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Authors
Wells, Molly
Pickus, Milton
Kennedy, Kurt
et al.

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Inorganic Materials Research Division, Lawrence Radiation Laboratory
and Department of Mineral Technology, University of California, Berkeley

The critical temperatures of superconducting intermetallic compounds can be empirically related to parameters such as crystal structure, composition, electron concentration and lattice parameter, as shown by Matthias et al.[1] In a recent review paper, Roberts[2] clearly demonstrated that for a given crystal structure the critical temperature is related to the electron concentration per unit volume. It is not obvious, however, that similar correlations exist for another large class of superconductors, viz., the transition metal-nonmetal compounds of the cubic (NaCℓ) crystal structure. Although some of the highest critical temperatures reported have been for compounds of this class, the relevant literature is scarce and often contradictory. A program to study systematically the carbides and the nitrides of the transition metals has been initiated at this laboratory.

The critical temperatures of solid solutions of TaC and NbC are reported in this paper. This system was chosen since Ta and Nb are in the same column of the periodic table and are, therefore, electronically similar; their sizes are identical in the elemental state; the interstitial element, carbon, is common to both compounds; both compounds are known to be superconductors; and they form a continuous series of solid solutions.

The powders of TaC and NbC, obtained from Fansteel Metallurgical Corporation, were ball-milled, leached with HCℓ to remove iron, and washed in distilled water to remove soluble chlorides. The powders were mixed by dry

tumbling and extruded with polystyrene as a binder as rods of 1/8" diameter. The polystyrene was then volatilized in purified helium at 375°C and the samples were given an initial four-hour firing in vacuum at 1350°C. The rods were then sintered for 60 hours at 2100°C. Homogenization was verified by means of x-ray diffraction. Critical temperatures were measured using the Meisner effect. Samples of TaC and NbC, processed in the same manner as the mixed powders, exhibited the same critical temperatures as the original unsintered powders indicating that no significant contamination or change in carbon content occurred in the processing.

The variation of critical temperature with composition is shown in Fig. 1. The critical temperature has a broad maximum in NbC-rich solutions. Also, at a given composition, the transition from normal to superconducting behavior is spread over a range of three to four degrees. Giorgi et al.[3] and Schröder[4] have shown that the critical temperature of transition metal carbides or nitrides is strongly dependent on stoichiometry. Giorgi, in a private communication, has suggested that the width of the transition zone at a given composition is also inversely related to stoichiometry. The possibility exists then that the maximum in critical temperature is a consequence of a variation in combined carbon. This is not the case, as shown by the linearity of the lattice parameter composition plot, shown in Fig. 2. The lattice parameters[5,6] and critical temperatures of the pure compounds correspond to the formulae $TaC_{.98}$ and $NbC_{.96}$. The agreement with Vegard's Law for the solid solutions proves that the stoichiometry was not inadvertently changed during processing.

A maximum in critical temperature has been reported for the NbC-MoC[7] system. Both NbC and MoC are superconductors forming cubic solid solutions up to 80% MoC.

In a system closely analogous to TaC-NbC, viz., HfN-ZrN, Giorgi et al.[8] have reported a linear variation of critical temperature and lattice parameter with composition. Both systems, TaC-NbC and HfN-ZrN, have metallic elements of the same size and electronic configuration, and a common interstitial element. Each system forms a continuous series of solid solutions which obey Vegard's Law and each system is composed of superconducting compounds. Yet, one exhibits a maximum in critical temperature while the other does not. It can be concluded from the limited data available that the parameters so successfully applied to intermetallic compounds, i.e., electron-to-atom ratio or electrons per unit volume, are insufficient criteria in the case of solid solutions of transition metal-interstitial element compounds.

Acknowledgment

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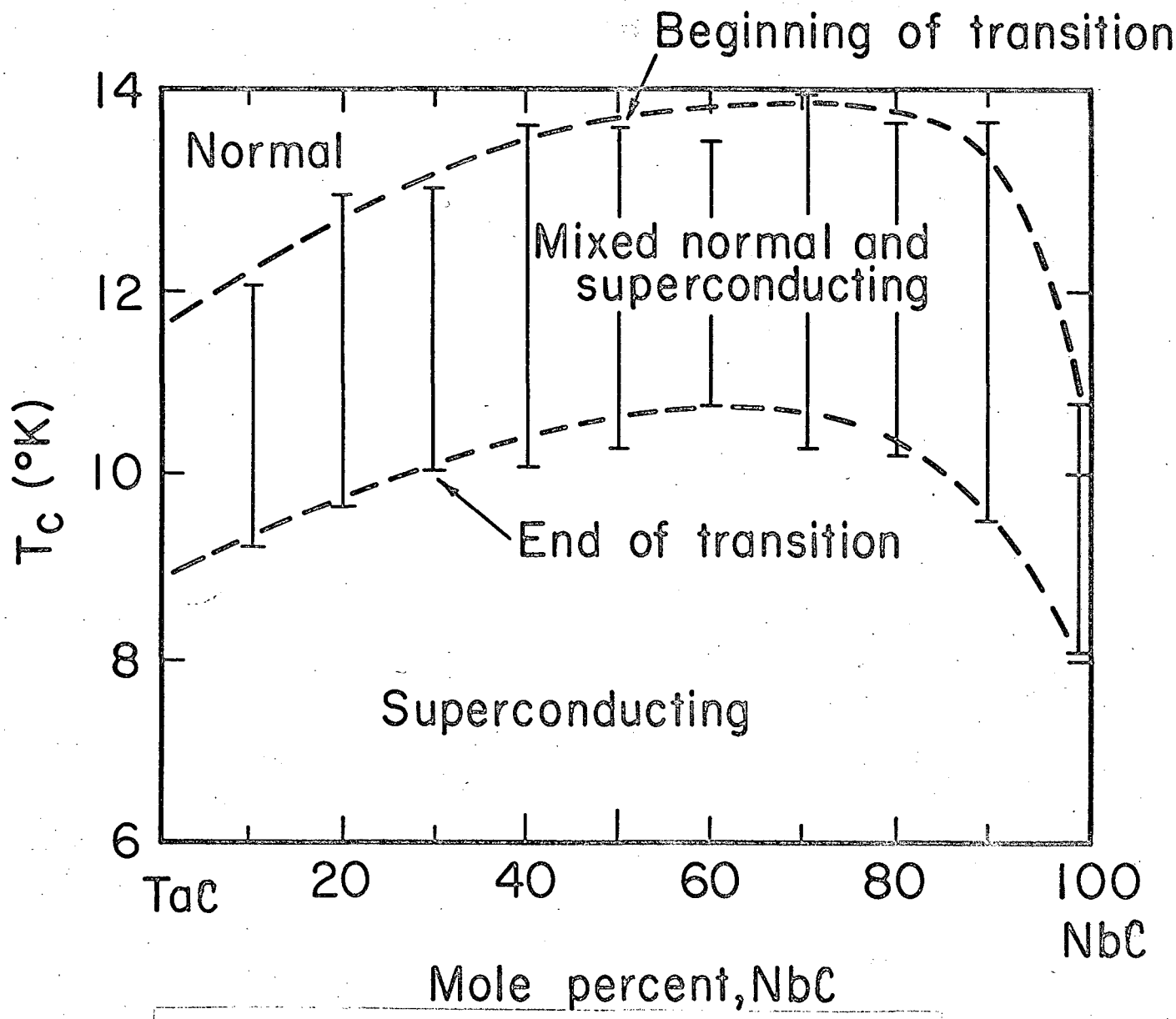

Fig. 1. The variation of critical temperature, T_c, with composition for the system TaC-NbC.

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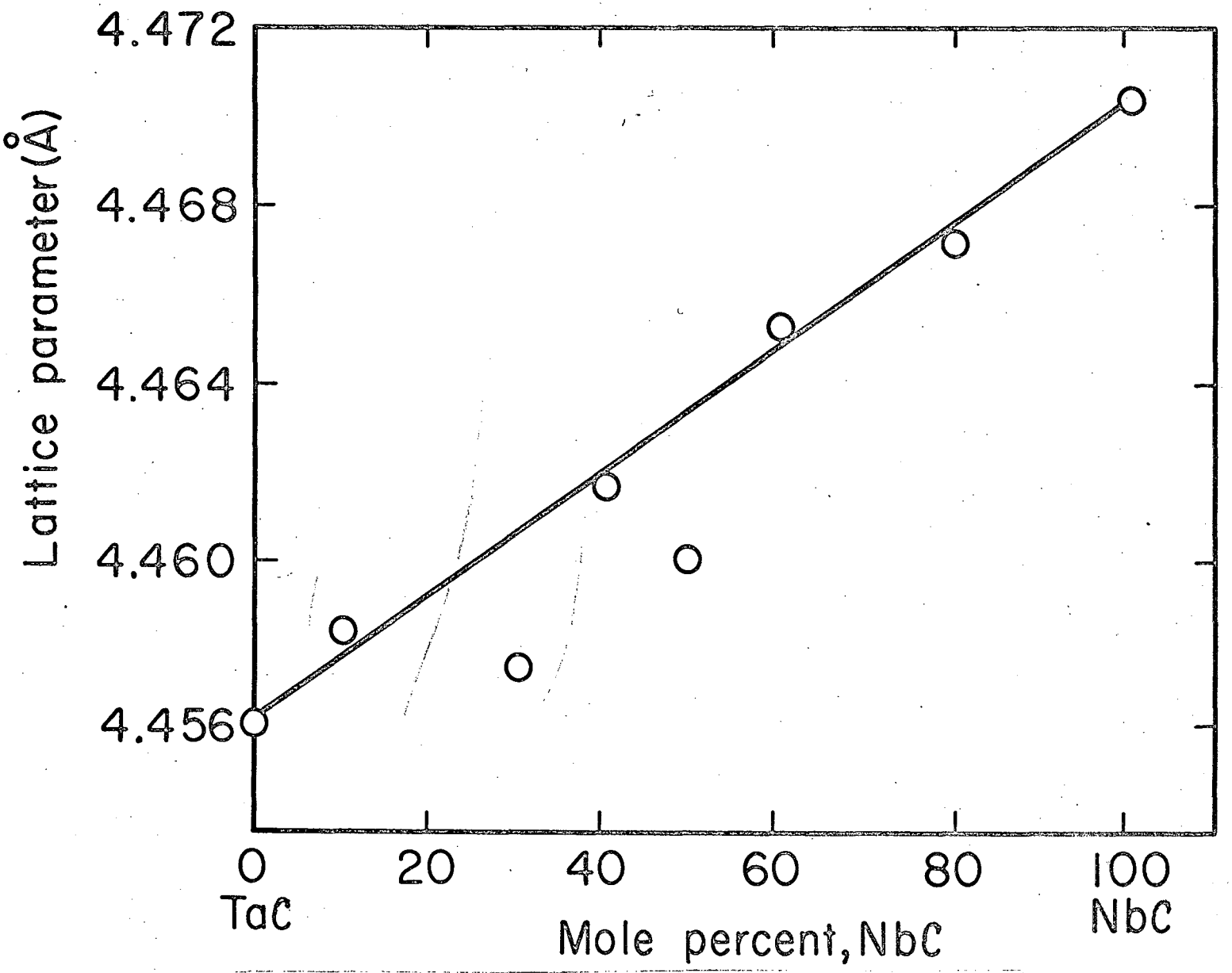

Fig. 2. The variation of lattice parameter with composition for the system TaC-NbC.

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