to track megafauna abundance, Gill *et al*. (*15*) showed that herbivore populations collapsed shortly before the onset of vegetation change and increased fire. The same chain of events may have occurred earlier in Australia, but over a time span too fine to be resolved by current dating methods. Recovery of ancient DNA from sediments has also revealed that mammoths and horses survived much later in North America than indicated by the fossil record, owing to the improbability of finding and dating fossils of the last survivors, especially in dwindling populations (*16*). Application of these types of approaches in Australia may further refine the timing of the events leading up to extinction and provide sharper insights into the likely drivers of this ecological catastrophe.

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17. The authors thank the artist, P. Trusler, and the Australian Postal Corporation for permission to reproduce the painting shown in the first figure. The original work is held in the National Philatelic Collection.

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GEOCHEMISTRY

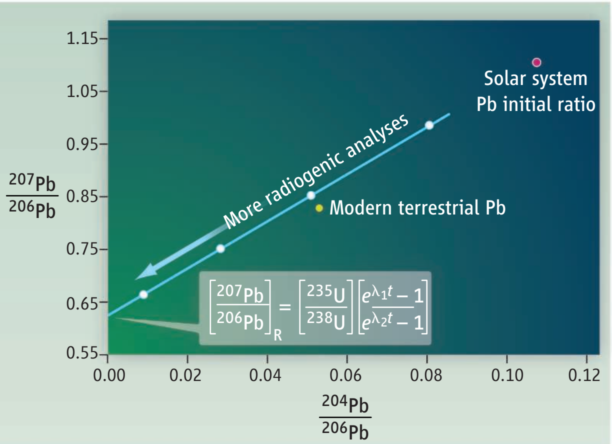
Adjusting the Solar System's Absolute Clock

The recent demonstration that a basic assumption in using isotope decay to measure the age of meteorites is incorrect has profound implications for dating the early solar system.

J. N. Connelly

Meteorites and their components provide the only means to study the circumstances and processes that gave rise to our solar system. But the task of unraveling our origins is by no means straightforward. A single undifferentiated meteorite—a chondrite—typically contains matrix and inclusions formed over a wide range of conditions and time before eventually being accreted into a single body. Understanding this complex assemblage, especially over the critical first 10 million years, allows the formulation of models of the spatially and temporally evolving thermochemical conditions that gave rise to the solar system. Geochronology—the determination of ages of events recorded by meteorites—provides the requisite temporal information. Although advances have been made over the past decade in this field, on page 449 of this issue, Brennecka *et al*. (*1*) present data suggesting that a basic assumption for the use of the U-Pb chronometer in geochronology, the golden spike for deep time, may be incorrect.

Two types of chronometers are used to measure the ages of ancient meteorites. So-called relative chronometers are based on the rapid decay [half-lives <10 million years (My)] of now extinct, short-lived radioisotopes that were created either just before or during the formation of the solar system. These relative chronometers, such as ^{41}Ca-^{41}K, ^{26}Al-^{26}Mg, ^{53}Mn-^{53}Cr, ^{60}Fe-^{60}Ni, and ^{182}Hf-^{182}W, require that the initial abundances of the parent radioisotope are known and that it was homogeneously distributed throughout the solar system. The short half-lives of these extinct nuclides have the potential to provide the most precise age estimates for the first 10 My. But the recent discovery of large-scale isotopic variability of nucleosynthetic origin for elements of contrasting volatility among different meteorite groups (*2*) casts doubts on the assumption of homogeneous distribution of short-lived radioisotopes.



Setting a date. In the inverse Pb-Pb diagram, the radiogenic $^{207}Pb/^{206}Pb$ ratio [$(^{207}Pb/^{206}Pb)_R$] can be calculated by projecting a line through data points with variable mixtures of radiogenic Pb and initial Pb to the *y* intercept, where the initial Pb is theoretically zero. This variability is created by strategically analyzing related fragments, minerals, or acid leachates with variable U/Pb ratios. (Subscript R refers to radiogenic; λ_1 and λ_2 represent the decay constants of ^{235}U and ^{238}U, respectively; and *t* represents time.)

Absolute chronometers are based on long-lived radioisotopes so that the present-day parent-daughter ratio in a sample provides an age in years before present with no assumptions necessary regarding the initial inventory of the parent radioisotope or its homogeneous distribution. Of the absolute chronometers, only the U-Pb system has half-lives and systematics suitable for resolving events in the first 10 My of the solar system. In addition to its high resolution, the U-Pb system is unique in a second way. Two isotopes of U, ^{235}U and ^{238}U, break down spontaneously at different rates (half-lives of 0.704 and 4.47 billion years) to produce ^{207}Pb and ^{206}Pb, respectively.

If it is accepted (and it has been) that the $^{238}U/^{235}U$ of all objects is 137.88, then one may calculate an absolute age knowing only the ratio of radioactively produced ^{207}Pb and ^{206}Pb (see the figure). This so-called Pb-Pb method is advantageous, if not necessary, because late alteration and/or laboratory treatment to remove ubiquitous contaminant terrestrial Pb typically alters the U/Pb ratio so that the measured parent-daughter

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ratios do not reflect those of a closed system. Because the Pb isotopic ratios remain unaffected by late alteration or laboratory treatment, the Pb-Pb age remains accurate despite any shifts in the U/Pb ratio that may have recently occurred.

It has long been assumed that the solar system inherited a fixed inventory of galactically derived ^{238}U and ^{235}U that was homogeneously distributed in the protosolar molecular cloud and that they were not measurably fractionated in natural systems. All ages reported in the literature today are based on this assumed fixed $^{238}U/^{235}U$ ratio. The isotopic composition of U is typically not measured in samples owing to the technical challenge of measuring the extreme $^{238}U/^{235}U$ ratio of small amounts of U sufficiently precisely and the lack of evidence, despite prior attempts (*3*), that this ratio varies measurably in meteorites. But Brennecka *et al*. demonstrate that this assumption of a fixed U ratio is incorrect for some of the solar system's oldest solids, calcium-aluminum–rich inclusions (CAIs), and that the ratio may vary up to 3000 parts per million from the accepted value of 137.88. This translates into a potential age offset of 5 My for a given radiogenic ratio $(^{207}Pb/^{206}Pb)_R$—or about 50% of the life span of the solar protoplanetary disk. They attribute this heterogeneity to the variable fractionation of short-lived ^{247}Cm that decays to ^{235}U, by using Nd as a geochemical proxy for the now extinct Cm.

Ages from a relative chronometer can be mapped into absolute time in the past when a single object is found to be suitable for both a Pb-Pb age and a relative age. For example, CAIs anchor the ^{26}Al-^{26}Mg system (*4*), whereas the differentiated basaltic angrite LEW 86010 provides the anchor for the ^{53}Mn-^{53}Cr system (*5*, *6*). If the short-lived nuclides were homogeneous and the $^{238}U/^{235}U$ ratio was consistent, all ages from different chronometers for samples that behaved as a closed system should be concordant. But they are not. For example, age offsets of up to 3 My exist between Pb-Pb ages and the available relative chronometers for some rapidly cooled volcanic meteorites (*7*).

Homogeneity of short-lived nuclides in the disk has commonly been singled out as the least robust assumption in geochronology, from which one can infer that the relative chronometers are most likely in error. But it is now possible that variations in the $^{238}U/^{235}U$ ratio in meteorites and their components may, at least in part, be to blame for the discordances. However, so far the offsets in the $^{238}U/^{235}U$ ratio of CAIs reported by Brennecka *et al*. will only make the discordance between Pb-Pb ages and the relative chronometers worse.

With the rapidly growing identifications of planets that orbit distant stars, and the tantalizing perspective of discovering an Earth-like world, understanding the sequence of events leading to the formation of the planetary bodies in our solar system has never been so relevant. Brennecka *et al*. convincingly relate for the first time the important discovery that U is isotopically variable in CAIs, implying that the currently accepted age for the formation of the solar system's first solids (*4*) may be incorrect. At the same time, they have defined a new benchmark for high precision and accurate geochronology: All future Pb-Pb studies must include $^{238}U/^{235}U$ ratios. Only then can we be certain that we have an internally consistent Pb-Pb chronometric database and a correct temporal framework within which to interpret meteorites and, in turn, understand the origins of our solar system.

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MATERIALS SCIENCE

Epitaxial Growth Writ Large

The technological goal of optimizing the controlled deposition of atomic monolayers is simplified by studying models of deposition of larger colloids.

Theodore L. Einstein[1] and Timothy J. Stasevich[2]

The performance of semiconductors in device applications often depends on their crystallinity—the grain boundaries and defects of a polycrystalline material interfere with transport of charge carriers. Single crystalline layers can be grown through epitaxy: Atoms are deposited from the gas phase on top of an existing crystal to form new layers. However, if the growth process is not well controlled or is too rapid, unwanted surface features, such as mounds, may form. Thus, the fabrication process relies heavily on monocrystalline growth of a single element. Models to find optimal conditions for this process have been studied for a long time (*1*, *2*) and have had to become increasingly sophisticated (*3*, *4*). Insights from related processes involving molecules or even larger particles can test our understanding of how epitaxy works and can be easier to observe directly. On page 445 of this issue, Ganapathy *et al*. (*5*) describe epitaxial growth with colloidal spheres some four orders of magnitude larger than atoms. Models developed for atomic epitaxy can describe these processes, despite colloid-colloid attractions arising in a way very different from atomic interactions.

Models of epitaxial growth must account for how adsorbed atoms, called adatoms, interact with each other and the surface template, as well as the effects of different incoming fluxes *F* of atoms and different growth temperatures. Adatoms are trapped by attractive forces in wells of the corrugated surface potential but move when they have enough energy to "hop" over these barriers (see the figure, panel A). A minimal model describing how adatoms move requires attractions between neighboring atoms (such as chemical bonds) to create the potential wells and energy barriers to describe the hopping process. Analysis of the energy barriers helps in estimating the thermal surface diffusion coefficient *D*. The model must also account for the greater difficulty of atoms dropping over a step edge, because they must break even more bonds. In the simplest picture, this leads to the so-called "Ehrlich-Schwoebel" (ES) barrier (see the figure, panel B) (*6*, *7*).

For colloidal particles, attractive forces arise from a "depletion interaction." Smaller surrounding polymer "depletants" have a hard time getting between closely spaced colloids,

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