

EXTINCT ^{44}Ti IN PRESOLAR GRAPHITE AND SiC: PROOF OF A SUPERNOVA ORIGIN

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Received 1996 January 25; accepted 1996 February 20

ABSTRACT

Large excesses in ^{44}Ca , from the radioactive decay of short-lived ^{44}Ti , have been observed in four low-density graphite grains and five SiC grains of type X extracted from the Murchison meteorite. Titanium-46, ^{49}Ti , and ^{50}Ti excesses were also observed in several of these grains. Because ^{44}Ti is only produced in supernovae, these grains must have a supernova origin. Moreover, Si-, C-, N-, Al-, O-, and Ti-isotopic compositions of the grains require a Type II supernova source, and indicate extensive and heterogeneous mixing of different supernova regions, including the nickel core.

Subject headings: dust, extinction — nuclear reactions, nucleosynthesis, abundances — supernovae: general

1. INTRODUCTION

Several types of circumstellar dust grains preserved in primitive meteorites at the time of solar system formation have been recently isolated and identified. They include diamond (Lewis et al. 1987), SiC (Bernatowicz et al. 1987; Tang & Anders 1988; Hoppe et al. 1994a), graphite (Amari et al. 1990), refractory carbides (Bernatowicz et al. 1991, 1996), Al_2O_3 (Huss et al. 1994; Nittler et al. 1994), and silicon nitride (Hoppe et al. 1994c; Nittler et al. 1995). Although the diamonds (1.6 nm) and refractory carbides (5–200 nm) are too small to be analyzed individually, ion microprobe isotopic analyses of individual grains of the other types show a wide diversity of isotopic compositions among grains and provide a wealth of information on nucleosynthesis and the physical and chemical conditions of their stellar sources. Among these grains, SiC and low-density graphite have been extensively studied, primarily because many of them are large ($>1\ \mu\text{m}$) and have relatively high trace element contents, allowing isotopic analysis of several elements on individual grains by ion probe mass spectrometry.

A small fraction ($\sim 1\%$) of presolar SiC, named grains X, show isotopic signatures distinct from those of all other SiC. They have isotopically heavy N ($^{15}\text{N}/^{14}\text{N}$ up to 23 times solar), high inferred $^{26}\text{Al}/^{27}\text{Al}$ ratios (on the order of 10^{-1}), ^{13}C depletions and enrichments, and excesses in ^{28}Si (up to 4 times solar) (Amari et al. 1992; Nittler et al. 1993, 1995; Hoppe et al. 1994b). In their original X-grain study, Amari et al. (1992) also found ^{49}Ti excesses in four grains X and in one grain, X2, a $^{44}\text{Ca}/^{40}\text{Ca}$ ratio 4 times as high as the solar value. They proposed a supernova origin for these grains. Many low-density graphite grains have Si-, C-, and N-isotopic signatures similar to those of the SiC grains X, and in addition large ^{18}O excesses up to 200 times solar (Amari, Zinner, & Lewis 1995b). To explain the isotopic ratios of these graphite grains, Zinner et al. (1995) investigated mixing between different zones in models of presupernova stars. They could successfully reproduce many of the isotopic ratios of the grains, although a

few discrepancies between the observed and predicted ratios remain (see Nittler et al. 1995 for a more detailed discussion).

Here we report large ^{44}Ca excesses in several SiC grains of type X and low-density graphite grains from the Murchison meteorite. These excesses are most likely due to the in situ decay of short-lived ^{44}Ti ($T_{1/2} = 58\ \text{yr}$), an isotope that is produced in the deep interiors of supernovae. The presence of ^{44}Ti in the above types of stellar dust in meteorites is the strongest evidence that these grains are supernova condensates and provides further evidence for deep and heterogeneous mixing in supernovae. We note that the possible existence of supernova condensates with large ^{44}Ca excesses from ^{44}Ti decay was predicted by Clayton (1975).

2. RESULTS AND DISCUSSION

We have previously reported Si, C, N, and Al isotopic ratios for SiC grains X from the Murchison 2–4 μm separate KJG (Nittler et al. 1995), and the same ratios plus those of O, Ca, and Ti for low-density graphite grains from the Murchison separates KE3 and KFA1 (density range $1.65\text{--}2.1\ \text{g cm}^{-3}$) (Amari et al. 1995b; Amari et al. 1995c). The grains were analyzed by secondary ion mass spectrometry, using previously described techniques (Zinner, Tang, & Anders 1989; Hoppe et al. 1994a). Although small grain sizes and/or low concentrations precluded Ca and Ti isotopic measurements in most of the grains, we succeeded in measuring these elements in a few SiC grains X (^{40}Ca , ^{44}Ca , and all Ti isotopes) and graphite grains (^{40}Ca , ^{42}Ca , ^{43}Ca , ^{44}Ca , and Ti isotopes). Five SiC grains and four graphite grains have large excesses in ^{44}Ca , relative to the solar $^{44}\text{Ca}/^{40}\text{Ca}$ ratio. In addition, four ^{44}Ca -enriched grains have ^{49}Ti excesses, two grains have ^{50}Ti excesses, and one grain has a ^{46}Ti excess. Isotopic data for the ^{44}Ca -enriched grains, including the previously reported data for SiC grains X2 (Amari et al. 1992) and X57 (Hoppe et al. 1996), are presented in Table 1. (See also Amari et al. 1995b and Nittler et al. 1995.)

Nucleosynthesis theory predicts that ^{44}Ca is produced in stars both by slow n -capture during He burning and from the radioactive precursor ^{44}Ti . Neutron capture, however, is predicted to produce even larger excesses of ^{42}Ca and ^{43}Ca than of ^{44}Ca . Since the $^{42}\text{Ca}/^{40}\text{Ca}$ and $^{43}\text{Ca}/^{40}\text{Ca}$ ratios in the graphite grains and in SiC grain X2 are normal within $2\ \sigma$ errors, the excess ^{44}Ca in these grains must be due to the in situ decay of ^{44}Ti . Moreover, several of the grains have ^{44}Ca excesses far greater than those predicted for n -capture (up to 14 times

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TABLE 1
ISOTOPIC DATA FOR GRAPHITE AND SiC GRAINS WITH ^{44}Ca EXCESSES
A.

Grain (Type) ^a	$^{12}\text{C}/^{13}\text{C}$	$^{14}\text{N}/^{15}\text{N}$	$^{16}\text{O}/^{17}\text{O}$	$^{16}\text{O}/^{18}\text{O}$	$^{26}\text{Al}/^{27}\text{Al}$
KE3a-321 (G)	136 ± 1	215 ± 10	916 ± 190	15.7 ± 0.8	0.104 ± 0.001
KE3a-322 (G)	147 ± 2	161 ± 9	2635 ± 312	57.9 ± 1.8	0.125 ± 0.001
KE3c-242 (G)	6.91 ± 0.02	257 ± 10	n.m. ^b	491 ± 28	0.0018 ± 0.0003
KFA1f-302 (G)	116.6 ± 0.5	398 ± 49	1962 ± 279	86 ± 4	<0.0008
KJGM2-66-3 (S)	74.7 ± 2.7	85.7 ± 2.5	n.m.	n.m.	0.136 ± 0.005
KJGM2-243-9 (S)	43.8 ± 0.9	20.5 ± 0.7	n.m.	n.m.	0.110 ± 0.060
KJGM2-290-2 (S)	49.0 ± 0.8	71.1 ± 1.9	n.m.	n.m.	0.155 ± 0.003
KJGM4-205-12 (S)	103 ± 1	88.8 ± 1.5	n.m.	n.m.	0.010 ± 0.009
KJGM4-271-3 (S)	26.1 ± 0.2	29.2 ± 0.3	n.m.	n.m.	0.173 ± 0.006
KJH X2 (S) ^c	304 ± 3	106 ± 1	n.m.	n.m.	0.23 ± 0.03
KJD X57 (S) ^d	n.m.	n.m.	n.m.	n.m.	0.11 ± 0.03
Solar	89	272	2610	499	...

B.

Grain	$\delta^{29}\text{Si}/^{28}\text{Si}$ (‰) ^e	$\delta^{30}\text{Si}/^{28}\text{Si}$	$\delta^{42}\text{Ca}/^{40}\text{Ca}$	$\delta^{43}\text{Ca}/^{40}\text{Ca}$	$\delta^{44}\text{Ca}/^{40}\text{Ca}$
KE3a-321	-214 ± 37	-314 ± 33	30 ± 19	72 ± 41	76 ± 14
KE3a-322	-120 ± 27	-179 ± 17	34 ± 33	69 ± 57	53 ± 23
KE3c-242	197 ± 149	77 ± 149	-28 ± 73	7 ± 114	194 ± 60
KFA1f-302	-272 ± 20	-349 ± 18	365 ± 440	2069 ± 1119	137063 ± 7824
KJGM2-66-3	-130 ± 11	-205 ± 12	n.m.	n.m.	2143 ± 566
KJGM2-243-9	-448 ± 16	-535 ± 17	n.m.	n.m.	2200 ± 849
KJGM2-290-2	-235 ± 8	-309 ± 11	n.m.	n.m.	371 ± 94
KJGM4-205-12	-310 ± 11	-419 ± 12	n.m.	n.m.	319 ± 19
KJGM4-271-3	-410 ± 11	-568 ± 13	n.m.	n.m.	4547 ± 849
KJH X2	-459 ± 2	-454 ± 3	-167 ± 80	390 ± 170	3040 ± 237
KJD X57	-747 ± 16	-475 ± 33	n.m.	n.m.	~19000

C.

Grain	$^{44}\text{Ti}/^{48}\text{Ti}$	$\delta^{46}\text{Ti}/^{48}\text{Ti}$	$\delta^{47}\text{Ti}/^{48}\text{Ti}$	$\delta^{49}\text{Ti}/^{48}\text{Ti}$	$\delta^{50}\text{Ti}/^{48}\text{Ti}$
KE3a-321	0.00106 ± 0.00022	22 ± 11	-5 ± 11	296 ± 13	n.m.
KE3a-322	0.0024 ± 0.0011	78 ± 27	-4 ± 26	576 ± 36	n.m.
KE3c-242	0.098 ± 0.038	16 ± 230	-271 ± 196	-4 ± 268	n.m.
KFA1f-302	0.00809 ± 0.00053	118 ± 10	26 ± 10	443 ± 12	205 ± 12
KJGM2-66-3	0.017 ± 0.005	267 ± 214	29 ± 239	978 ± 333	631 ± 303
KJGM2-243-9	0.06 ± 0.03	-391 ± 643	925 ± 714	-733 ± 589	-298 ± 600
KJGM2-290-2	0.0033 ± 0.0008	52 ± 84	26 ± 62	77 ± 81	318 ± 92
KJGM4-205-12	~0.024	-40 ± 56	-57 ± 49	25 ± 42	-55 ± 37
KJGM4-271-3	0.018 ± 0.004	108 ± 140	126 ± 156	153 ± 171	43 ± 167
KJH X2	0.159 ± 0.017	82 ± 48	23 ± 50	239 ± 58	-52 ± 38
KJD X57	0.37 ± 0.11	n.m.	n.m.	n.m.	n.m.

^a G indicates graphite; S indicates SiC.

^b n.m.: not measured.

^c Amari et al. 1992.

^d Hoppe et al. 1996.

^e $\delta A/B \equiv [(A/B)_{\text{Sample}}/(A/B)_{\text{Terr}} - 1] \times 1000$.

solar in the 25 M_{\odot} model of Meyer, Weaver, & Woosley 1995). One such graphite grain, KFA1f-302, has a $^{44}\text{Ca}/^{40}\text{Ca}$ ratio 138 times the solar ratio. SEM examination of this grain showed that it contains small ($\sim 0.5 \mu\text{m}$) subgrains of TiC (Fig. 1 [Pl. L4]). These observations suggest that essentially all of this grain's ^{44}Ca comes from ^{44}Ti , which had condensed into the TiC subgrains. Although we did not measure ^{42}Ca or ^{43}Ca in the new KJG SiC grains X, their ^{44}Ca is probably due to ^{44}Ti as well. Using the Ti/Ca ratios measured in the grains, we infer initial $^{44}\text{Ti}/^{48}\text{Ti}$ ratios between 0.001 and 0.06 (Table 1 and Fig. 2). The true $^{44}\text{Ti}/^{48}\text{Ti}$ ratio for SiC grain KJG-205-12 is uncertain because of the presence of unknown amounts of Ca and/or Ti from a nearby hibonite ($\text{CaAl}_{12}\text{O}_{19}$) grain on the sample mount.

Titanium-44 is predicted to be produced in the innermost

layers of supernovae, by α -rich freeze-out from nuclear statistical equilibrium in Type II and Ib supernovae and by explosive He-burning in sub-Chandrasekhar mass models of Type Ia (see, e.g., Woosley & Weaver 1995a; Timmes et al. 1996; Woosley, Taam, & Weaver 1986; Woosley & Weaver 1994). Iyudin et al. (1994) reported the detection of ^{44}Ti γ -rays by the COMPTEL telescope on the *Compton Gamma Ray Observatory* (CGRO) from the supernova remnant Cas A. Searches by the OSSE telescope (also on CGRO) did not confirm the detection, however (The et al. 1995; Timmes et al. 1996). Estimates of the ^{44}Ti production in SN 1987A predict a γ -ray line flux below the detection limit of current instruments, but there is evidence that ^{44}Ti decay powers the bolometric light curve after 1500 days (Timmes et al. 1996). The SiC grains X and graphite grains have extremely high inferred initial

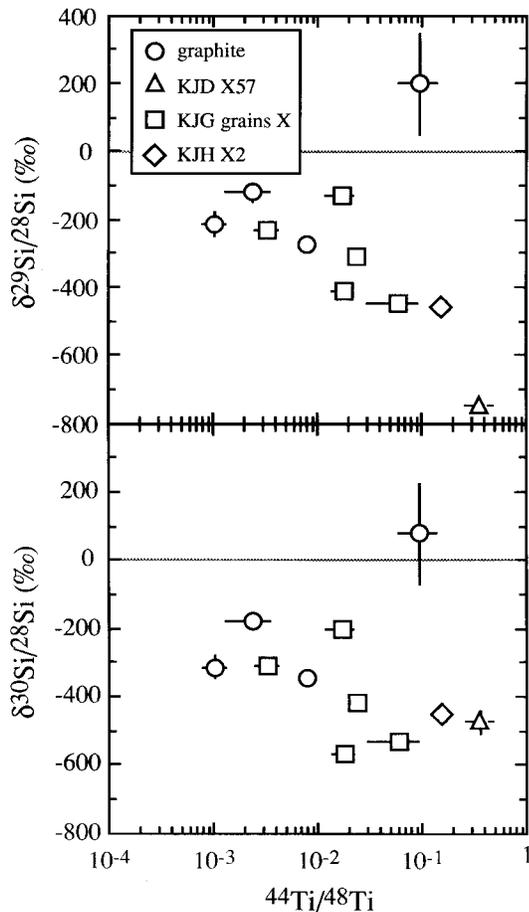


FIG. 2.—Si isotopic ratios plotted as δ -values (see Table 1 for definition), vs. inferred $^{44}\text{Ti}/^{48}\text{Ti}$ ratios for ^{44}Ca -enriched graphite and SiC grains from Murchison separates KE3, KFA1, and KJG. Also shown are previously reported data for SiC grains KJH X2 (Amari et al. 1992) and KJD X57 (Strebel et al. 1995; Hoppe et al. 1996). Most of the grains have excesses in ^{28}Si , relative to the heavier Si isotopes (negative δ -values). The $\delta^i\text{Si}$ -values generally become more negative with higher $^{44}\text{Ti}/^{48}\text{Ti}$ ratios, consistent with an origin of the ^{44}Ti in the ^{28}Si -rich inner zones of Type II supernovae.

$^{26}\text{Al}/^{27}\text{Al}$ ratios, and several have ^{13}C enrichments. These compositions point to a contribution of H-burnt material, and large ^{18}O excesses in the graphite require a massive star origin (Amari et al. 1995b). Thus, an origin in Type I supernovae, which are deficient in H (all Type I) and are believed to originate from low-mass objects (Type Ia), is ruled out for these grains. This leaves supernovae of Type II as the most likely source, provided that the ^{44}Ti can be mixed with carbon-rich layers where graphite and SiC can form. Here we use supernova models of Woosley & Weaver (1995a) to qualitatively explain the isotopic compositions of the grains and put constraints on supernova mixing. These models predict isotopic and elemental yields from different zones within the exploding massive star; following Meyer et al. (1995), we label these zones by their most abundant species. For example, the He/N zone contains the products of H-burning, the He/C zone contains the products of partial He-burning, and so on. Since we have previously discussed in detail the Si, C, N, O, and Al compositions of these grains and the associated agreements and discrepancies with models (Nittler et al. 1995), here we will primarily concentrate on the Ca and Ti

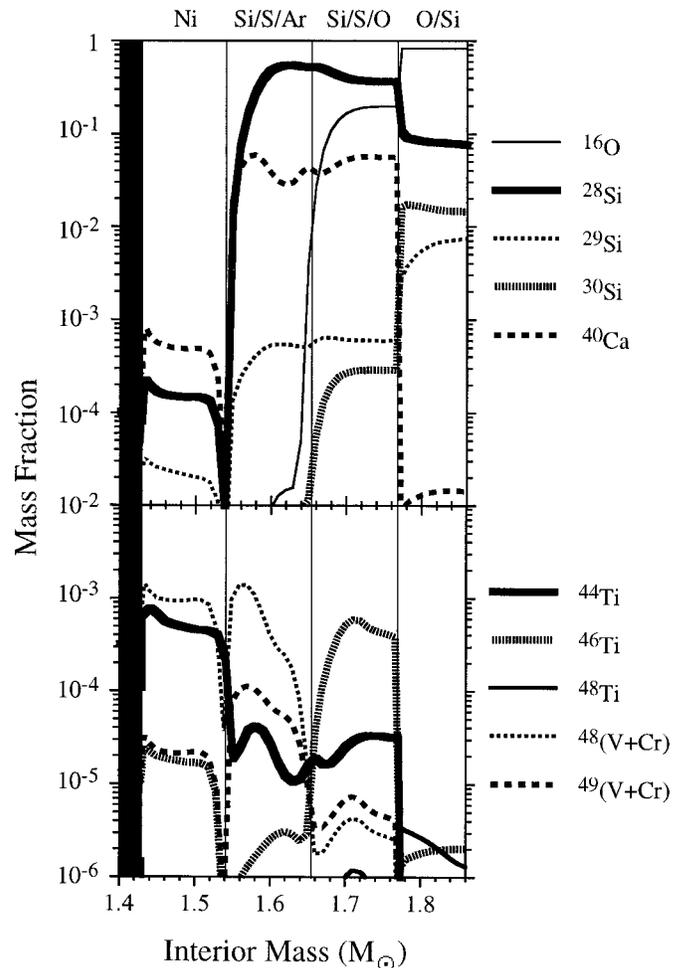


FIG. 3.—Predicted mass fractions of selected isotopes in the innermost layers of a $15 M_{\odot}$ supernova model (Woosley & Weaver 1995a). ^{44}Ti is abundant in the Ni core and in the Si/S/Ar and Si/S/O zones.

isotopic compositions. We note, however, that all supernova models still have many uncertainties. These include uncertain reaction rates, in particular that of $^{12}\text{C}(\alpha, \gamma)^{16}\text{O}$ and the weak interaction rates important for the latest stages of nucleosynthesis and core collapse, as well as the treatment of convection and mixing. In addition, the nucleosynthesis of many heavier isotopes (including ^{44}Ti) depends strongly not only on the nuclear reaction rates employed, but also on the details of the explosion itself (Woosley & Weaver 1995b; Thielemann, Nomoto, & Hashimoto 1995).

Figure 3 shows the compositions predicted for the inner layers of a $15 M_{\odot}$ star (Woosley & Weaver 1995a). ^{44}Ti is present in the core and in the overlying ^{28}Si -rich (from O-burning) zones. With the exception of the unusual graphite grain KE3c-242, all of the grains with inferred ^{44}Ti also have large ^{28}Si excesses (Fig. 2), consistent with a common origin of the two isotopes in these zones. In order to have ^{44}Ti and ^{28}Si excesses in a region with $\text{C}/\text{O} > 1$, required for graphite and SiC formation, some material from the inner zones must be mixed with material from the C-rich He/C zone. Low $^{14}\text{N}/^{15}\text{N}$ and $^{16}\text{O}/^{18}\text{O}$ ratios are predicted for this zone as well. To explain the high $^{26}\text{Al}/^{27}\text{Al}$ ratios and enrichments in ^{17}O and ^{13}C observed in many of the grains, material from the H-burnt He/N zone must also be included in the mix. However, such

mixing must be quite selective to avoid too large a contribution from the O-rich zones intermediate between the He/C zone and the Si zones, which would tip the C/O balance in favor of oxygen. Observations of supernova SN 1987A do provide evidence for extensive mixing of the ejecta (Shigeyama & Nomoto 1990), and infrared excess indicate grain formation in the ejecta a year after the explosion (Colgan et al. 1994). Also, hydrodynamical calculations of supernova explosions predict macroscopic mixing of different regions due to shock-driven Rayleigh-Taylor instabilities, resulting in clumps of material with distinct chemical compositions (Müller, Fryxell, & Arnett 1991; Herant & Woosley 1994). These calculations, at least qualitatively, show that the mixing is nonhomogeneous. It is possible to have lumps from interior zones injected into exterior zones without having all the intermediate material homogeneously mixed, just as required by the grain data to avoid an overabundance of oxygen. It remains to be seen, however, whether future hydrodynamical simulations carried to smaller scales and three dimensions will be able to quantitatively reproduce the microscopic mixing required to explain the grain data.

From the observed ^{46}Ti and ^{49}Ti excesses in the grains we can make rough estimates how much of the ^{44}Ti excesses come from the Si/S/O, Si/S/Ar, and Ni zones, respectively. Titanium-44 from the first of these zones would be accompanied by large amounts of ^{46}Ti (see Fig. 3), and ^{44}Ti from the Si/S/Ar zone by ^{49}V and ^{49}Cr that decay into ^{49}Ti . Chromium-49 ($T_{1/2} = 42$ minutes) has certainly decayed into ^{49}V ($T_{1/2} = 337$ days) before any grains form, but the latter could still be present at the time of grain formation. An uncertainty that enters here is possible fractionation between the chemical elements V and Ti during their deposition into graphite or SiC grains. The uncertainties become even larger, because of fractionation between Ti and Si, if we want to obtain additional constraints from the Si isotopic compositions (^{44}Ti from

the Si/S/O and Si/S/Ar zones is accompanied by almost pure ^{28}Si).

Most grains with evidence for ^{44}Ti have ^{49}Ti excesses. These could be from either n -capture or the decay of ^{49}V . If V is not fractionated from Ti, which at least for SiC generally seems to be the case (Amari et al. 1995a), grains with $^{44}\text{Ti}/^{48}\text{Ti}$ ratios larger than 0.05 would have $\delta^{49}\text{Ti}$ -values larger than 1000‰ if the ^{44}Ti comes from the Si/S/Ar layer of a $15 M_{\odot}$ supernova. Similarly, contributions from the Si/S/O zone are ruled out for all grains with $^{44}\text{Ti}/^{48}\text{Ti} > 0.01$, since they would have $\delta^{46}\text{Ti} > 300$ ‰. Constraints are even more stringent for supernovae of higher mass. This means that, at least for the grains with the highest $^{44}\text{Ti}/^{48}\text{Ti}$ ratios, a substantial fraction of the ^{44}Ti has to come from the Ni-rich core of the star. This conclusion is probably also true for grain KJD X57 because the $^{44}\text{Ti}/^{48}\text{Ti}$ ratio is high enough in the core, but not in the Si/S/Ar zone. However, because ^{46}Ti data are lacking for this grain, a contribution from the Si/S/O zone cannot strictly speaking be excluded, although it would have resulted in $\delta^{46}\text{Ti}$ -value of more than 10,000‰. Finally, a strong fractionation between Ca and Ti must have taken place because both Si/S zones are extremely rich in ^{40}Ca (Fig. 3) and ^{44}Ca excesses in the grains would be negligible if ^{44}Ti from these zones were accompanied by proportionate amounts of ^{40}Ca . Indeed, in at least some graphite grains, the Ti is present in the form of TiC subgrains, and trace element measurements in mainstream (i.e., not type X) SiC grains have shown that Ca is strongly depleted relative to Ti (Amari et al. 1995a).

We acknowledge the key role of E. Anders in developing the procedures used to isolate presolar dust in meteorites. L. N. thanks NASA for a GSRP fellowship. This work was supported by NASA grants NAGW 3371, NAGW 2525, NAG5 2843, and NSF grant 91-15367.

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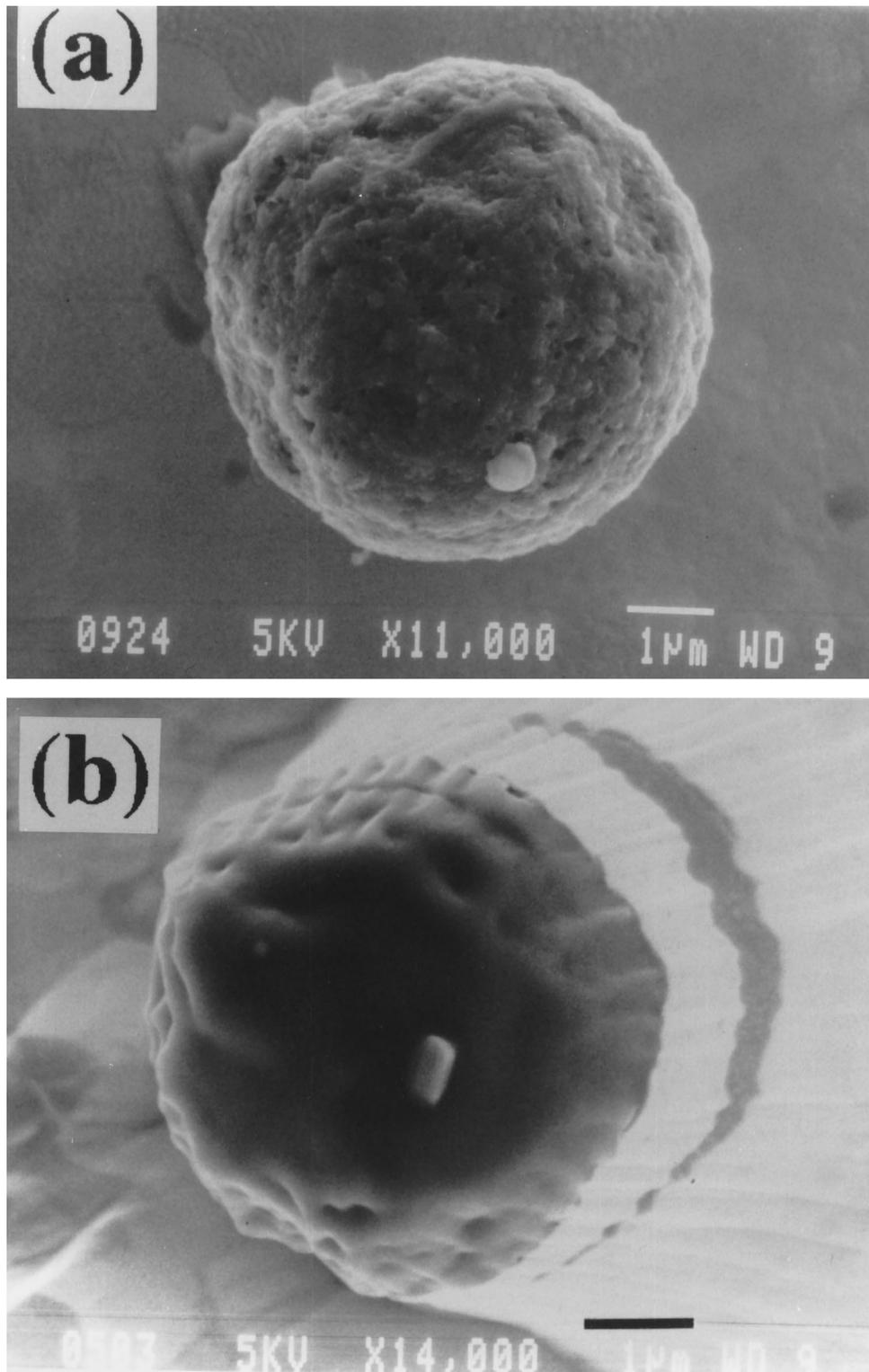


FIG. 1.—Scanning electron micrographs of the ^{44}Ca -enriched graphite grain KFA1f-302, before (a) and after (b) sputtering in the ion microprobe. Removal of the grain's surface layers revealed a $\sim 0.5\ \mu\text{m}$ subgrain of TiC (visible in b), suggesting that the large excess of ^{44}Ca observed in this grain originated as the radioactive progenitor ^{44}Ti , which condensed into TiC.