The close agreement of all Pb-Pb dates at ca. 4.6 Ga requires that disturbances causing U-Pb age discordances of up to 90% took place in relatively recent time. Whereas $^{206}Pb/^{204}Pb$ and $^{238}U/^{204}Pb$ values of the fragments vary widely between 9.41 and 43.54, and between 0.09 and 28.6, respectively, κ_{Pb} and κ_m values cluster between 2.9 and 5.7 (between 3.4 and 4.4 for chondrite fragments) and most agree well between methods. No simple relationship is observed between either $^{238}U/^{204}Pb$ or κ and degree of discordance for the chondrite fragments. If their excess radiogenic Pb was acquired on the parent body, or as contamination by terrestrial Pb, a Th/U different from that which produced the *in situ* Pb within the fragment might be expected; however, such demonstrably different κ_{Pb} and κ_m values have not yet been found.

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