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$^{142}Nd/^{144}Nd$ IN BULK PLANETARY RESERVOIRS, THE PROBLEM OF INCOMPLETE MIXING OF INTERSTELLAR COMPONENTS AND SIGNIFICANCE OF VERY HIGH PRECISION $^{145}Nd/^{144}Nd$ MEASUREMENTS;

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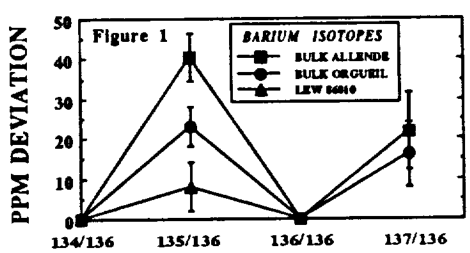
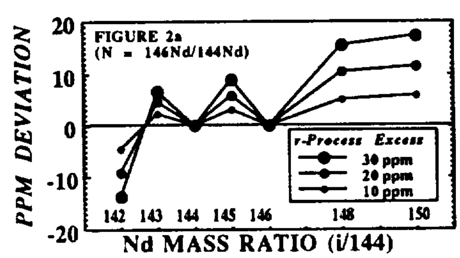
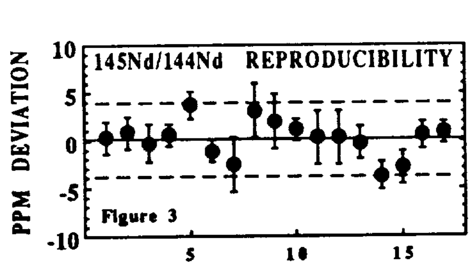
Introduction: Apart from the challenge of very high precision $^{142}Nd/^{144}Nd$ ratio measurement, accurate applications of the coupled $^{146,147}Sm$-$^{142,143}Nd$ systematics [1] in planetary differentiation studies require very precise knowledge of the present-day (post-^{146}Sm decay) $^{142}Nd/^{144}Nd$ ratios of bulk planetary objects (BP). The coupled systematics yield model ages for the time of formation of Sm/Nd-fractionated reservoirs by differentiation of Sm/Nd-unfractionated bulk planetary reservoirs [1,2,3,4]. Estimates of $(^{142}Nd/^{144}Nd)_{BP}$ and $(^{143}Nd/^{144}Nd)_{BP}$ therefore provide the critical baseline relative to which these model ages are referenced. In the ^{147}Sm-^{143}Nd systematics, $^{143}Nd/^{144}Nd$ variations are mostly large (>200 ppm), and therefore small (*viz.*, <20 ppm) variations in initial $^{143}Nd/^{144}Nd$ ratios generally can be ignored. However, in the case of ^{146}Sm-^{142}Nd, the range of $^{142}Nd/^{144}Nd$ divergence for differentiated planetary reservoirs is much smaller (<~100 ppm). Consequently $^{146,147}Sm$-$^{142,143}Nd$ model ages are sensitive to small variations in bulk planetary $^{142}Nd/^{144}Nd$ (both present-day and initial). One major unanswered question is whether or not Nd shelf standards (CIT Ndβ/Ames metal, La Jolla, NASA-JSC/Ames metal) have $^{142}Nd/^{144}Nd$ identical to the bulk Earth or otherwise might record some degree of radiogenic evolution in an early-fractionated reservoir. Our discussions of early Earth differentiation based on $^{142}Nd/^{144}Nd$ in Isua and Acasta samples [1,5] have employed a working assumption: $(^{142}Nd/^{144}Nd)_{Nd\beta} = (^{142}Nd/^{144}Nd)_{\text{Bulk Earth}}$. This requires experimental justification and is apparently contradicted by chondrite $^{142}Nd/^{144}Nd$ measurements [3,4], which have been interpreted to indicate: $(^{142}Nd/^{144}Nd)_{\text{JSC/Ames metal}} = ((^{142}Nd/^{144}Nd)_{CHUR} + 35\pm8$ ppm) [4]. At present, interpretations of the early Earth and Moon hinge largely on this issue. Because Ba in bulk chondrite samples exhibits similar magnitude nuclear anomalies [6], attributable to incomplete mixing of interstellar components, a critical question is whether or not nuclear effects are also present in $^{142}Nd/^{144}Nd$, both in bulk chondrites and between planetary objects. In general, $^{142}Nd/^{144}Nd$ variations between bulk planetary reservoirs could result from four possibilities: (i) *ab initio* ("nuclear") variation of initial $^{142}Nd/^{144}Nd$; (ii) *ab initio* variation of $(^{146}Sm/^{144}Sm)_{BP}$; (iii) variation in $(Sm/Nd)_{BP}$; and (iv) radiogenic evolution of $^{142}Nd/^{144}Nd$ in an Sm/Nd-fractionated "parental" reservoir prior to formation of the Moon putatively by giant impact. The first two possibilities result from incomplete mixing of interstellar components and the third from cosmochemical fractionation of the rare earths. Possibility (ii) is an unlikely problem because isotopic variability on the planetary scale due to incomplete mixing is a small effect---apparently <100 ppm in the Ba-REE region. At this level, initial $^{146}Sm/^{144}Sm$ heterogeneity will not be significant, either as a chronometric bias (from error in the $^{146}Sm/^{144}Sm$ ratio) or via the post-decay shift in bulk planetary $^{142}Nd/^{144}Nd$. (A 100 ppm shift in initial $^{146}Sm/^{144}Sm$ amounts to only a 28 ppb shift in $^{142}Nd/^{144}Nd$). A larger nebular heterogeneity scale in $^{146}Sm/^{144}Sm$ would be expected if ^{146}Sm was synthesized "locally" by protosolar flare spallation, but the low abundances of ^{53}Mn and ^{92}Nb in the early solar system preclude more than ~2% local spallogenic production of the *ab initio* ^{146}Sm budget [7]. By an indirect argument, planetary-scale Sm/Nd variability (possibility iii) also appears not to be a significant problem. The degree of moderately volatile/refractory element fraction between the Earth, Moon, Mars, EPB, bulk solar system and EPB, can be gauged from inferred bulk Mn/Cr ratios [*cf.*, 8]. The bulk Earth and Moon exhibit the strongest inferred Mn/Cr fractionations---about equal to bulk Allende. High precision measurements of $^{147}Sm/^{144}Nd$ in large homogenized samples of Allende [9,10] differ from the "CHUR" average ($^{147}Sm/^{144}Nd = 0.1967$ [9]) only by a very small fractionation factor ($^{147}Sm/^{144}Nd = 0.1964$; $f_{Sm/Nd} = -0.0015$). Assuming Sm/Nd variation between bulk planetary reservoirs to be limited to the range $f_{Sm/Nd} = \pm0.0020$, bias relative to CHUR will be less than a total range of 46 ppm and 1.1 ppm in ε_{143Nd} and ε_{142Nd}, respectively---which is not significant. Discussions of (iv) are critically dependent on a satisfactory evaluation of (i) and determination of an appropriate $^{142}Nd/^{144}Nd$ bulk Earth value from Sm/Nd-unfractionated meteorites, adjusted for nuclear effects. Here we show that the problem of planetary-scale nuclear effects can be satisfactorily addressed by coupling high precision $^{142}Nd/^{144}Nd$ measurements with $^{145}Nd/^{144}Nd$ measurements using the decomposition of Nd into s- and r-process components obtained from SiC studies [11].

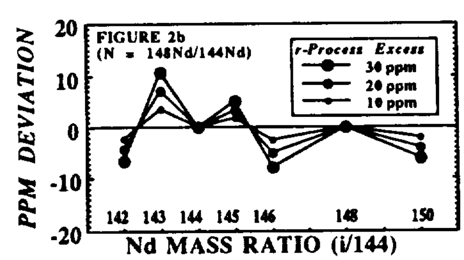
Nuclear Effects in Ba and Nd due to Incomplete Nebular Mixing: R-excess type anomalies in Ba, Nd and Sm isotopes are well-known from studies of the highly anomalous "FUN" inclusion EK1-4-1 [12]. More recently, similar effects at <50 ppm levels have been resolved in Ba in large bulk samples of Orgueil and Allende, and a strong hint of an anomaly at the ~8±6 (2σ) ppm level was observed in 4 high quality runs of the LEW 86010 angrite [6; Fig. 1]. The presence of isotopic anomalies in Ba in bulk chondrites and an achondrite indicates that similar effects are probably also present in Nd. Measurements of $^{142}Nd/^{144}Nd$ in Sm/Nd-unfractionated meteorites, therefore, probably do not provide an accurate measure of the CHUR/bulk Earth value without correction for nuclear differences relative to the bulk Earth. The data required for these corrections are: (a) a measure of nuclear effects in Nd and (b) the relative proportions of the r- and s-process contributions across the Nd masses. Modelling of r/s-type nuclear anomalies begins with a determination of the r-process excess parameters: $\eta_{r,i} = \varepsilon_i(N_{\Sigma\ r+s}/N_r)_i$, where ε_i is the abundance anomaly in the nuclide i (due to an excess in its r-process component) and N refers to atom number proportions [13]. It can be seen that abundance anomalies are demagnified in proportion to the fraction of s-process component in any nuclide for a given r-process excess. For self consistency, $\eta_{r,i}$ should agree for all masses of the same element. (Excesses may agree between nuclides of different elements, but this is not necessarily the case.) In practice, anomalies are determined in normalized ratios which involve a cumulate of the effects at 3 or 4 masses. However for Ba, an s-only mass-136 index and $^{134}Ba/^{136}Ba$ normalization can be utilized, and the magnitudes of the r-excesses are then obtained simply from the $^{135}Ba/^{136}Ba$ and $^{137}Ba/^{136}Ba$ ratio anomalies multiplied by $(N_{\Sigma\, r+s}/N_r)_i$ for masses 135 and 137. From the Mainz Murchison SiC s-process decomposition [11], we obtain multipliers of 1.2 and 2.0 for masses 135 and 137, respectively. The product of these and the measured anomalies yield self consistent r-excesses of 48±~10 ppm and 44±~14 ppm for the Allende $^{135}Ba/^{136}Ba$ and $^{137}Ba/^{136}Ba$ anomalies, and 28±~8 ppm and 32±~11 ppm for Orgueil (Fig. 1). An r-excess of 10±8 ppm is inferred from the LEW 80610 $^{135}Ba/^{136}Ba$ data. Figures 2a and 2b show the expected anomaly patterns in $^{i}Nd/^{144}Nd$ for r-excesses of 10, 20 and 30 ppm, normalized to $^{146}Nd/^{144}Nd$ and $^{148}Nd/^{144}Nd$, respectively, using the Mainz Nd decomposition: {$(N_{\Sigma\, r+s}/N_r)_i$: (142, 143, 144, 145, 146, 148, 150) = (1.00/0.00, 1.44±0.01, 2.15±0.09, 1.33±0.01, 2.21±0.03, 1.07±0.01, 1.00)} [11]. It can be seen that the signs of the $^{142}Nd/^{144}Nd$ anomalies are negative for both normalizations and that their magnitudes are reduced by factors of 2.2 and 4.5, respectively, w.r.t. the r-excess. The -35±8 ppm deviation in $^{142}Nd/^{144}Nd$ relative to the NASA standard (identical to Ndβ) reported by [4] for bulk chondrites corresponds to excesses of 77 ppm relative to $^{146}Nd/^{144}Nd$ and 1.6 ε-units relative to $^{148}Nd/^{144}Nd$.

Monitoring Nuclear Effects with Very High Precision $^{145}Nd/^{144}Nd$ Measurements: Figure 2 also shows that r-excesses in the 10-30 ppm range produce small anomalies of 3-9 ppm in $^{145}Nd/^{144}Nd$. If these small effects can be resolved, appropriate corrections can be applied to $^{142}Nd/^{144}Nd$. Larger effects will be present in $^{148}Nd/^{144}Nd$ and $^{150}Nd/^{144}Nd$ for the $^{146}Nd/^{144}Nd$ normalization, but these ratios are difficult to measure to high precision. Figure 3 shows reproducibility for 17 dynamic mode $^{145}Nd/^{144}Nd$ measurements of Ndβ (7) and terrestrial samples (10), exponentially normalized to $^{146}Nd/^{144}Nd$ = 0.724134. $2\sigma_p$ of these data is ±3.9 ppm ($2\sigma_m$ = ±0.95 ppm, about a mean value of $^{145}Nd/^{144}Nd$ = 0.34894096), demonstrating a capability for resolving shifts in $^{145}Nd/^{144}Nd$ down to 2 ppm, 2σ, with quadruplicate measurements. This corresponds to a ±3 ppm control on r *vs.* s polarization-type anomalies in $^{146}Nd/^{144}Nd$-normalized $^{142}Nd/^{144}Nd$ ratios due to incomplete nebular mixing. Applications of this method to chondrites, eucrites, lunar rocks and SNC meteorites will provide strong constraints on the bulk planetary $^{142}Nd/^{144}Nd$ values and contribute significantly towards understanding early planetary differentiation processes.









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