Growth and Characterization of High-Quality $YTiO_3$ Single Crystals: Minimizing Ti^{4+} Containing Impurities and TiN Formation

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Table S1. The details of structure refinement for $YTiO_3$ single crystals at room temperature. The code 2392602, 2392601, and 2392600 are assigned to $YTiO_3$ single crystals grown under different conditions: (A) from the nominal composition $YTiO_3$ at a growth rate of 10mm/h; (B) from the nominal composition $YTiO_{2.925}$ at a growth rate of 4 mm/h; (C) from the nominal composition $YTiO_{2.85}$ at a growth rate of 4 mm/h.

CCDC Reference	2392602	2392601	2392600
Chemical formula	$YTiO_3$	$YTiO_3$	$YTiO_3$
M_r	184.81	184.81	184.81
Crystal system	Orthorhombic	Orthorhombic	Orthorhombic
Space group	*Pnma*	*Pnma*	*Pnma*
Temperature (K)	299	299	299
a (Å)	5.6807(3)	5.6860(3)	5.6995(1)
b (Å)	7.6204(4)	7.6108(3)	7.6297(2)
c (Å)	5.3391(2)	5.3378(2)	5.3455(1)
V (Å^3)	231.13 (2)	230.99 (2)	232.45 (1)
Z	4	4	4
λ (Å)	0.71073	0.71073	0.71073
μ (mm^{-1})	28.14	28.15	27.98
Density (Mg/m^3)	5.311	5.314	5.281
Crystal size (mm)	0.1 × 0.04 × 0.03	0.09 × 0.03 × 0.01	0.16 × 0.14 × 0.12
T_{min}, T_{max}	0.460, 0.819	0.453, 0.873	0.229, 0.408
No. of measured, independent, and observed [$I > 2\sigma(I)$] reflections	1607, 378, 355	1558, 373, 352	1596, 380, 370
Absorption	Gaussian	Gaussian	Gaussian
Completeness (%)	99.6	98.7	100.0
R_{int}	0.016	0.017	0.014
$R_{[F2 > 2\sigma(F2)]}$, $wR_{(F2)}$	0.019, 0.051	0.020, 0.053	0.016, 0.041
S (GOOF)	1.14	1.07	1.08
No. of reflections	378	373	380
No. of parameters	28	28	29
$\Delta\rho_{max}$, $\Delta\rho_{min}$ (e Å−3)	1.20, −0.60	0.83, −0.78	0.82, −1.08

Thermogravimetric analysis (TGA) was conducted using a Setaram Labsys TGA-DSC thermal analyzer. The black $YTiO_3$ powder previously used for X-ray diffraction measurements (refer to Figure 6 in the paper) was analyzed to assess the oxygen content in the single crystals. As discussed in the paper, $YTiO_3$ single crystals grown from a nominal composition of $YTiO_{2.925}$ at a growth rate of 4 mm/h are free from impurity phases. A sample of the black powder, with a mass of 227.7 mg, was placed in a crucible and heated to 1100 °C in flowing air at a rate of 5 °C/min, with the temperature maintained at 1100 °C for 2 hours.

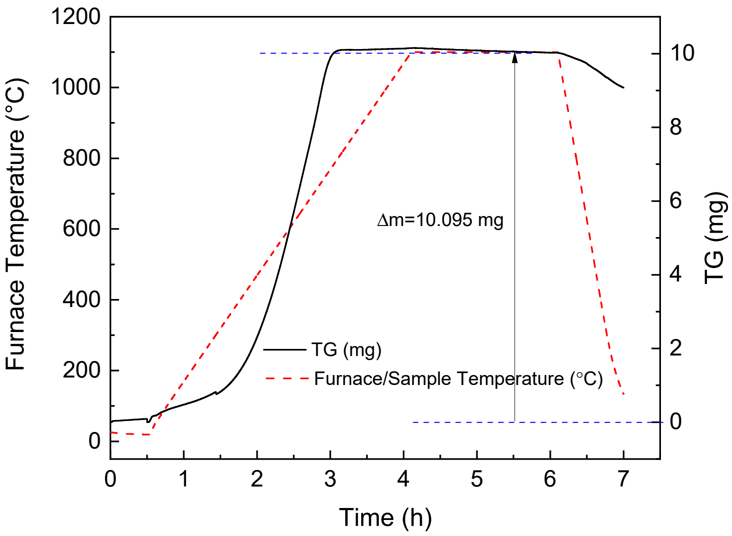

Figure S1. Mass change of $YTiO_3$ powder during heating under air up to 1100 °C.

Figure S1 illustrates the mass change of $YTiO_3$ powder during heating. Above 400 °C, the mass increases almost linearly, reaching a plateau around 800 °C. After the experiment, the black powder changes to a white color, confirmed to be $Y_2Ti_2O_7$. The mass of the white powder, measured at room temperature, is 9.6 mg—slightly less than the 10.095 mg observed in the TGA measurement.

The transformation involves the following chemical reaction during heating:

$$YTiO_{3+x} + (0.25\text{-}x/2)O_2 \rightarrow YTiO_{3.5}.$$

The $YTiO_3$ compound is oxidized during heating and transforms into the $Y_2Ti_2O_7$ ($YTiO_{3.5}$) phase. The mass gain corresponds to $(0.25-x/2)O_2$. Based on the two values, Δm = 10.095 mg and 9.6 mg, x is calculated to be -0.0126 and 0.0126, respectively. Thus, the oxygen content in the single crystals is determined to be between 3 - 0.0126 and 3 + 0.0126. The electronic analytical balance we used has a readability of 0.0001g (0.1 mg). The accuracy of balance in the thermal analyzer is superior to the electronic analytical balance. However, the large thermal drift was induced by the changes in ambient temperature for the magnets and associated coils and by some internal parts dilatation. The error is estimated to be 0.1 mg from the electronic analytical balance, and the value 3 + 0.0126 is favorable. Combining this with the powder and single-crystal XRD measurements, we conclude that the oxygen content in $YTiO_3$ single crystals is unambiguously stoichiometric, with an O content of 3.