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Enhancement in superconducting transition temperature (T_c) and upper critical field (H_{c2}) in new Yb-doped $Ce_{1-x}Yb_xO_{0.9}F_{0.1}FeAs$ superconductors

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ABSTRACT

We report the synthesis and characterization of new oxypnictides of the type $Ce_{1-x}Yb_xO_{0.9}F_{0.1}FeAs$ ('x' = 0.3, 0.4 and 0.5) for the first time. All these compounds crystallize in the tetragonal ZrCuSiAs type structure (space group: *P*4/*nmm*). Reduction in both the lattice parameters (**a** and **c**) was observed on substitution of smaller Yb ions at Ce sites in $CeO_{0.9}F_{0.1}FeAs$. All these compounds were found to be superconducting with maximum T_c of 48.7 K for 'x' = 0.5 composition which is highest among Ce(O/F)FeAs based superconductors. Temperature dependence of resistivity under magnetic field has been studied to evaluate the upper critical field ($H_{c2}(0)$) of these superconductors. Enhancement in upper critical field (H_{c2}) was observed for these Yb doped $Ce_{1-x}Yb_xO_{0.9}F_{0.1}FeAs$ superconductors (max $H_{c2}(0) = \sim 142$ T for 'x' = 0.3). The average coherence length of all Yb doped samples was estimated to be ~23 Å.

1. Introduction

The recent discovery of superconductivity at 26 K in an oxypnictide La(O/F)FeAs [1] received enormous attention among solid state chemists and condensed matter physicist. Following this report, many new pnictogen and chalcogen based superconducting families were discovered with the general formula of AFFeAs, AFe_2As_2, AFeAs, $A_4M_2M'_2Pn_2O_6$, $A_3M_2M'_2Pn_2O_5$ and FeSe/Te. Compounds with the general formula LnOFeAs (1111 system) adopt the tetragonal ZrCuSiAs type structure (space group: *P*4/*nmm*) [2] which is a filled variant of well known PbFCl structure (space group: *P*4/*nmm*). Its structure consists alternate layers of namely [La–O] (charge reservoir) and [Fe–As] (conducting layer) [3]. Undoped CeOFeAs is an antiferromagnetic semimetal and exhibits structural transition from tetragonal (space group: *P*4/*nmm*) to orthorhombic structure (space group: *Cmma*) [4]. Resistivity and magnetic studies of LnOFeAs shows an anomaly at 150–160 K due to structural transition followed by a spin density wave due to antiferromagnetic ordering of iron spins. Superconductivity in these pnictides can be induced by suppressing structural transition and antiferromagnetism through electron or hole doping [1,5–8]. Electrons in LnOFeAs can be doped through substitution of fluoride ions at oxygen site [1,5] while holes are doped through substitution of strontium ions at rare-earth site [7,8]. Enhancement in superconducting transition temperature of La(O/F)FeAs was achieved by replacing La by other smaller rare-earth metals like Ce [9], Pr [10], Nd [11], Sm [12] etc. with maximum T_c of 55 K in Sm(O/F)FeAs [13]. The hole doped $Ln_{1-x}Sr_xOFeAs$ superconductors show maximum T_c of 25 K in $La_{0.8}Sr_{0.2}OFeAs$ [7]. The substitution of Co at Fe sites in LnOFeAs also dopes electrons and induces superconductivity [14–16] with maximum T_c of 15 K in $SmOFe_{0.9}Co_{0.1}As$ [15].

In this report we have investigated the effect of substitution of smaller ytterbium ions at Ce site in $CeO_{0.9}F_{0.1}FeAs$ superconductor on its structural and superconducting properties. We have also estimated upper critical field of these novel superconductors.

2. Experimental

Polycrystalline samples with nominal compositions of $Ce_{1-x}Yb_xO_{0.9}F_{0.1}FeAs$ ('x' = 0.3, 0.4 and 0.5) were synthesized by a two step solid state method using high purity Ce, CeO_2, Yb_2O_3, CeF_3, and FeAs as starting materials. FeAs was obtained by reacting Fe chips and As powder at 800 °C for 24 h. The raw materials were taken according to stoichiometric ratio and then sealed in evacuated silica ampoules (10^{-4} torr) and heated at 900 °C for 30 h. The powder was then compacted (5 tons) and the disks were wrapped in Ta foil, sealed in evacuated silica ampoules and heated at 1150 °C for 30 h. All the chemical manipulations were performed in an Argon-filled glove box. The samples were characterized by powder X-ray diffraction using Cu Kα radiation. Refined lattice parameters were calculated by least square fit method.

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The temperature dependence of normal state resistivity (ρ) of Yb-doped $Ce_{1-x}Yb_xO_{0.9}F_{0.1}FeAs$ ('x' = 0.3, 0.4 and 0.5) polycrystalline samples were measured by conventional four probe technique using PPMS (Quantum Design, USA) with an excitation current of 1 mA. For resistivity measurements under applied magnetic field (0–4 T) data were recorded during warming runs at a rate of 1 K/min. The dc magnetic susceptibility as a function of temperature was also performed by PPMS–VSM module under zero field cooling with applied field of 5 Oe at temperature between 2 and 300 K.

3. Results and discussion

Powder X-ray diffraction patterns for $Ce_{1-x}Yb_xO_{0.9}F_{0.1}FeAs$ ('x' = 0.3, 0.4 and 0.5) are shown in Fig. 1. Majority of the observed reflections could be indexed on the basis of tetragonal CeOFeAs structure (space group: $P4/nmm$). Along with major tetragonal CeOFeAs phase, some amount of Yb_2O_3, Fe_2As and CeAs were also observed as secondary phases. Both the lattice parameters (**a** and **c**) were found to be decreasing with increase in Yb substitution (Fig. 2) which is expected since the ionic size of Yb^{3+}(0.985 Å) is smaller as compared to Ce^{3+} (1.143 Å) in eightfold coordination. Hence the substitution of Yb-ions in $CeO_{0.9}F_{0.1}FeAs$ increases the chemical pressure. The lattice parameters for all the Yb-doped compositions were compared and found to be smaller than that of the parent $CeO_{0.9}F_{0.1}FeAs$ (a = 3.991(1) Å and c = 8.613(3) Å) reported earlier [17]. This indicates successful substitution of Yb at the rare earth sites. 'x' = 0.3 composition (a = 3.991 Å and c = 8.625 Å) however does not follow the trend, indicating a deviation of stoichiometry from the loaded composition. The variation of resistivity as a function of temperature for $Ce_{1-x}Yb_xO_{0.9}F_{0.1}FeAs$ ('x' = 0.3, 0.4 and 0.5) is shown in Fig. 3a. For 'x' = 0.3 composition temperature dependence of resistivity show metallic behavior before a superconducting transition at 42.7 K which is higher than the ytterbium free $CeO_{0.9}F_{0.1}FeAs$ (T_c = 38 K [17]). The superconducting transition temperature (T_c) was determined by the intersection of two extrapolated lines from 90% and 10% of the resistivity curve (schematically shown in Fig. 3b). Further increase in Yb substitution (x) in $Ce_{1-x}Yb_xO_{0.9}F_{0.1}FeAs$ leads to gradual enhancement of T_c to 47.1 K and 48.7 K for 'x' = 0.4 and 'x' = 0.5 respectively. The maximum T_c was observed for 'x' = 0.5 (48.7 K) composition which is highest among the Ce(O/F)FeAs superconductors [9,17]. The calculated residual resistivity values ($RRR = R_{300K}/R_{50K}$) were found to be 3.6, 4.7 and 3.5 for 'x' = 0.3, 0.4 and 0.5 compositions, respectively. The small value of RRR indicates inhomogeneity in samples. Decrease in lattice parameters (chemical pressure) through substitution at Ln sites leading to increase in T_c as has been reported earlier [18,19]. On comparison with other superconducting Ln-1111 phases [18,20] it is clear that the oxygen-deficient and yttrium substituted Ln-1111 phases show a gradual increase in T_c with decrease in 'a'- lattice parameter up to a certain level of doping. Reduction in the lattice parameters on substitution of smaller Ln ions lead to shorter Fe–As bond lengths which increases the Fe–As hybridization and changes the bandwidth leading to the enhancement in T_c. The temperature

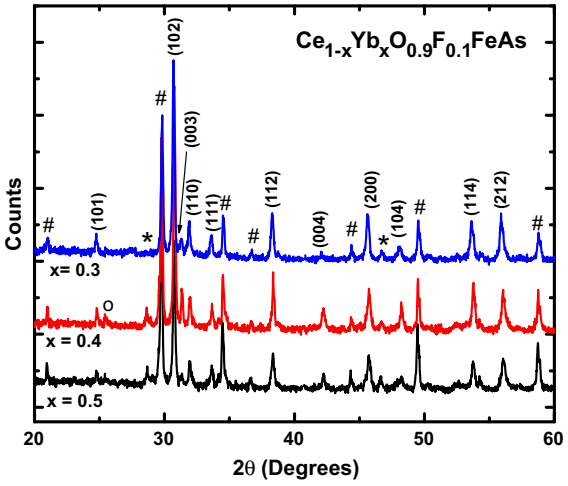

Fig. 1. Powder X-ray diffraction patterns of $Ce_{1-x}Yb_xO_{0.9}F_{0.1}FeAs$. The impurity phases are Fe_2As (∗), Yb_2O_3 (#) and CeAs (0).

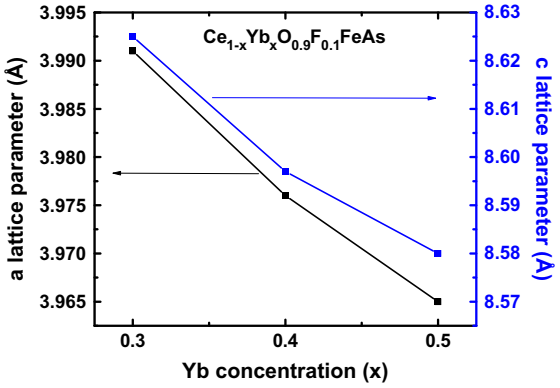

Fig. 2. Plot of variation of lattice parameters (**a** and **c**) with Ytterbium content (x) in $Ce_{1-x}Yb_xO_{0.9}F_{0.1}FeAs$.

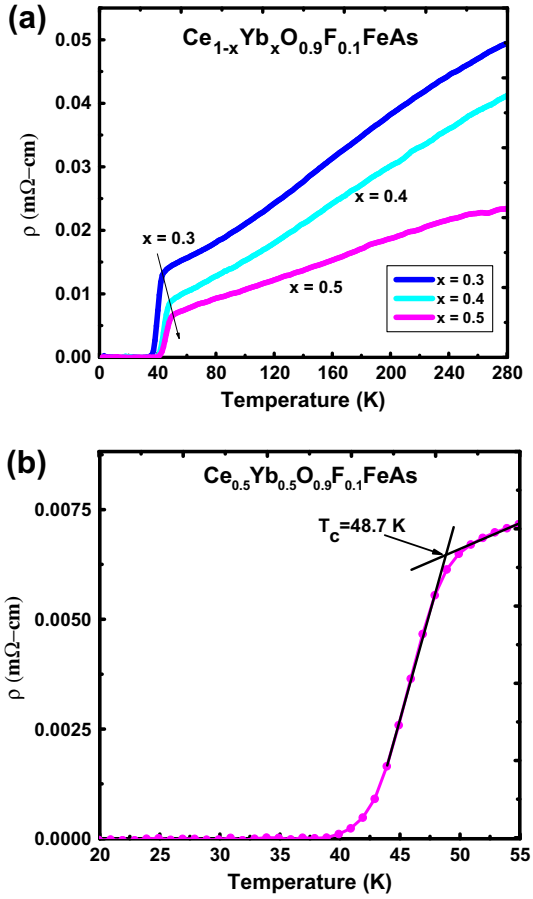

Fig. 3. Temperature dependence of resistivity (ρ) for (a) $Ce_{1-x}Yb_xO_{0.9}F_{0.1}FeAs$ ('x' = 0.3, 0.4 and 0.5) and (b) ρ vs T plot for $Ce_{0.5}Yb_{0.5}O_{0.9}F_{0.1}FeAs$ showing the criterion for evaluating T_c.