

**Synthesis and superconductivity of intermetallic compounds  $Y_2Ni_x B_{8-x} C_2$ ,  $YNi_x Cu_{2-x} B_2C$ , and  $YNi_x Cu_{2-x} Si_2C$**

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# Synthesis and superconductivity of intermetallic compounds $Y_2Ni_xB_{8-x}C_2$ , $YNi_xCu_{2-x}B_2C$ , and $YNi_xCu_{2-x}Si_2C$

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Borocarbide and silicocarbide intermetallic  $Y_2Ni_xB_{8-x}C_2$  ( $x=1, 2, 3, 5$ ),  $YNiCuB_2C$ , and  $YNi_xCu_{2-x}Si_2C$  ( $x=0, 2$ ) were prepared and their superconductivity was studied. The results indicated that with the change of the ratio of Ni to B in  $Y_2Ni_xB_{8-x}C_2$ , secondary phases were introduced into  $YNi_2B_2C$  phase. A  $T_c$  of 15.0 K was observed for the compounds  $x=2, 3$ , and 5, which originated from the major phase  $YNi_2B_2C$  in these samples. Partial substitution of Ni by Cu reduced the  $T_c$  to 11.0 K for  $YNiCuB_2C$ . For Si substitution system, no bulk superconductivity was found in  $YNi_2Si_2C$  and  $YCu_2Si_2C$ . But a minor superconducting phase ( $T_c \sim 4$  K) with a volume fraction of the order of near 1% was observed in them. It was noticed that for the temperature-dependent magnetization of superconducting compounds containing Ni, a significant difference between zero-field cooling and field cooling curves before  $T_c$  was always observed, which was tentatively attributed to Ni-containing impurity. © 1996 American Institute of Physics. [S0021-8979(96)02308-7]

## I. INTRODUCTION

Superconductivity has been discovered recently in borocarbide intermetallic  $YNi_2B_2C$  with  $T_c=15.6$  K.<sup>1</sup> Like  $LuNi_2B_2C$ ,  $YNi_2B_2C$  displays a tetragonal body-centered layerlike structure with a  $I_4/mmm$  symmetry.<sup>1,2</sup> It may be viewed as alternate stackings of the NaCl-type (YC) and the inverse PbO-type ( $Ni_2B_2$ ) layers. In the  $Ni_2B_2$  layers, a Ni atom is tetrahedrally coordinated by four boron atoms. It is interesting to inspect the possibility of increasing  $T_c$  in this system through substitution of Ni and B by other elements such as Cu and Si and through changing the ratio of Ni and B. In this work, borocarbide and silicocarbide intermetallic  $Y_2Ni_xB_{8-x}C_2$  ( $x=1, 2, 3, 5$ ),  $YNiCuB_2C$ , and  $YNi_xCu_{2-x}Si_2C$  ( $x=0, 2$ ) have been prepared and their superconductivity is studied.

## II. EXPERIMENT

Samples were prepared by the arc-melt technique. The starting materials were Y (Aldrich, 99.9%), Ni (Aldrich, 99.95%), Cu (Aldrich, 99.99%), B (99.999%), C (99.9%), and Si (99.9%). Stoichiometric amounts of the starting materials were melted under argon atmosphere on a standard water-cooled copper hearth at least four times, with the melted button turned over between melts in order to ensure homogeneity. The overall loss in weight of the samples during arc melting was less 1%.

The magnetic susceptibility was measured by a SQUID magnetometer (Quantum Design). The resistance was determined by the standard four-lead method. The x-ray diffraction patterns of samples were obtained on a SCINTAG powder diffractometer using  $Cu K\alpha$  radiation.

## III. RESULTS AND DISCUSSION

For  $Y_2Ni_xB_{8-x}C_2$  ( $x=1, 2, 3, 5$ ) system, their x-ray diffraction patterns indicated that all samples were multiphased. Except for the sample with  $x=1$ , all other three samples

showed major diffraction patterns characteristic of  $YNi_2B_2C$ . Figure 1(a) shows the normalized x-ray diffraction patterns of  $Y_2Ni_3B_5C_2$ . One can see that the main peaks can be attributed to the  $YNi_2B_2C$  phase with lattice parameters  $a=3.53$  Å and  $c=10.57$  Å, agreeing with the data in Ref. 1. For  $Y_2Ni_2B_6C_2$  ( $YNiB_3C$ ) and  $Y_2Ni_5B_3C_2$  samples, their major diffraction patterns can also be indexed with the  $YNi_2B_2C$  of the same lattice parameters. For the sample

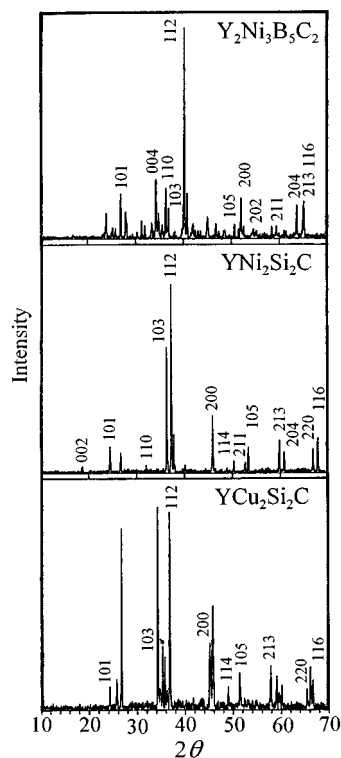


FIG. 1. The x-ray diffraction patterns of  $Y_2Ni_3B_5C_2$ ,  $YNi_2Si_2C$ , and  $YCu_2Si_2C$ .

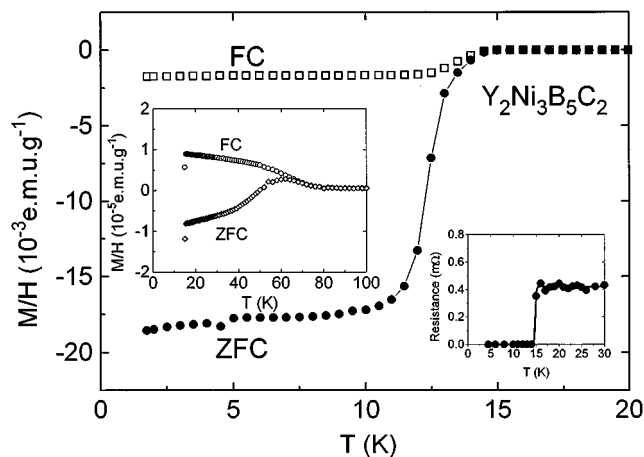


FIG. 2. Temperature-dependent magnetization (applied field  $H = 10$  G) for  $Y_2Ni_3B_5C_2$ . The left inset, expanded view showing the magnetization before  $T_c$ . The right inset shows the resistance for this sample.

$Y_2NiB_7C_2$ , no  $YNi_2B_2C$  phase was observed. The analysis of the secondary phases in these samples has been proven to be a difficult task and will be discussed elsewhere. The above results were consistent with those of Buchgeister *et al.*<sup>3</sup> and Chakoumakos and Paranthaman,<sup>4</sup> who reported that excess amounts of both Ni and B introduced secondary phases into the samples. These results also confirmed that defects such as planar defects or intergrowths were hard to exist in the  $YNi_2B_2C$  structure.<sup>2</sup>

The x-ray diffraction pattern of Cu-substituted  $YNiCuB_2C$  consisted of a major pattern with tetragonal structure with lattice parameters  $a = 3.54$  Å and  $c = 10.63$  Å and some additional peaks which could not be identified. Gangopadhyay *et al.*<sup>5</sup> found that there was limited solubility in  $YNi_2B_2C$  for Cu and that with increasing the doping level of Cu, the impurity peaks grew stronger. Our data agreed with their results.

Silicocarbides  $YNi_2Si_2C$  and  $YCu_2Si_2C$  were prepared as shown in Figs. 1(b) and 1(c). In Fig. 1(b), most of the diffraction peaks can be indexed to the  $YNi_2B_2C$ -type structure with lattice parameters  $a = 3.94$  Å and  $c = 9.55$  Å. This suggests that a new silicocarbide intermetallic  $YNi_2Si_2C$  compound with  $YNi_2B_2C$ -type structure was formed. For  $YCu_2Si_2C$ , the diffraction pattern [Fig. 1(c)] showed that this sample was multiphased. The main peaks were identified to the  $YNi_2B_2C$ -type structure with  $a = 3.95$  Å and  $c = 9.98$  Å. There also existed peaks of secondary phases in Fig. 1(c) whose identification is needed. The result of  $YCu_2Si_2C$  is similar to that of Cu-substituted  $YNiCuB_2C$  in which pure, Cu-containing intermetallic compounds with a  $YNi_2B_2C$ -type structure are difficult to synthesize by means of an arc-melting technique.

Figure 2 shows the temperature dependence of the magnetization measured at 10 G and the resistance for the  $Y_2Ni_3B_5C_2$  sample. The superconducting transition is seen at 15.0 K with a broader transition width as compared with that of the pure  $YNi_2B_2C$  sample.<sup>1</sup> The sample cooled in zero field shows a magnetic shielding equal to  $\sim 80\%$ – $100\%$  of that of a perfect superconductor. On cooling in the field, it

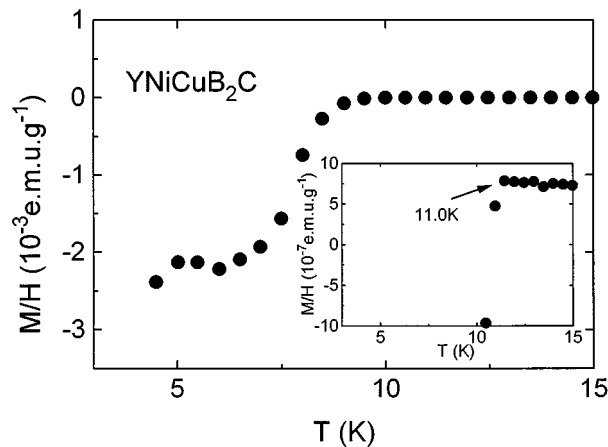


FIG. 3. Temperature-dependent magnetization ( $H = 20$  G) for  $YNiCuB_2C$ , showing the superconducting transition. The data were obtained during warming after zero-field cooling.

displays a Meissner effect of  $\sim 10\%$  of that expected for perfect diamagnetism. The broader transition width may result from the secondary phases already seen in the x-ray pattern which separate the  $YNi_2B_2C$  phase grains so that weak links form. We found that before  $T_c$ , positive, up-grown field cooling and zero-field cooling curves were always observed in these Ni-containing samples. As shown in the left inset of Fig. 2, the zero-field cooling curve shows a broad maximum at  $\sim 60$  K. More interestingly, for this sample, its ZFC susceptibility became negative at a lower temperature. Although ac susceptibility conducted on the sample indicated that this negative ZFC dc susceptibility was not due to a superconducting phase transition, its origin is yet

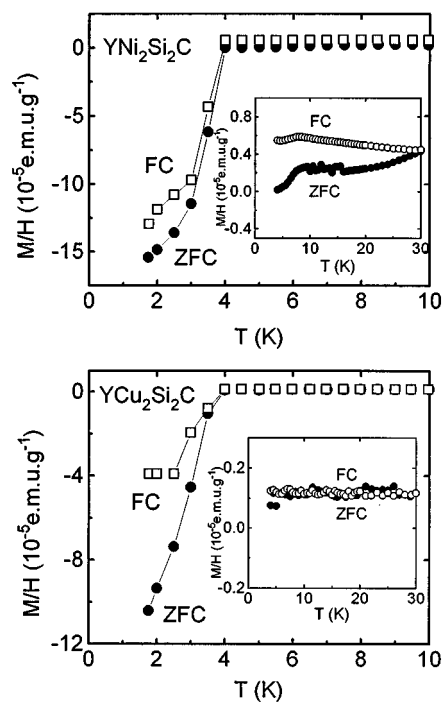


FIG. 4. Temperature-dependent magnetization ( $H = 10$  G) for  $YNi_2Si_2C$  and  $YCu_2Si_2C$ . The insets, expanded view showing the magnetization before  $T_c$ .

to be found out. Its FC susceptibility, on the other hand, continued to increase with decreasing temperature until  $T_c$  was reached. This novel behavior is most likely to be associated with a Ni-containing impurity. For  $Y_2Ni_2B_6C_2$  ( $YNiB_3C$ ) and  $Y_2Ni_5B_3C_2$  samples, they possess the same onset temperature at 15.0 K with a broader transition width. This result is consistent with the x-ray data which indicate that  $YNi_2B_2C$  is the major phase in these samples. For the sample  $Y_2NiB_7C_2$ , no superconductivity was found. Its susceptibility (in the order of  $10^{-5}$ ) seems to follow the Curie–Weiss law. Such a behavior may be due to a residual magnetic moment of Ni.<sup>6</sup>

The temperature-dependent magnetization data measured at 20 G for Cu-substituted  $YNiCuB_2C$  are given in Fig. 3. The shield signal corresponds to  $\sim 25\%$  of perfect diamagnetism. The inset to Fig. 3 shows an onset temperature at 11.0 K, indicating that Cu substitution for Ni reduces  $T_c$ , agreeing with that of Gangopadhyay *et al.*<sup>5</sup>

Superconductivity in small quantities can be observed in silicocarbides  $YNi_2Si_2C$  and  $YCu_2Si_2C$ . The temperature dependence of the magnetization at 10 G for both samples is shown in Fig. 4. The onset temperature is  $\sim 4$  K for both samples. The magnitudes of the shielding and Meissner effect signals correspond to  $\sim 1.5\%$  and  $\sim 1.2\%$  of perfect diamagnetism for  $YNi_2Si_2C$  and  $\sim 1\%$  and  $\sim 0.4\%$  for  $YCu_2Si_2C$ , respectively. This suggests that the superconducting phase is a minor phase with a volume fraction of the order of 1%. From the insets of Fig. 4, the hysteresis between field cooling and zero-field cooling curves before  $T_c$  can also be seen in  $YNi_2Si_2C$ . While such curves were not observed in  $YCu_2Si_2C$ , which is Ni free, further suggesting that this history-dependent hysteretic magnetic behavior is associated with a Ni-containing impurity.

#### IV. CONCLUSIONS

Deviation from the stoichiometric composition of  $YNi_2B_2C$  by changing the ratio of Ni to B introduced impurity phases into the major phase  $YNi_2B_2C$ . In  $Y_2Ni_xB_{8-x}C_2$  ( $x=2, 3, 5$ ), the major phase  $YNi_2B_2C$  was responsible for superconductivity with  $T_c=15.0$  K. For Cu-substituted  $YNiCuB_2C$ , an expansion in lattice was observed due to the substitution of Ni with Cu. Secondary phases emerged because of the limited solubility of Cu in this compound. The Cu substitution reduced the  $T_c$  to 11 K, in agreement with the argument that the Fermi level in the parent compound lies very near to a peak in the density of states so that further enhancement in  $T_c$  by chemical modification is rather unlikely.<sup>5</sup> For new silicocarbides  $YNi_2Si_2C$  and  $YCu_2Si_2C$ , they were not bulk superconductors. But there existed superconducting minor phases with  $T_c \approx 4$  K.

#### ACKNOWLEDGMENTS

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<sup>4</sup>B. C. Chakoumakos and M. Paranthaman, Physica C **227**, 143 (1994).

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