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RADIOACTIVE CHEMICAL KINETICS OF LARGE SUPERNOVA DUST. D. D. Clayton, B. S. Meyer and L.- S. The, Dept of Physics and Astronomy, Clemson Univ, Clemson SC 29634-0978

Supernovae are profoundly radioactive. Accordingly, we seek a new picture for radioactive regulation of the condensation of dust within their comoving, expanding, and cooling gaseous interiors. Such supernova condensates (SUNOCONs 1,2) from presolar galactic supernovae are recovered from meteorites (graphite, SiC) and are identified by predicted (3) excess ^{26}Mg , ^{41}K , and ^{44}Ca from post-event decay of ^{26}Al , ^{41}Ca and ^{44}Ti (4). Their isotopic compositions suggest mixed-shell material. This suggests two puzzles: 1. *Why are the discovered SUNOCONs so large?* 2. *Where are the expected more numerous ones from unmixed supernova shells?* We propose that the large sizes and the mineralized structures of SUNOCONs result from five controls: *population control, cooling, admixed seed grains, catalysis, and entropic arrow.* The constraints are driven by radioactivity and rapid decline of gas density, which maintains atomic abundances far from thermal equilibrium. Central to the carbide SUNOCONs is the CO molecule, whose disruption by Compton electrons (5) maintains free carbon that enables graphite or SiC growth (2).

Diffusive mixing at the molecular level is impossible for bulk supernova gases during the time available (about 2 years) for SUNOCON growth. Mixed compositions occur only along velocity-plume and penetrated-shell interfaces. That most mass cannot be mixed can be seen by considering dimensions of a supernova shell. The shell of thickness L contains about one solar mass of atoms, about 10^{56} atoms. Characteristic density at 200d is $n=10^{10}$ atoms cm^{-3} ; therefore the shell volume is 10^{46} cm^3 . Its radius R (at $v=2000$ km/sec) at 200d (when condensation begins) is 4×10^{15} cm, so its thickness L must be at least 10^{14} cm (several percent of shell radius) to account for its volume. Therefore, if diffusive mixing is to be effective, diffusion distances during a few years must be a significant fraction of $L=10^{14}$ cm. The diffusion root-mean-square distance can be estimated from random-walk theory: $\langle x^2 \rangle^{1/2} = D = l(N/3)^{1/2}$, where $l=(\sigma n)^{-1}$ is the mean free path, N is the number of scatters, $\sigma = 10^{-16}$ cm^2 is the scattering cross section and $n=10^{10}$ cm^{-3} is the number density near 200d. This yields mean free path $l=10^6$ cm. With thermal speed near 10^6 cm/sec, the time between collisions is 1 s, yielding $N=t$ for the number of collisions during time t . Thus $D=10^6 \text{cm} (t/3)^{1/2} = 5 \times 10^9$ cm for $t=3$ yr, by which time the condensation will effectively have been truncated by the t^{-3} time dependence of the atom density. One sees that the fraction of the shell that has been polluted by diffusion is $\bullet M/M = D/L = 5 \times 10^{-5}$. Therefore, SUNOCONs from the shell M might have been expected to be 10^4 times more abundant than those from the small mixed mass $\bullet M$. This is clearly not the case among the large ones, because the isotopes in both large graphite and SiC SUNOCONs seem to require matter from different shells in virtually all that have been discovered (4). We advance five kinetic controls for the large SUNOCONs:

Population Control: If the number of small grains is large,

they cannot grow to large size before depleting the condensible atoms. To grow large they must be rare. Since observed SUNOCONs contain 10^{11} to 10^{16} atoms, their seed nucleations must be limited to abundances less than 10^{-11} and 10^{-16} of the condensible atoms. *Population control* is affected through chemical reactions that suppress the abundance of molecular nucleations. Although counterintuitive, rapid destruction of nucleations may often be required in order to enable large final grain size. Lack of population control of Al_2O_3 nucleations has been seen (4) as probable explanation of the absence of large SUNOCONs of this type from the OSi shell, where Al bathed in O are both very abundant. The Al_2O_3 nucleations become too numerous and must remain small owing to their exhaustion of condensible atoms. A more subtle example of population control showed (2) that oxidation of C_n suppresses the number of graphite grains condensed, which allows each to finally grow to large size. The "CO trap" actually assists the growth of very large graphite particles, for without it one would have instead a very large number of small graphite particles. Analogous kinetic population-control factors may favor mixed-shell compositions. This is a kinetic phenomenon, not one of equilibrium. C and O are very much out of equilibrium owing to the CO Compton-electron dissociation in supernovae, causing free abundances of both C and O to be simultaneously large, which is not possible in equilibrium at 2000K.

Cooling rates: Even percent levels of CO create such effective cooling through $\bullet v=1$ and $\bullet v=2$ vibrational levels that ambient T is lowered from near 4500K to near 1800K at 200d (5). Such kinetic temperatures may represent the difference between instability and stability of dust. Earlier condensation produces bigger grains owing to the t^{-3} dependence of the gas density. Grain temperature is another difficult question because grains maintain cooler temperature than the gas, although they are attacked by hot atoms. The cooling effect of CO and population control together may explain the rarity of large SUNOCONs from the HeC shell (the only large C-rich mass in SNII). The He^+ ions (from radioactivity) destroy CO too rapidly for its abundance to be able cool the shell, so that C_n itself must be the coolant; furthermore less dominant oxidation in the HeC shell results in a larger abundance for C_n chain molecules, rendering growth to large size problematic. In similar way the SiO molecules (and subsequent grains) may cool the OSi shell, in which abundant O is fusing to Si at explosion time. Consider the abundance transition in the SiO shell near $n(\text{O}) = n(\text{Si})$, which are the two dominant abundances (6). The ratio O/C near 10^3 decimates C_n abundance by severe oxidation for about a year. The steady-state CO abundance remains a few percent of less abundant C, too few to cool this shell. Cooling here relies on the SiO vibrational transitions. A mass near $10^{-3} M_{\text{sun}}$ of SiO, as seen in SN1987A, can be produced in this shell (7). A change in the free O/C ratio occurs after SiO provides the first step to abundant

small Si_nO_m molecules, which seed SiO_2 and MgSiO_3 grain condensation into many small grains, partially depleting O and Si. During this transition the Si_nC_n molecules become increasingly more abundant, allowing SiC grains to condense in small number abundance, but that small number (population control) allows each to grow to micron size by depleting remaining Si and all C as SiC grains. The carbon atoms are alternately trapped as CO and liberated by fast electrons until depletion of O to O/C values less than 10. This OSi shell is very promising for a source of SiC-X grains in SNII, because its $^{28}\text{Si}/^{29}\text{Si}$ is only fourfold solar, ^{13}C renders the C only moderately ^{12}C -rich, $^{26}\text{Al}/^{27}\text{Al}$ is near 0.01, and it is ^{15}N -rich from the neutrino reactions on the O (6). Radioactive destruction of CO and population control may in this way conspire for kinetic growth of SiC-X in the O-dominated OSi shell. Silicate SUNOCON population was much larger, however, so that their final sizes became too small for detection within current automated scans of meteorite grains.

TiC seed grains: We calculated final mass fractions of key nuclei within the alpha-rich freezeout central zone. They are appropriate for condensation of both TiC grains and FeNi grains. ^{44}Ti is quite abundant, as is ^{12}C . The carbon abundance vastly exceeds that of O, so CO can not be a population control for carbides. The two Ti isotopes make it slightly more abundant than C. This zone, whose temperature may be cooled by FeNi nanograins and TiC molecular vibrations, will condense abundant TiC grains, exhausting Ti and C; but these will not be very large because they are too numerous owing to lack of population control. However, when turbulently admixed into condensing graphite in the CO shell or into the OSi shell, rapid growth of graphite or SiC-X containing both TiC and (Fe,Ni)C subgrains may result. Predicted is that these TiC are ^{44}Ca -rich (3) and the FeC is deficient in ^{54}Fe and ^{58}Fe (3). Dynamic importance of such seed grains may be seen by considering the rarity of nucleations for SiC-X condensation in the OSi shell. So severe is oxidation there that the abundance of Si_nC_n is held very low, perhaps too low for the number of large SiC grains to be significant. But when small TiC grains are admixed having sizes sufficient for C association to be faster than oxidation, they may deplete remaining C (along with Si) as SiC of large size.

Catalysis: Catalytic assistance to growth of large grains is the most difficult mechanism to be specific about; but its

possibility should be recognized. Both graphite and SiC SUNOCONs are N-rich, suggesting that ^{15}N may somehow catalyze the C growth in O-rich gas. If catalytic cycles using N can effectively enhance the C association rate (or reduce the O oxidation rate) for small C_n or Si_nC_n molecules, they increase the abundance of carbon molecules in conditions of severe oxidation. Molecule CN has greater binding than has C_2 , suggesting that it may play an important role at the highest (earliest) temperatures.

Entropic Arrow: These four kinetic constraints are far from obvious, and many workers have long regarded thermodynamic equilibrium as the explanation of these grains. Although that approach has good success for AGB stardust (8), it fails for SUNOCONs (2). Thermodynamic equilibrium is also unable to predict the sizes and compositions of SUNOCONs or the fraction of matter that can condense in the time available. However, the entropic thrust toward minimization of the free energy does sort adhered surface atoms on grains toward mineralic compositions. Our thesis that magnesian silicates and silicon carbide may grow *simultaneously* in the OSi zone, for example, requires the action of this principle to maintain integrity of those two mineral species. This aspect resembles the usual chemical equilibrium; but it is limited by the numbers of surface reactions that can occur. Our condensation picture factors into nonequilibrium molecular kinetics to establish nucleations for the growth of solids and subsequent entropic guiding of their constitutions, retaining C and Si preferentially in SiC grains and the O, Mg and Si in the silicates as atoms are resorted by surface reactions.

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