PLATINUM IN PRESOLAR NANODIAMOND RESIDUES – AN AMS APPROACH. U. Ott[1], S. Merchel[1,2], K. Knie[3], G. Korschinek[3], Th. Faestermann[3], G. Rugel[3] and A. Wallner[3]. [1]Max-Planck-Institut für Chemie (Otto-Hahn-Institut), Becherweg 27, D-55128 Mainz, Germany (ott@mpch-mainz.mpg.de); [2]present address: BAM, D-12205 Berlin, Germany; [3]Technische Universität München, Physik Department E15, D-85747 Garching, Germany.

Summary: Platinum isotopes have been measured in presolar nanodiamond residues from the Allende and Murchison meteorites using accelerator mass spectrometry (AMS). We infer an upper limit to the abundance of ^{198}Pt-H of ~1x10^{14} atoms/g, which is on the order of what is expected from an extrapolation of the apparently mass dependent abundance trend of the associated noble gases.

Introduction: Presolar nanodiamonds are the most enigmatic type of presolar grain found in primitive meteorites (e.g., [1,2]). The inference about a presolar origin of at least part of them only rests on the isotopic analysis of trace elements such as noble gases (e.g., [3,4]). However, typically just one out of a million diamond grains contains an atom of these, and it is not possible to quantify the fraction of diamonds that are truly presolar. The major hints come from elements that are located in the Solar System abundance distribution on the first two r-process abundance peaks associated with magic neutron numbers N=50 (Kr) and N=82 (Te, Xe) [3-6]. It seemed worthwhile, therefore, to search for correlated effects in elements of the third peak (N=126) such as platinum.

Experimental: AMS was chosen to determine the isotopic composition of platinum in two diamond residues, in order to avoid the potential problem of molecular and isobaric interferences. While this is also well achieved by RIMS, which also can have considerably higher detection efficiency (e.g. [7]), AMS is more easily adapted to measurement of a new element, and it seemed worthwhile to explore its applicability. Moreover, since unlike the case of SiC and graphite, diamonds cannot be analyzed individually anyway, detection efficiency is of lesser importance in their study.

The experimental set-up was similar to that used for the measurement of actinides [8]. Diamonds were mixed with high purity Ag powder and pressed in Ag cathodes, which were introduced into a sputter ion source. Negative Pt ions were extracted and, after a first magnetic mass selection, injected into the tandem accelerator, running at a terminal voltage of 9.5 MV. The stripping process in the accelerator's terminal provides an entire molecular breakup, thus any molecular background can be rejected on the high-energy side of the AMS system. After the accelerator, Pt ions in the 8^+ charge state were selected. To switch between the different Pt isotopes, the terminal voltage was adjusted for an equal magnetic rigidity of the ions on the high-energy side; on the low energy side the injection magnet's field was changed.

Two different types of standards were measured alongside the diamond residues: Pt metal and terrestrial Pt co-precipitated with AlO(OH) and ignited at 900°C to Al_2O_3 "contaminated with Pt".

Results and Discussion: Measured isotopic ratios are normalized to ^{198}Pt, with corrections applied only for the difference in yields in the stripping process [9] are shown in Fig. 1.

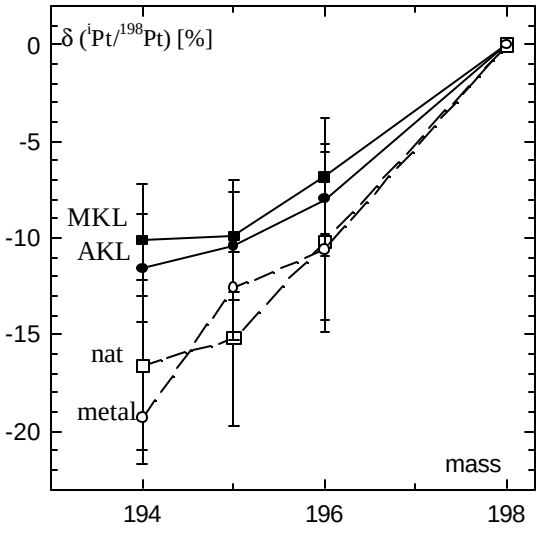

Fig. 1. Platinum isotopic composition (deviation from normal [12] in %) measured by AMS for two diamond residues and two terrestrial samples (see text).

Errors include both statistical errors and an estimated 2 % uncertainty of systematic effects, quadratically added (1 σ). Data are for two classically prepared samples (similar to [10]), one from Allende (AKL; 35 ppm Pt as determined by INAA [11]) and one from Murchison (MKL; <10 ppm Pt). In addition, results of the measurement of the two terrestrial standards are shown. Measured isotopic ratios - both of the meteorite and the terrestrial samples - differ significantly from the normal Pt isotopic composition [12] although they agree with each other within the estimated uncertainties. This is not quite unexpected in AMS. For example, in the Munich tandem accelerator system the terminal voltage has to be adjusted for each isotope, which leads to differences in transmission. Also, matrix dependent fractionation effects may occur, e.g. in the ion source. Interestingly the diamond

residue samples are much closer to the normal terrestrial values than the standards. They give almost identical values, one possible reason being that they were immediately run after each other, and instrumental and matrix-dependent effects in both cases were rather similar.

For estimating the amount of Pt-H possibly present in the nanodiamonds, a comparison of the two different meteoritic diamond samples is most useful. This is because the Pt concentration in AKL (35 ppm) is at least a factor of three higher than that in MKL (<10 ppm). Since dilution by extraneous material is so much lower in the case of MKL, the existence of an isotopic effect of sufficient size would (with the reasonable assumption that they are equal in diamonds from both meteorites) show up as a difference relative to AKL. As evident from Fig. 1, all ratios measured in the two samples nominally agree within 2 %. Agreement is within 3-4 % if our estimated systematic errors are taken into account, but this may be an overestimation since some of the systematic uncertainties must cancel if the comparison is made relative to a suitable standard. Overall, we estimate that 5 % is probably a safe upper limit to the overabundance of ^{198}Pt. With an upper limit for Pt in MKL of <10 ppm, this then translates into an upper limit for the abundance of ^{198}Pt of $<1.1 \times 10^{14}$ atoms/g diamond

This is about a factor 50 higher than the observed $\sim 2 \times 10^{12}$ atoms/g of excess ^{136}Xe. Nevertheless, it is an important constraint. While neighboring elements such as Te and Xe appear to be present in presolar nanodiamonds almost unfractionated relative to suggested production ratios [6], the noble gases overall suggest a trend of regularly increasing abundance with mass [11,13]. As shown by these workers, solar-normalized noble gas abundances in diamonds define an approximately straight line in a log (fractionation factor f relative to solar) vs. mass plot. Fig. 2 shows the corresponding trend based on the relative abundances of Ar, Kr, and Xe together with abundances of trace elements heavier than Kr determined in four of our samples by INAA [11]: the two classically prepared samples measured for platinum isotopes and two corresponding residues obtained by a different kind of chemistry, i.e. one designed to reduce the level of interfering PGE not hosted by the diamonds proper (AMW and MMW). Assuming the abundances for non-noble gas elements follow the same trend as the noble gases, the vertical distance from the line is an indication for the degree of "pollution". To guide the eye, the dashed lines in Fig. 2 show the effects of dilution by factors of 10 and 100, respectively. Assuming Pt-H is indeed more abundant than Xe-H by the factor implied from Fig. 2, we may just have missed seeing an isotopic effect in our AMS measurement. While some improvement of accuracy in AMS appears possible, it will be crucial to further reduce the abundance of non-diamond Pt in the samples to be analyzed. Only then will it be possible to arrive at a value (or upper limit) that will allow to test the predictions of the models devised to explain the H component in nanodiamond trace elements and to confirm or negate the abundance trend suggested by the noble gases and shown in Fig. 2.

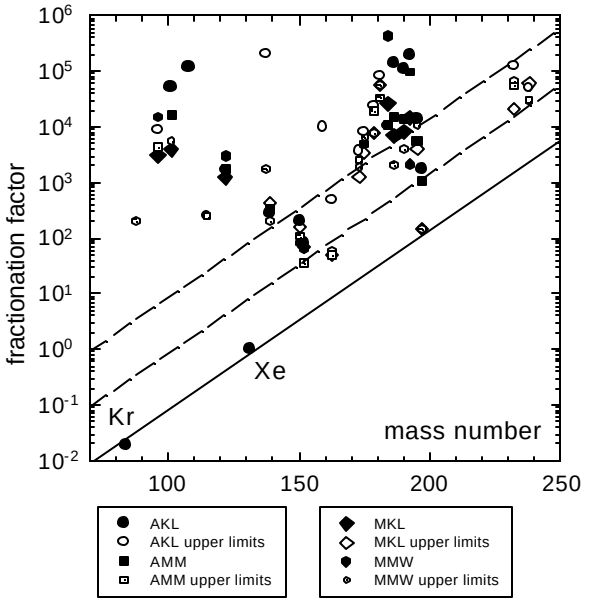

Fig. 2. Trace element abundances in nanodiamonds determined by INAA [11] compared to the abundance trend suggested by the noble gases Ar, Kr and Xe (solid line - see text). The dashed lines show the effect of dilution by extraneous material by factors 10 and 100, respectively.

References: [1] Zinner E. (1998) *Ann. Rev. Earth Planet. Sci. 26*, 147-188. [2] Hoppe P. and Zinner E. (2000) *JGR-Space Physics 105*, 10371-10385. [3] Lewis R.S. et al. (1987) *Nature 326*, 160-162. [4] Huss G.R. and Lewis R.S. (1994) *Meteoritics 29*, 791-810. [5] Richter S. et al. (1998) *Nature 391*, 261-263. [6] Maas R. et al. (2001) *Meteorit. Planet. Sci. 36*, 849-858. [7] Savina M.R. et al. (2003) *Geochim. Cosmochim. Acta*, in press. [8] Wallner C. et al. (2000) *Nucl. Instr. Meth. B 172*, 333-337. [9] Sayer R.O (1977) *Rev. Phys. Appl. 12*, 1543-1546. [10] Amari S., et al. (1994) *Geochim. Cosmochim. Acta 58*, 459-470 [11] Merchel S. et al. (2000) *Meteorit. Planet. Sci. 35*, A108-A109. [12] De Bièvre P. and Taylor P.D.P. (1993) *Int. Journ. Mass Spectr. Ion Proc. 123*, 149-166. [13] Ott U. et al. (1981) *Geochim. Cosmochim. Acta 45*, 1751-1788.