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Tantalum Carbide Formed by the Carbonization of Ta_2O_5 in an Argon Plasma Arc

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Synopsis. The formation of tantalum carbide by the carbonization of Ta_2O_5 with graphite by heating in an argon plasma arc was studied. Chemical and X-ray analyses showed that the product obtained was either stoichiometric or a defect carbide with the formula of TaC_{1-x}. The products were found to be superconductors.

Tantalum carbide is known to exist as from $TaC_{0.99}$ to $TaC_{0.85}$.[1] Giorgi *et al.*[2] found that the superconducting transition temperature decreased from 9.7 K to below 2.04 K as the carbon content decreased from 49.5 to 46.5 at%. In this case, tantalum carbide samples were prepared by heating mixtures of the elemental powders in a graphite crucible at 1850 °C under a vacuum of 10^{-5} Torr, with reaction times varying from 2 to 24 hr.

The present authors[3] previously formed niobium carbide by the carbonization of Nb_2O_5 with graphite by heating in an argon plasma arc and found that the product was a superconductor. Tantalum carbide was obtained by the carbonization of Ta_2O_5 with graphite in the same way. The products were confirmed, their properties, such as the density and the superconducting transition temperature, will be described in this note.

Ta_2O_5 (purity above 99.9%; H. C. Stark, Berlin) and graphite (spectroscopic grade; Tokai Electrode Mfg. Co.) were mixed in the desired molar ratio (C/Ta_2O_5). About one gram of the mixture was pressed into the tablet. Purified argon was used as the plasma gas. The plasma arc furnace and heating procedure were the same as have been described previously.[3,4] The temperature of the sample in the plasma arc was about 2900 °C. Heating was continued for 3 min. The products were investigated by X-ray diffraction and chemical analysis. The density of the products was measured picnometrically. The superconducting transition temperatures of some products were measured by means of 21 Hz mutual-inductance bridge and a Ge thermometer.

The chemical analysis data of the products with various C/Ta_2O_5 values are given in Table 1, while the X-ray analysis data of the products given in Table 2. The product obtained by $6.5 \leq C/Ta_2O_5 \leq 7.0$ was either stoichiometric TaC or a defect carbide with the formula TaC_{1-x}. Tantalum carbide, which has an fcc crystal structure, had a homogeneity range from 50.0 at% C to 43.5 at% C. The sum of the Ta and C contents of the products reached 100 wt%. It seems that only a little oxygen is present in the products in each case.* The lattice parameter of tantalum carbide, a, gradually increased with an increace in the carbon content. The product containing 50 at% C had a lattice parameter of 4.455 Å, which was comparable to that of the $TaC_{1.0}$ formed by the carbonization of metallic tantalum.[5]

TABLE 2. X-RAY ANALYSIS DATA OF PRODUCTS LISTED IN TABLE 1

Sample No.	Identified compounds	a of TaC (Å)
1	TaC, Ta_2C	4.419
2	TaC	4.421
3	TaC	4.435
4	TaC	4.441
5	TaC	4.448
6	TaC	4.450
7	TaC	4.455
8	TaC, C	4.456

TABLE 1. CHEMICAL ANALYSIS DATA OF PRODUCTS BY THE CARBONIZATION OF VARIOUS C/TaO IN AN ARGON PLASMA ARC

Sample No.	C/Ta_2O_5	Ta content		Combined C content		Free C content		$\sum$ (Ta, C) (wt %)
		(wt %)	(at %)	(wt %)	(at %)	(wt %)	(at %)	
1	6.4	96.5	58.5	4.46	41.5	—	—	100.1
2	6.5	95.2	56.5	4.48	43.5	—	—	100.1
3	6.6	94.4	53.0	5.56	47.0	—	—	100.1
4	6.7	94.2	52.0	5.77	48.0	—	—	100.0
5	6.8	94.1	51.1	5.95	48.9	—	—	100.1
6	6.9	94.0	50.5	6.04	49.5	—	—	100.0
7	7.0	93.8	50.0	6.22	50.0	—	—	100.0
8	7.1	93.0	46.6	6.18	46.7	0.89	6.70	100.1

* In the NbC formed by the carbonization of Nb_2O_5, the oxygen content in the product was found to be 0.008 wt% by radioactivation analysis.[3]

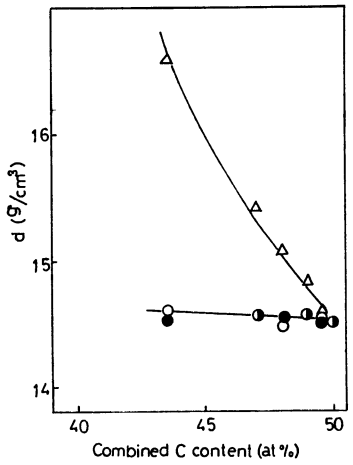

Fig. 1. Density *vs.* combined carbon content.
○: measured value, ●: calculated value assuming that the carbon position was vacant, △: calculated value assuming that the carbon position was substituted by Nb.

The relation between the density of the product at 25 °C and the combined carbon content is shown in Fig. 1. The density was about 14.5 g/cm³ for the product containing 50 at% C. This value agreed with the value calculated for stoichiometric TaC from the X-ray diffraction data. The density for TaC_{1-x} also agreed with that determined by means of X-ray diffraction, if we assumed that some carbon positions in the TaC lattice were vacant.

The relation between the superconducting transition temperature, T_c, of the product and the combined carbon content is shown in Fig. 2. The highest transition temperature was reached for the stoichiometric composition; it was 10.1 K. T_c decreased from 10.1 K to less than 4.2 K as the carbon content decreased from 50 to 47 at% C. This is similar to the decrease observed for tantalum carbide made by the carbonization

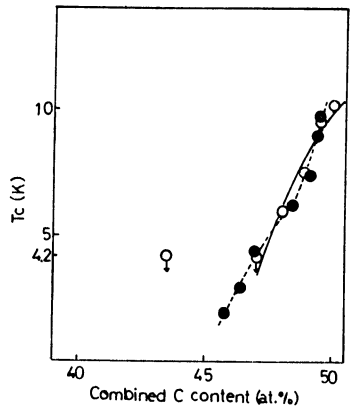

Fig. 2. T_c *vs.* combined carbon content.
○: this experiment, ●: Giorge *et al.*[2]

of metallic tantalum.[2]

The small amount of oxygen contained in the carbide remarkably decreases the T_c.[6] The oxygen content of the product formed in this work seems to be small. Therefore, it may be concluded that increasing the number of vacant carbon sites in the TaC lattice decreases the T_c. This conclusion is also supported by the results of the density measurement.

References

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