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Magnetic Studies on $Sc_yU_{1-y}O_{2+x}$ Solid Solutions

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Magnetic susceptibilities of $Sc_yU_{1-y}O_{2+x}$ solid solutions have been measured from 2.7 K to room temperature. The magnetic moment and Weiss constant have been determined in the temperature range in which the Curie–Weiss law holds. For the solid solutions showing antiferromagnetic transition, the Néel temperature has also been determined. The substitution of Sc^{3+} for U^{4+} was found to effect not only magnetic dilution of UO_2, but also oxidation of U^{4+} to U^{5+}. Excess oxygen ions which entered the interstitial sites, weakened the antiferromagnetic interaction between uranium ions and oxidized U^{4+} to U^{5+}. The effect of oxygen vacancies on the antiferromagnetic interaction was small in the concentration range of this experiment (0.8 a/o). © 1986 Academic Press, Inc.

Introduction

Investigations on UO_2–Sc_2O_3 system are limited (*1–5*), and the magnetic study concerning this system seems to be done only by Kemmler-Sack *et al.* (*4*), who measured the magnetic susceptibility of $Sc_{0.5}U_{0.5}O_2$ from liquid-nitrogen temperature to 473 K and obtained the magnetic moment of U^{5+} from the Curie law.

The magnetic behavior of UO_2–Sc_2O_3 solid solutions is complicated. Substitution of Sc^{3+} for U^{4+} in UO_2 lattice results in not only magnetic dilution of UO_2, but also oxidation of uranium ions to higher valence state according to the electrical neutrality condition. In the case of hyper- or hypostoichiometric solid solutions, the effect of excess or deficient oxygens must also be considered.

In the present study, we prepared $Sc_yU_{1-y}O_{2+x}$ solid solutions which have the same fluorite-type structure as UO_2, and measured their magnetic susceptibilities from 2.7 K to room temperature. The effects of Sc^{3+} and excess or deficient oxygens on the magnetic properties of solid solutions were investigated by using specimens with various x and y values.

Experimental

1. Sample Preparation

As the starting materials, UO_2 and Sc_2O_3 were used. Before use, the uranium dioxide was reduced to a stoichiometric composition in flowing hydrogen at ca. 1000°C and scandium sesquioxide was heated in air at ca. 850°C.

The UO_2 and Sc_2O_3 were weighed to the intended atom ratios of uranium and scandium. After being finely ground, the mix-

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TABLE I

LATTICE PARAMETER OF $Sc_yU_{1-y}O_{2+x}$ SOLID SOLUTIONS

Solid solution	Lattice parameter (Å)	Preparation condition
$Sc_{0.05}U_{0.95}O_{1.984}$	5.4530	I
$Sc_{0.05}U_{0.95}O_{1.998}$	5.4487	II
$Sc_{0.10}U_{0.90}O_{2.001}$	5.4265	II
$Sc_{0.05}U_{0.95}O_{2.177}$	5.4229	III
$Sc_{0.10}U_{0.90}O_{2.145}$	5.3990	III
$Sc_{0.15}U_{0.85}O_{2.117}$	5.3839	III

tures were pressed into pellets and then heated under either of the following three kinds of conditions:

1. The reaction in an induction furnace *in vacuo* (2×10^{-3} Torr) at 1600°C. This condition will be referred to as Condition I.
2. The reaction in the induction furnace in flowing helium at 1400°C. This condition will be referred to as Condition II.
3. The reaction in a SiC resistance furnace in flowing helium at 1400°C. Since a vinyl tube was used in this case for helium gas introduction, the partial pressure of oxygen in the gas is significantly higher than that of Condition II. This condition will be referred to as Condition III.

After cooling to room temperature, the samples were crushed into powder, repressed, and reacted under the same conditions to make the reaction complete. These procedures were repeated twice.

2. *Analysis*

2.1. X-Ray diffraction analysis. X-Ray diffraction study on the solid solutions was performed using Cu$K\alpha$ radiation with a Philips PW 1390 diffractometer with curved graphite monochromator. The lattice parameter of the samples was determined by Nelson–Riley extrapolation method applied to the diffraction lines above 80° (2θ).

2.2. Determination of oxygen in solid solutions. The amount of oxygen in the solid solutions was determined by the back-titration method (*6, 7*). The sample was dissolved in excess cerium(IV) sulfate solution. The cerium(IV) sulfate solution was standardized with stoichiometric UO_2. The excess cerium(IV) was titrated against standard iron(II) ammonium sulfate solution with ferroin indicator. The oxygen amount was determined for predetermined Sc/U ratio and sample weight.

3. *Magnetic Susceptibility Measurement*

Magnetic susceptibility was measured by the Faraday-type torsion balance in the temperature range from 2.7 K to room temperature. The apparatus was calibrated with Mn-Tutton's salt ($\chi_g = 10{,}980 \times 10^{-6}/(T + 0.7)$) as a standard. The temperature of the sample was measured by "normal" Ag vs Au–0.07 at.% Fe thermocouple (*8*) and Au–Co vs Cu thermocouple. Details of the experimental procedure have been described elsewhere (*9*).

Results and Discussion

The results of X-ray diffraction analysis showed that cubic solid solutions with fluorite structure were formed for all samples in this study. The lattice parameter and the composition of the samples prepared are listed in Table I. The solid solution prepared *in vacuo* at 1600°C (Condition I) is oxygen-deficient. In Fig. 1 the lattice parameter is plotted as a function of Sc^{3+} concentration. Solid solutions prepared in the induction furnace in flowing helium (Condition II) are nearly stoichiometric and the lattice parameter decreases linearly with increasing Sc^{3+} concentration from that of UO_2, which could be followed by an equation, $a_0\ (\text{Å}) = 5.4704 - 0.438\, y$. Solid solutions prepared in the SiC furnace (Condition III) are hyperstoichiometric, and their lattice parameter is no longer connected straight to that of UO_2 but decreases almost

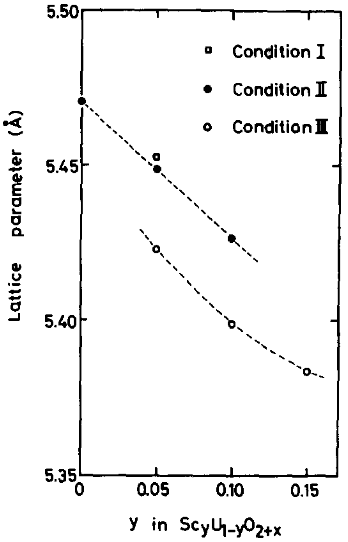

FIG. 1. Lattice parameter vs Sc^{3+} concentration.

TABLE II

MAGNETIC PARAMETERS FOR $Sc_yU_{1-y}O_{2+x}$ SOLID SOLUTIONS

Solid solution	C	μ_{eff} (B.M.)	θ (K)	T_N (K)	Mean valency of U
UO_2[a]	1.217	3.12	−220.0	31.0	4.000
$Sc_{0.05}U_{0.95}O_{1.984}$	1.111	2.98	−206.7	24.0	4.019
$Sc_{0.05}U_{0.95}O_{1.998}$	1.095	2.96	−207.0	24.0	4.048
$Sc_{0.10}U_{0.90}O_{2.001}$	1.021	2.86	−190.2	17.2	4.113
$Sc_{0.05}U_{0.95}O_{2.177}$	0.785	2.51	−118.4	—	4.425
$Sc_{0.10}U_{0.90}O_{2.145}$	0.763	2.47	−118.6	—	4.433
$Sc_{0.15}U_{0.85}O_{2.117}$	0.742	2.44	−118.1	6.0	4.452

[a] Parameters for UO_2 are given in Ref. (*9*).

linearly with increasing Sc^{3+} concentration.

The temperature dependence of magnetic susceptibility per mole of uranium for the solid solution prepared under Condition I, i.e., $Sc_{0.05}U_{0.95}O_{1.984}$ is shown in Fig. 2. Antiferromagnetic transition occurs at T_N = 24.0 K, and below 10 K the magnetic susceptibility does not change materially with decreasing temperature. This behavior is similar to that of UO_2–ThO_2 solid solutions (*9, 10*) or UO_2–ZrO_2 solid solutions (*11*). From the reciprocal susceptibility vs temperature curve, the Curie–Weiss law was found to hold in the temperature range from ca. 50 K to room temperature. The effective magnetic moment (μ_{eff}) was 2.98 B.M. and the Weiss constant (θ) was −206.7 K. Magnetic parameters (μ_{eff}, θ, and T_N) of the solid solutions are listed in Table II.

Figure 3 shows the temperature depen-

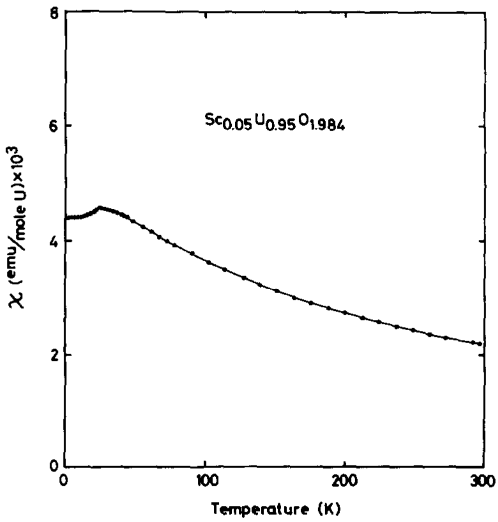

FIG. 2. Magnetic susceptibility of $Sc_{0.05}U_{0.95}O_{1.984}$ vs temperature.

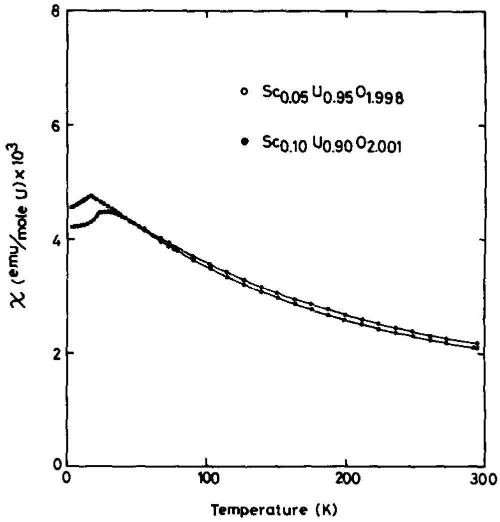

FIG. 3. Magnetic susceptibility of $Sc_{0.05}U_{0.95}O_{1.998}$ and $Sc_{0.10}U_{0.90}O_{2.001}$ vs temperature.

dence of magnetic susceptibility per mole of uranium for the solid solutions prepared under Condition II, i.e., $Sc_{0.05}U_{0.95}O_{1.998}$ and $Sc_{0.10}U_{0.90}O_{2.001}$. Since both the oxygen amounts in these two solid solutions are very near to 2, it is appropriate to examine the effects of diamagnetic Sc^{3+} on the magnetic properties by these solid solutions. From the fact that the Néel temperature (T_N) decreases from 31.0 K for UO_2 with increasing Sc^{3+} concentration, the magnetic interaction between uranium ions is found to be weakened with increasing Sc^{3+} concentration. The effective magnetic moment and Weiss constant given in Table II also decrease from those of UO_2 with the dilution. This result is the same as those of UO_2–ThO_2 solid solutions (*9, 12*) and UO_2–ZrO_2 solid solutions (*11*).

By comparing the magnetic susceptibility data for $Sc_{0.05}U_{0.95}O_{1.984}$ with those for $Sc_{0.05}U_{0.95}O_{1.998}$, the effect of oxygen vacancy on magnetic properties can be discussed. With the change from $Sc_{0.05}U_{0.95}O_{1.998}$ to $Sc_{0.05}U_{0.95}O_{1.984}$, the ratio of oxygen vacancy increases from 0.1 a/o to 0.8 a/o, but the Néel temperature and Weiss constant which are measures of magnitude of antiferromagnetic interaction, do not appreciably change. That is to say, T_N was 24.0 K for both the specimens and θ differed only from −207.0 to −206.7 K. Accordingly, it can be said that the oxygen vacancy to this extent does not affect the antiferromagnetism of uranium ions. As the oxygen amount in these solid solutions is larger than that calculated for the case that the valence state of uranium is +4 only ($Sc_{0.05}U_{0.95}O_{1.975}$), some of the uranium ions are considered to be oxidized to either U^{5+} or U^{6+}. Since the uranium in $Sc_{0.05}U_{0.95}O_{1.998}$ is more oxidized than in $Sc_{0.05}U_{0.95}O_{1.984}$, the magnetic moment of uranium ion in the former solid solution is expected to be smaller than that in the latter. The experimental results show that the value of magnetic moment decreases from 2.98 B.M. for $Sc_{0.05}U_{0.95}O_{1.984}$ to 2.96 B.M. for $Sc_{0.05}U_{0.95}O_{1.998}$. The largest difference in magnetic behavior of $Sc_{0.05}U_{0.95}O_{1.984}$ and $Sc_{0.05}U_{0.95}O_{1.998}$ is the temperature dependence of magnetic susceptibility below the Néel temperature. In the former solid solution, the susceptibility is almost constant in some temperature range below T_N, while in the latter solid solution, it decreases with decreasing temperature still in this range. The reason for this difference is not very clear, but one possibility is that the decrease of the susceptibility with decreasing temperature seems to correspond to the magnetic exchange interaction in which U^{5+} ions participate, i.e., the interaction either between U^{5+} ions or between U^{4+} and U^{5+}. A similar phenomenon has been observed also in $Y_yU_{1-y}O_2$ solid solutions (*13*), but not in UO_2–ThO_2 solid solutions (*9, 10*) or UO_2–ZrO_2 solid solutions (*11*).

The temperature dependence of magnetic susceptibility for hyperstoichiometric solid solutions prepared under Condition III is shown in Fig. 4. Maxima are not found in the magnetic susceptibility–temperature curves for $Sc_{0.05}U_{0.95}O_{2.177}$ and $Sc_{0.10}U_{0.90}O_{2.145}$. This means that antiferromagnetic transition does not occur for the hyperstoichiometric solid solutions. If this magnetic behavior is compared with $Sc_{0.05}U_{0.95}O_{1.998}$ and $Sc_{0.05}U_{0.95}O_{2.001}$ which are almost stoichiometric, the excess oxygen ions which are located at interstitial sites are seen to play a role to weaken the antiferromagnetic interactions between uranium ions. The Curie–Weiss law holds in the temperature range from ca. 50 K to room temperature, and the magnetic moment and Weiss constant are $\mu_{eff} = 2.51$ B.M. and $\theta = -118.4$ K for $Sc_{0.05}U_{0.95}O_{2.177}$, and $\mu_{eff} = 2.47$ B.M. and $\theta = -118.6$ K for $Sc_{0.10}U_{0.90}O_{2.145}$. The magnetic moments of these solid solutions are considerably small compared with those of solid solutions which are hypostoichiometric or nearly stoichiometric. This fact shows that the uranium ions are further

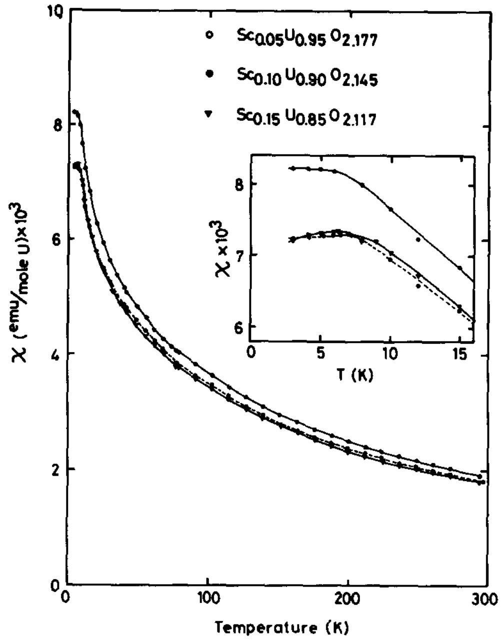

FIG 4. Magnetic susceptibility of $Sc_{0.05}U_{0.95}O_{2.177}$, $Sc_{0.10}U_{0.90}O_{2.145}$, and $Sc_{0.15}U_{0.85}O_{2.117}$ vs temperature. The inset shows details of the low-temperature region.

oxidized by the excess oxygen ions in addition to the substitution of Sc^{3+} for U^{4+} to maintain the electrical neutrality in the solid solutions. As the amount of excess oxygen ions decreases with increasing Sc^{3+} concentration under the same partial pressure of oxygen, antiferromagnetic transition is expected to be observed for solid solutions in which the Sc^{3+} concentration is higher than some critical value. This was observed in $Sc_{0.15}U_{0.95}O_{2.117}$ at 6.2 K.

The magnetic moment and Weiss constant of each solid solution are plotted against Sc^{3+} concentration in Fig. 5. Both the magnetic moment and Weiss constant of the solid solutions prepared under Condition II can be connected smoothly to those of UO_2, which is the same as in the change of the lattice parameter. On the other hand, the magnetic moment and Weiss constant of the samples prepared under Condition III seem hardly to be connected to those of UO_2. It is noteworthy, however, that the Weiss constant is almost unchanged in the lower three samples in Table II, where the oxygen amount decreases with increasing Sc^{3+} concentration. The reason is considered as follows: With increasing Sc^{3+} concentration, the mean valency of uranium in these samples increases from 4.425 to 4.452, which means an increase in the amount of U^{5+} as will be described below. Since the energy difference between the ground state and the excited state in case of U^{5+} under a crystalline field with cubic symmetry is much larger than that in case of U^{4+}, the Weiss constant of U^{5+} is usually smaller than that of U^{4+}. In addition, the decrease in the excess oxygen amount with increasing Sc^{3+} concentration serves to increase the Weiss constant since the excess oxygen ions weaken the antiferromagnetic interactions between uranium ions and this constant is a measure of the strength of these interactions. The essentially unchanged Weiss constant can be ascribed to the offset of the two effects above. The magnetic moment is found to decrease with increasing Sc^{3+} concentration, which

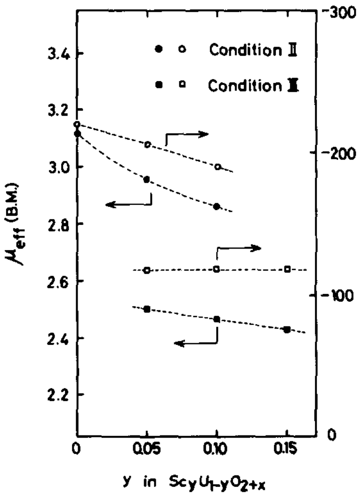

FIG. 5. Magnetic moment and Weiss constant vs Sc^{3+} concentration.

reflects the oxidation of uranium by substitution of Sc^{3+}.

On the basis of the results of the magnetic susceptibility measurements, the effect of excess oxygen ions which enter interstitial sites will be first considered. We compare the magnetic properties of solid solutions with the same y value and different x values, i.e., $Sc_{0.05}U_{0.95}O_{1.998}$ which is nearly stoichiometric and $Sc_{0.05}U_{0.95}O_{2.177}$. Assuming that the U^{4+} ions are oxidized to hexavalent state, the ions in the solid solutions are

$$Sc_y^{3+}U_{1-x-1.5y}^{4+}U_{x+0.5y}^{6+}O_{2+x}^{2-}. \qquad (1)$$

The ratios, U^{4+}/U_{total}, are calculated from Eq. (1) as follows:

for $Sc_{0.05}U_{0.95}O_{1.998}$,

$$C_1' = U^{4+}/U_{\text{total}} = 0.927/0.95 = 0.976;$$

for $Sc_{0.05}U_{0.95}O_{2.177}$,

$$C_2' = U^{4+}/U_{\text{total}} = 0.748/0.95 = 0.787.$$

Since U^{6+} ions are diamagnetic, the ratios of U^{4+} are considered to be proportional to the Curie constants, C, which are also listed in Table II. As the concentration of Sc^{3+} is equal in both the solid solutions, the effect of U^{4+} is considered to be the same as a first approximation. The Curie constants for $Sc_{0.05}U_{0.95}O_{1.998}$, C_1, and for $Sc_{0.05}U_{0.95}O_{2.177}$, C_2, are 1.095 and 0.785, respectively. If the mechanism shown in Eq. (1) is correct, the relation $C_1/C_2 = C_1'/C_2'$ must hold. Actually, the ratio experimentally obtained, C_1/C_2, does not accord with the calculated ratio, C_1'/C_2'. This result means that the uranium ions are not oxidized in the way shown in Eq. (1).

Next, the case that the oxidation of uranium proceeds from U^{4+} to U^{5+} will be considered. In this case, the following ions should comprise the solid solutions:

$$Sc_y^{3+}U_{1-2x-2y}^{4+}U_{2x+y}^{5+}O_{2+x}^{2-}. \qquad (2)$$

Here, since both U^{4+} and U^{5+} ions contribute to the paramagnetism, the method above is ineffective.

For the magnetic susceptibilities of two solid solutions with the same y value but different x values (to be referred to as $\chi(S.1)$ and $\chi(S.2)$ and here S.1 and S.2 correspond to $Sc_{0.05}U_{0.95}O_{1.998}$ and $Sc_{0.05}U_{0.95}O_{2.177}$, respectively), the following two equations hold,

$$\chi(S.1) = (1 - 2x_1 - 2y)\chi(U^{4+}) + (2x_1 + y)\chi(U^{5+}), \qquad (3)$$

$$\chi(S.2) = (1 - 2x_2 - 2y)\chi(U^{4+}) + (2x_2 + y)\chi(U^{5+}), \qquad (4)$$

where $\chi(U^{4+})$ and $\chi(U^{5+})$ are the magnetic susceptibilities of U^{4+} and U^{5+}, respectively. By eliminating $\chi(U^{4+})$ from Eqs. (3) and (4), $\chi(U^{5+})$ is expressed as

$$\chi(U^{5+}) = \frac{1}{2(1 - y)(x_1 - x_2)} \times [(1 - 2x_2 - 2y)\chi(S.1) - (1 - 2x_1 - 2y)\chi(S.2)]. \qquad (5)$$

In this case, $x_1 = -0.002$, $x_2 = 0.177$, and $y = 0.05$. Therefore,

$$\chi(U^{5+}) = 2.658\chi(S.2) - 1.605\chi(S.1). \qquad (6)$$

Using the susceptibilities experimentally obtained for $\chi(S.1)$ and $\chi(S.2)$, the magnetic susceptibility for U^{5+} was obtained. Its reciprocal susceptibility vs temperature curve is shown in Fig. 6. It is found that the Curie–Weiss law holds in the entire temperature range studied. Its effective magnetic moment is calculated to be 2.00 B.M.

The stoichiometric UO_2 has a cubic fluorite structure. In the solid solutions also, a central uranium ion is considered to be surrounded by eight oxygen ions located at the corners of a cube, as a first approximation. The ground state of U^{5+} ion with one unpaired electron is $^2F_{5/2}$. In a crystalline field with cubic symmetry, this multiplet state splits into two states, doublet state Γ_7 and quartet state Γ_8 (in Bethe's notation) *(14)*. The difference in energy between these two states is usually large (~4000 cm^{-1}) *(15,*

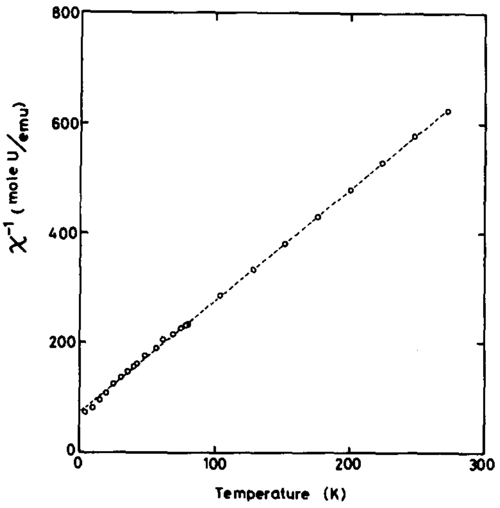

FIG. 6. Reciprocal magnetic susceptibility for U^{5+} vs temperature. U^{5+} is formed due to excess oxygen ion.

16). The ground state is Γ_8 and its wavefunctions are

$$\sqrt{5/6}\,|\pm \tfrac{5}{2}\rangle + \sqrt{1/6}\,|\mp \tfrac{3}{2}\rangle, \quad |\pm \tfrac{1}{2}\rangle.$$

If only this quartet state Γ_8 contributes to the paramagnetism, magnetic susceptibility of the solid solution is calculated as

$$\chi = \frac{Ng^2\beta^2}{kT}\cdot\frac{65}{36}, \qquad (7)$$

and the effective magnetic moment is 2.00 B.M. which is consistent with the value experimentally obtained. This fact shows that when excess oxygen ions enter the lattice, oxidation of uranium proceeds from U^{4+} to U^{5+} and that as a first approximation, the change in symmetry of crystalline field by the nonstoichiometric oxygen ions has a negligibly small effect on the magnetic moment.

Next, the effect of substitution of Sc^{3+} on the magnetic properties of the solid solutions will be considered. Since Sc^{3+} is a diamagnetic ion, substitution of the scandium ion for uranium ion results in magnetic dilution as described above. Oxidation of the uranium ion due to the substitution of trivalent scandium ion proceeds simultaneously to maintain the electrical neutrality in the solids. Since the samples prepared under Condition II are almost stoichiometric in oxygen amounts, the effect of excess oxygen ions would be very small. The effect of Sc^{3+} can be discussed by comparing the magnetic properties of $Sc_{0.05}U_{0.95}O_{1.998}$ and $Sc_{0.10}U_{0.90}O_{2.001}$. The possibility of oxidation to U^{6+} can logically be excluded as we have shown in the above paragraph that U^{4+} and U^{5+} exist in the same solid solution $Sc_{0.05}U_{0.95}O_{1.998}$. For the solid solutions, $Sc_{0.05}U_{0.95}O_{1.998}$ and $Sc_{0.10}U_{0.90}O_{2.001}$ (to be referred to as S.3 and S.4, respectively), the following two equations hold,

$$\chi(\text{S.3}) = (1 - 2x_3 - 2y_3)\chi(U^{4+}) + (2x_3 + y_3)\chi(U^{5+}), \qquad (8)$$

$$\chi(\text{S.4}) = (1 - 2x_4 - 2y_4)\chi'(U^{4+}) + (2x_4 + y_4)\chi'(U^{5+}). \qquad (9)$$

In treating these two equations, the problem is that the substitution of scandium results in not only oxidation of uranium but also diminution of antiferromagnetic interactions between the uranium ions. That is, the difference in magnitude of magnetic dilution also varies the magnetic susceptibility and therefore the effective magnetic moment, which is shown in UO_2–ThO_2 solid solutions (*9, 12, 17, 18*) and UO_2–ZrO_2 solid solutions (*11*). To take the dilution effect on magnetic susceptibility into consideration, it is desirable to compare, for example, the magnetic properties of $Sc_{0.10}U_{0.90}O_{2.001}$ with those of the solid solution in which some diamagnetic tetravalent ions replace the uranium ions in the same concentration with Sc^{3+}. However, comparison of the magnetic properties of $Sc_{0.10}U_{0.90}O_{2.001}$ with those of $Zr_{0.10}U_{0.90}O_{2.001}$ is inappropriate, because the magnetic dilution effect of even Zr^{4+} on magnetic susceptibilities is much stronger than that of Sc^{3+} (*19*).

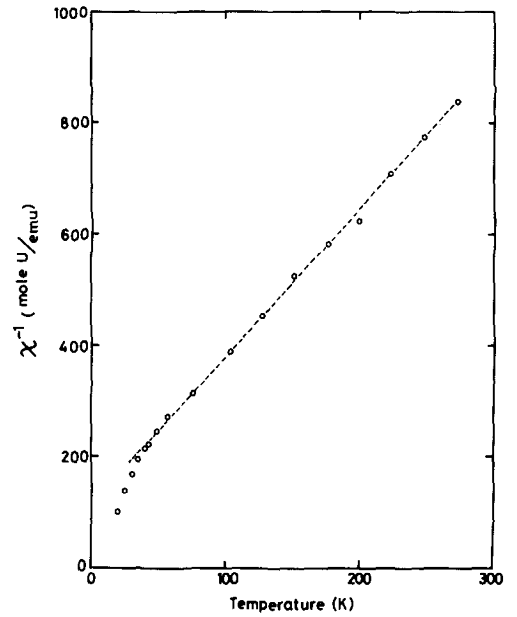

FIG. 7. Reciprocal magnetic susceptibility for U^{5+} vs temperature. U^{5+} is formed due to substitution of Sc^{3+} for U^{4+}.

Since the difference in Sc^{3+} concentration is small, it can be assumed as a first approximation that $\chi'(U^{4+})$ is equal to $\chi(U^{4+})$ and that $\chi'(U^{5+})$ is equal to $\chi(U^{5+})$. The susceptibility of U^{5+} will be

$$\chi(U^{5+}) = \frac{1}{2(x_3 - x_4) + (y_3 - y_4) - 2(x_3y_4 - x_4y_3)} \times [(1 - 2x_4 - 2y_4)\chi(S.3) - (1 - 2x_3 - 2y_3)\chi(S.4)]. \quad (10)$$

By substituting the values of x_3, x_4, y_3, and y_4 into Eq. (10), the following equation is obtained:

$$\chi(U^{5+}) = 16.29\chi(S.4) - 14.38\chi(S.3). \quad (11)$$

By using the experimental susceptibilities for $\chi(S.3)$ and $\chi(S.4)$, the magnetic susceptibility for U^{5+} can be obtained. Figure 7 shows its reciprocal susceptibility vs temperature curve. It is seen that the Curie–Weiss law holds from ca. 40 K to room temperature. The effective magnetic moment is 1.74 B.M. This value is a little lower than that calculated for $5f^1$ electronic configuration in a crystalline field with cubic symmetry, but this magnitude of magnetic moment is observed in uranates of alkaline earth elements with fluorite structure MU_2O_6 (M = Ca, Sr, Ba) (*20*). Therefore, it can be said that the substitution of Sc^{3+} for U^{4+} causes the oxidation of U^{4+} to U^{5+} and decreases effective magnetic moment in the solid solution. The reason that the effective magnetic moment determined for U^{5+} which is oxidized due to substitution of Sc^{3+} is not consistent with the value of 2.00 B.M. is considered to be inequality of magnetic dilution effect in $Sc_{0.05}U_{0.95}O_{1.998}$ and $Sc_{0.10}U_{0.90}O_{2.001}$.

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