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COMMUNICATION

Single Crystalline Ternary Mixed Metal Oxide 1-Dimensional Nanostructures of Ir$_{1-x}$Ru$_x$V$_2$O$_2$ by Vapour Phase Transport

Hayoung Jung, Sung Hee Chun, Jeesoo Seok, Yu Lim Kim, Seung Joon Lee, Myung Hwa Kim, and Nam-Suk Lee

We report the growth of the single crystalline ternary mixed metal oxide nanowires consisting of Ir, Ru, V, and O via a vapour phase transport process by carefully controlling the relative ratios of IrO$_2$, RuO$_2$, and VO$_2$ precursors, which have the tetragonal rutile crystal structure with the similar lattice parameters for the first time. We clearly observed that various compositions of Ir$_{1-x}$Ru$_x$V$_2$O$_2$ nanowires were readily modulated by randomly incorporating Ir$^{4+}$, Ru$^{4+}$, and V$^{4+}$ metal ions into the tetragonal lattice unit owing to the similar ionic sizes as confirmed from STEM elementary mapping as well as X-ray diffraction pattern for analysis of the crystal structure of mixed metal oxide nanowires.

INTRODUCTION

The synthetic strategy incorporating two or more transition metals into a single metal oxide lattice unit has been a long standing issue to develop new functional materials for building blocks in a variety of industrial electrochemical processes for alternative energy applications and as heterogeneous catalysts. The main reason is that the favourable enhancements of their chemical, mechanical, electrical and optical properties could be achieved compared with those of the single component metal oxides. Particularly, the nanostructured mixed metal oxides containing with novel transition metals such as Ir, Ru, Pt, etc... are currently one of the extremely promising candidates as a highly efficient electrode material and electrocatalysts in electrochemical devices owing to its superior electrocatalytic activity, high electrical conductivity, chemical stability, and excellent diffusion barrier properties. However, very limited synthetic approaches such as sol-gel process, thermal decomposition, and reactive sputtering have been previously employed to mostly prepare the binary mixed metal oxide thin films or nanoparticles in order to enhance the electrochemical activity as well as stability in electrochemical devices. Unfortunately, mixed metal oxides obtained from these methods often represent the significant local inhomogeneity in the crystal structure which can be poorly defined crystal structures rather than single phases. Thus, it is still of great challenge to make well-defined single crystalline mixed metal oxides in even ternary systems to be useful for a variety of applications.

On the other hand, we recently introduced that highly single crystalline Ir$_x$Ru$_{1-x}$O$_2$ and Ru$_{1-x}$V$_2$O$_2$ mixed metal oxide nanowires can be prepared via a simple physical vapour transport process by controlling the relative ratios of two precursors, IrO$_2$ and RuO$_2$, VO$_2$ and RuO$_2$, respectively. In our previous work, interestingly, since both metal oxides have the same tetragonal crystal structure and the similar ionic radius in the tetravalent ionic state, the binary mixture systems readily formed even a continuous solid solution over the whole or partial compositional range between two metal oxides. In fact, this simplicity of the nanowire growth suggests a certain degree of generality so that it could be readily extended to the synthesis of a variety of mixed metal oxide systems including ternary or even quaternary metal oxide systems.

In this communication, therefore, we introduce the first time attempt to synthesize the ternary mixed metal oxide of 1-dimensional Ir$_{1-x}$Ru$_x$V$_2$O$_2$ nanostructures in the form of a single crystal structure via a simple vapour phase transport process by carefully controlling the relative ratios of IrO$_2$, RuO$_2$, and VO$_2$ precursors, respectively. It is likely because three metal oxides have the same crystal structure in a tetragonal phase as well as the similar ionic radius, 0.0640 nm for Ir$^{4+}$, 0.0600 nm for V$^{4+}$ and 0.0620 nm for Ru$^{4+}$, respectively. They could readily share the same site on the cationic sub-lattice of a tetragonal phase in the crystal structure. Furthermore, a new synthetic platform for obtaining highly single crystalline ternary metal oxide nanostructures is

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carefully explored by confirming of their crystal structures based on HRTEM, STEM, XPS and XRD analyses.
**EXPERIMENTAL**

We prepared highly single crystalline Ir$_{1-x}$Ru$_x$V$_2$O$_2$ ternary mixed metal oxide nanowires directly on a Si(001) wafer by a vapour phase transport process starting from the mixtures of IrO$_2$, RuO$_2$, VO$_2$ powders without purification and any foreign catalyst under atmospheric pressure as illustrated in Fig. 1. Specifically, Ir$_{1-x}$Ru$_x$V$_2$O$_2$ nanowires were synthesized by the vapour phase transport process in a single zone horizontal quartz tube furnace, 2.5 cm in diameter and 60 cm long under atmospheric pressure in Fig. 1. First, various weight percent of fine meshed RuO$_2$, and IrO$_2$ (99.9%, Aldrich) powders were mixed and then sonicated for homogenous mixing of two precursors. Various compositions of IrO$_2$-RuO$_2$ mixed metal oxide precursors were taken and then loaded at the centre of a 6 cm long quartz boat as given in Table 1. The quartz boat containing IrO$_2$-RuO$_2$ mixed metal oxide precursors was placed at the centre of the quartz tube furnace which is the highest temperature region. In addition, various compositions of VO$_2$ powder on another quartz boat were located at 5 cm downstream of IrO$_2$-RuO$_2$ mixed metal oxide precursors as given in Table 1. Furthermore, the growth substrate on a quartz boat was also introduced into the furnace at a point approximately ~15 cm downstream of the IrO$_2$-RuO$_2$ mixture powder source location. The quartz boat and mixed oxide charge were cleaned to get rid of impurities by first placing them at the centre of the quartz tube furnace under He 300 SCCM before heating. After that, the furnace temperature was rapidly increased to a temperature in the range of 1050°C with flowing He (99.999%) carrier gas of 400 SCCM at the rate of 100°C/min. The nanowire growth was proceed as flowing high purity He (99.999%, 365 ~ 450 SCCM) and O$_2$ (99.9%, 3~15 SCCM) for 1 h as given in Table 1. The temperature of the region which obtained the nanowires on the substrate was measured by ~700°C. The furnace was finally allowed to cool to room temperature under the same flow rate of He. The product collected on the Si(001) substrate was collected and characterized by scanning electron microscopy (SEM) and micro Raman spectroscopy. X-ray diffraction (XRD) pattern was obtained using Rigaku diffractometer with Ni filtered Cu K$_\alpha$ radiation, $\lambda = 0.15418$ nm at 25°C. Ir$_{1-x}$Ru$_x$V$_2$O$_2$ nanowires were also imaged by high resolution transmission electron microscopy (HRTEM, Cs-corrected STEM, JEOL JEM-2100F) at 200 kV. Samples for TEM imaging were prepared by simply transferring some of nanowires to a TEM grid.

Table 1. The detailed experimental conditions and chemical compositions of various Ir$_{1-x}$Ru$_x$V$_2$O$_2$ mixed metal oxide nanowires estimated by HRTEM-EDS measurements. Note that error bars were carefully estimated by measuring 10 different nanowires for each composition.

<table>
<thead>
<tr>
<th>Weight of Precursor (mg)</th>
<th>Gas Flow (SCCM)</th>
<th>Ir$_{1-x}$Ru$_x$V$_2$O$_2$</th>
<th>1-x-y</th>
<th>x</th>
<th>y</th>
</tr>
</thead>
<tbody>
<tr>
<td>IrO$_2$ &amp; RuO$_2$ &amp; VO$_2$</td>
<td>450 &amp; 3</td>
<td>2.9 &amp; 1.7 &amp; 1.3</td>
<td>0.06 &amp; 0.41 &amp; 0.53 &amp; ($\pm$0.02) &amp; ($\pm$0.02) &amp; ($\pm$0.03)</td>
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<tr>
<td>4.8 &amp; 1.7 &amp; 0.7</td>
<td>0.10 &amp; 0.36 &amp; 0.54 &amp; ($\pm$0.08) &amp; ($\pm$0.06) &amp; ($\pm$0.02)</td>
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<tr>
<td>3.2 &amp; 2.3 &amp; 2.3</td>
<td>0.12 &amp; 0.34 &amp; 0.54 &amp; ($\pm$0.03) &amp; ($\pm$0.06) &amp; ($\pm$0.03)</td>
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<tr>
<td>3.1 &amp; 5.7 &amp; 1.2</td>
<td>0.14 &amp; 0.74 &amp; 0.12 &amp; ($\pm$0.01) &amp; ($\pm$0.02) &amp; ($\pm$0.01)</td>
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<tr>
<td>3.1 &amp; 1.6 &amp; 1.2</td>
<td>0.23 &amp; 0.34 &amp; 0.43 &amp; ($\pm$0.01) &amp; ($\pm$0.01) &amp; ($\pm$0.02)</td>
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<tr>
<td>5.1 &amp; 3.9 &amp; 1.5</td>
<td>0.39 &amp; 0.38 &amp; 0.23 &amp; ($\pm$0.05) &amp; ($\pm$0.05) &amp; ($\pm$0.02)</td>
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<tr>
<td>5.3 &amp; 1.9 &amp; 2.4</td>
<td>0.40 &amp; 0.44 &amp; 0.16 &amp; ($\pm$0.06) &amp; ($\pm$0.07) &amp; ($\pm$0.02)</td>
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**RESULTS AND DISCUSSION**

Fig. 2 and Fig. S1 presents typical SEM images for various compositions of iridium-ruthenium-vanadium contained ternary nanostructures directly grown on a Si substrate. High magnification SEM images clearly represent that nanowires mostly were grown along the basal plane of a Si substrate and they also show a rectangular cross-section with well-defined sharp facets at the end of tip of nanowires (Fig. 2 and Fig. S1). The average lateral dimension and the length of nanowires for all compositions are approximately 200 nm and up to 3 µm,
nanowires was rarely obtained. The shape of products at high oxygen flow resembles microsized crystals with sharp facets. The concentration of oxygen gas was thus a critical factor to obtain the high aspect ratios of Ir$_{1-x}$Ru$_x$V$_y$O$_2$ ternary mixed metal oxide nanowires. In addition, since the vapour pressure of intermediate species of vanadium formed inside quartz tube is relatively the higher vapour pressure compared to that of the mixture precursor of IrO$_2$ and RuO$_2$. The location of VO$_2$ precursor was then placed at lower temperature region in a quartz tube for controlling the content of vanadium in Ir$_{1-x}$Ru$_x$V$_y$O$_2$ ternary mixed metal oxide nanowires.

Fig. 3 and Fig. S2 show XRD patterns for as grown mixed metal oxide nanowires for various compositions of IrO$_2$-RuO$_2$-VO$_2$ ternary system, indicating of the highly single crystalline nature. Especially, All XRD patterns for Ir$_{1-x}$Ru$_x$V$_y$O$_2$ ternary mixed metal oxide nanowires represent only a few sharp main peaks, near 28°, 40° and 57°, which are attributed to (110), (200), and (220) crystallographic planes, closely matched with the rutile structure of pure IrO$_2$, RuO$_2$ and VO$_2$ in the tetragonal phase from the standard data (JCPDS 15-0870 for IrO$_2$, JCPDS 40-1290 for RuO$_2$ and JCPDS 44-0253 for VO$_2$) 15-17. Strong intensity peak of the (110) plane for all Ir$_{1-x}$Ru$_x$V$_y$O$_2$ ternary mixed metal oxide nanowires could be rationalized by considering the in-plane preferential growth of the rectangular cross-section nanowires on a Si substrate. On the other hand, the mixed metal oxide nanowires with the high content of Ru show out of plane growth similar with the growth of pure RuO$_2$ nanowires in Fig. S2. In the case of Ir$_{1-x}$Ru$_x$V$_y$O$_2$ ternary mixed metal oxide nanowires as shown in Fig. 3, the X-ray scattering signals are mostly originated from the top crystal plane of nanowires, which corresponds to a (110) crystallographic plane of the tetragonal structure with the rectangular cross-section. Additionally, few indexed peaks indicate that the homogeneous solid solution of three metal oxides could be achieved in some range of compositions. Furthermore, Table S1 exhibits the variations of the d-spacing for a (110) indexed crystal plane with respect to the relative contents of three metals of Ir, Ru, and V in the ternary mixed

![Fig. 2 SEM images of as-grown iridium-ruthenium-vanadium ternary mixed metal oxide nanowires on a Si(001) substrate by a vapour phase transport process. (a) and (b) Ir$_{1.12}$Ru$_{2.34}$V$_{0.52}$O$_2$, (c) and (d) Ir$_{0.95}$Ru$_{2.25}$V$_{0.54}$O$_2$, (e) and (f) Ir$_{1.12}$Ru$_{2.34}$V$_{0.52}$O$_2$, (g) and (h) Ir$_{1.12}$Ru$_{2.25}$V$_{0.54}$O$_2$, (i) and (j) Ir$_{2.46}$Ru$_{0.40}$V$_{0.54}$O$_2$.](image1)

respectively. At a first glance, since nanowire’s tips do not have any catalyst particles, it might be reasonably expected that the vapour-solid (VS) growth could be responsible for the growth process of ternary iridium-ruthenium-vanadium ternary oxide nanowires. Furthermore, the composition of three metal contained nanowires were quantitatively determined by careful HRTEM-EDS measurements in Table 1. Interestingly, the vanadium content of as grown mixed metal oxide nanowires is highly dependent of oxygen gas flow compared with iridium and ruthenium as shown in Table 1. Our results indicate the content of vanadium is relatively low at higher flow rate of oxygen gas in Ir$_{1-x}$Ru$_x$V$_y$O$_2$ ternary mixed metal oxide nanowires. Without oxygen flow, the growth of
oxide nanowires. The d-spacings for a (110) indexed crystal plane were directly obtained from XRD measurements and calculated values were estimated with the reference data of pure metal oxides of IrO$_2$, RuO$_2$, and VO$_2$ from JCPDS by using the following equation.

\[
d_{x\text{IrO}_2, x\text{RuO}_2, x\text{VO}_2} = x_{\text{IrO}_2}d_{\text{IrO}_2} + x_{\text{RuO}_2}d_{\text{RuO}_2} + x_{\text{VO}_2}d_{\text{VO}_2}
\]

where x is the partial fraction in terms of atomic percent and d is the pure d-spacing of IrO$_2$, RuO$_2$, and VO$_2$ for a (110) plane from JCPDS. The results thus are in close values with the prediction of Vegard’s rule for an ideal solid solution.$^{18,19}$

The crystalline structure of Ir$_{1-x}$Ru$_x$V$_y$O$_2$ ternary mixed metal oxide nanowires was confirmed with HRTEM measurement and the chemical compositions were carefully determined by EDS-elemental mapping analysis in a single Ir$_{1-x}$Ru$_x$V$_y$O$_2$ ternary mixed oxide nanowire as shown in Fig. 4 and Fig S3. Fig. 4a and 4b represent a low magnification TEM image and a high resolution TEM image of a single Ir$_{0.12}$Ru$_{0.34}$V$_{0.54}$O$_2$ nanowire along the [111] zone axis, indicating that a nanowire has the single crystalline nature without the existence of the core/shell structures or alternative layer structures. In Fig. 4b, the interplanar distances of adjacent planes are clearly identified by 0.2368 nm and 0.2198 nm, which are the similar values for the (011) and (020) planes of the tetragonal structures of IrO$_2$, RuO$_2$ and VO$_2$ unit cells, respectively. The results show the spacing of the (011) and (020) planes are slightly shifted along the incorporation of the relative amounts of three metals into the tetragonal lattice compared with those of three pure metal oxides, reasonably consistent with the results of XRD patterns. The growth direction of a nanowire is carefully estimated by along the [001] direction from the Fast Fourier Transform (FFT) of the lattice resolved image in Fig. 4b (inset). It is quite reasonable because the c-

Fig. 4 (a) The low magnification TEM image and (b) the lattice resolved HRTEM image of a single Ir$_{0.12}$Ru$_{0.34}$V$_{0.54}$O$_2$ ternary mixed metal oxide nanowire. Inset represents the Fast Fourier Transform (FFT) of the lattice resolved image. (c) High angle annular dark field (HAADF) STEM image of a single Ir$_{0.12}$Ru$_{0.34}$V$_{0.54}$O$_2$ ternary mixed metal oxide nanowire. (d) EDS-elemental mapping analysis for Ir(M), Ru(L) and Ir(M) atoms of a single Ir$_{0.12}$Ru$_{0.34}$V$_{0.54}$O$_2$ ternary mixed metal oxide nanowire.
axis in the tetragonal structure is generally the preferential growth direction. Additionally, Fig. 5c and 4d show a high angle annular dark field (HAADF) STEM image of a single $\text{IrO}_x\text{Ru}_y\text{V}_z\text{O}_{2x+y+2z}$ ternary mixed metal oxide nanowire and the corresponding spatial distribution of Ru, V, and O atoms examined with EDS-element mapping analysis. All atoms of a single $\text{IrO}_x\text{Ru}_y\text{V}_z\text{O}_{2x+y+2z}$ ternary mixed metal oxide nanowire are homogeneously distributed through the whole part of the nanowire. This result thus supports that a homogenous solid solution of a $\text{IrO}_x\text{Ru}_y\text{O}_{2x+y}$ ternary mixed metal oxide system can be formed, quite consistent with the result of XRD measurements. On the other hand, it should be noted that the segregation of domains at much smaller length scales cannot be completely ruled out as dimers or nanoscale clusters due to the limited spatial resolution of EDS.

Furthermore, the high resolution XPS measurements reveal the surface composition and the chemical states of elements in $\text{IrO}_x\text{Ru}_y\text{V}_z\text{O}_{2x+y+2z}$ ternary mixed metal oxide nanowires in Fig. 5 and Fig. S4. The XPS spectrum for Ir 4f in Fig. 5a shows two predominant peaks at the binding energy of 61.9 eV and 64.9 eV, which are readily assigned to the 4f$^7/2$ and 4f$^5$ spin orbit doublet pair of IrO$_2$ consistent with the literature values, respectively. It is immediately apparent that although the broad band of carbon contained species of C 1s is located at around 285.3 eV, the peak at 281.1 eV for Ru 3d is also the characteristic binding energy of RuO$_2$ in Fig. 5b. For XPS spectrum of V 2p in Fig. 5c, it could be reasonable that the slightly broad peak at 517.1 eV corresponds to VO$_2$ species for V 2p$^3/2$, but the presence of higher oxidation states of V like V$_2$O$_5$ are not completely excluded based on the broad feature around 517.6 eV. It is likely because there might be the contribution from many droplet-like structures near nanowires which are identified as V$_2$O$_5$ similar to the case of the growth of VO$_2$. On the other hand, while the sharp peak at 530.0 eV for O 1s is originated from the direct binding of oxygen with metals in the crystal lattice, the broad feature around 533.7 eV could be assigned to various oxygen bounded species on the surface. It implies that the surface chemical states of $\text{IrO}_x\text{Ru}_y\text{V}_z\text{O}_{2x+y+2z}$ ternary mixed metal oxide nanowires mainly consist of $\text{Ir}^{4+}$, $\text{Ru}^{4+}$, and $\text{V}^{4+}$ ions as confirmed by HRTEM and XRD measurements.

As mentioned before, an interesting observation is that many droplet-like structures near nanowires are seen and then the droplet density is dramatically depleted near nanowires similar to the case of VO$_2$ pure nanowires and $\text{Ru}_{1-x}\text{V}_x\text{O}_2$ mixed metal oxide nanowires we reported in Fig. 2. It suggests that the nanowire growth process might be closely related to the consumption of the supercooled liquid nanodroplets of V$_2$O$_5$ intermediate species as already reported in pure VO$_2$ and Ru$_{1-x}\text{V}_x\text{O}_2$ nanowire growth. For a ternary mixture system of $\text{IrO}_x\text{Ru}_y\text{V}_z\text{O}_{2x+y+2z}$, in fact, it is reasonably expected that it is possible to have some compositional ranges to be continuous solid solutions between three metal oxides. They could readily share the same site on the cationic sub-lattice of a tetragonal phase in the crystal structure owing to the same crystal structure as well as the similar ionic radius. Thus, the preferential nucleation of gas phase $\text{IrO}_x\text{Ru}_y\text{V}_z\text{O}_{2x+y+2z}$ precursors, which can be formed by the oxidation of VO$_2$, IrO$_2$, and RuO$_2$ precursors under the tiny amount of O$_2$ gas flow at high temperature, plays a crucial role in the formation of the ternary mixed metal oxide nanowires. In fact, the gas phase $\text{IrO}_x\text{Ru}_y\text{O}_{2x+y}$, and $\text{V}_2$O$_5$ intermediate species can be effectively generated at high temperature and then they are much more volatile than those of RuO$_2$, IrO$_2$, and VO$_2$ at high temperature. It should be pointed out that the nanowire growth is highly dependent of the flow of the oxygen gas at the growth temperature so that the well-defined $\text{IrO}_x\text{Ru}_y\text{V}_z\text{O}_{2x+y+2z}$ nanowires was only found within a limited range of the oxygen flow rate from 3 to 15 SCCM in our study. It would be thus expected that the most volatile V$_2$O$_5$ species could be the form.

![Fig. 5 X-ray Photoelectron Spectroscopy (XPS) data of $\text{IrO}_x\text{Ru}_y\text{V}_z\text{O}_{2x+y+2z}$ mixed metal oxide nanowires for (a) Ir 4f, (b) Ru 3d, and (c) V 2p.](image-url)
of nanodroplets initially on a substrate, which are strongly suppressed in the freezing point compared to that of bulk due to their small size and interactions with the substrate.\textsuperscript{17,18} In the next process, IrO\textsubscript{3} and RuO\textsubscript{2} precursors could be dissolved into nanodroplets of V\textsubscript{2}O\textsubscript{5} to be a liquid mixture solution. Ostwald ripening or coalescence process could take place to further grow these mixed nanodroplets and it then might be supercooled before starting crystallization on the substrate. Finally, these mixed nanodroplets proceed the crystallization along the favourable growth direction of c-axis in tetragonal crystal phase, with the continuous feedback of the mixed nanodroplets from the condensation of gas phase precursors as illustrated in Fig. 6. Although the real-time mechanistic study such as in situ TEM or GISAXS (grazing incident small angle X-ray scattering) technique is required to elucidate the growth mechanism, present results propose that the growth of Ir\textsubscript{1-x},Ru\textsubscript{x}V\textsubscript{2}O\textsubscript{5} nanowires could be slightly different from the conventional vapour-liquid-solid (VLS) and vapour-solid (VS) growth.\textsuperscript{17} It rather closes to the growth of pure V\textsubscript{2}O\textsubscript{5} nanowires and Ru\textsubscript{1},V\textsubscript{2}O\textsubscript{5} mixed metal oxide nanowires, which are involved in the formation of supercooled liquid nanodroplets of V\textsubscript{2}O\textsubscript{5} species. Consequently, various compositions of Ir\textsubscript{1-x},Ru\textsubscript{x}V\textsubscript{2}O\textsubscript{5} ternary mixed metal oxide nanowires were successfully grown via the simple vapour phase transport process without catalysts by just controlling relative weight ratios of three precursors before the growth process and the flow of oxygen gas at the growth temperature. Since this synthetic strategy is simple, prompt, and versatile, it makes possible to extend for obtaining a variety of ternary mixed metal oxides or even quaternary mixed metal oxides toward applications in the field of high performance catalysts.

Fig. 6 A schematic figure of the nanowire growth process on a Si/SiO\textsubscript{2} wafer.

Conclusions

We describe the novel synthesis of highly crystalline Ir\textsubscript{1-x},Ru\textsubscript{x}V\textsubscript{2}O\textsubscript{5} ternary mixed metal oxide nanowires in various compositions via a simple vapour phase transport process originated from IrO\textsubscript{3}, RuO\textsubscript{2}, and V\textsubscript{2}O\textsubscript{5} precursors. The results represent that various compositions of Ir\textsubscript{1-x},Ru\textsubscript{x}V\textsubscript{2}O\textsubscript{5} nanowires were readily modulated by randomly incorporating of Ir\textsuperscript{4+}, Ru\textsuperscript{3+}, and V\textsuperscript{5+} metal ions into the same tetragonal lattice unit due to the similar ionic sizes. The crystalline nature of Ir\textsubscript{1-x},Ru\textsubscript{x}V\textsubscript{2}O\textsubscript{5} ternary mixed metal oxide nanowires was also confirmed from XRD as well as HRTEM analysis. In addition, it could be suggested that the nanodroplets like V\textsubscript{2}O\textsubscript{5} intermediate species may play a crucial role in growing Ir\textsubscript{1-x},Ru\textsubscript{x}V\textsubscript{2}O\textsubscript{5} ternary mixed metal oxide nanowires, different from the conventional growth mechanisms such as VLS and VS.

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References

Highly single crystalline Ir$_{1-x}$Ru$_x$V$_y$O$_2$ nanowires were grown from metal oxide precursors on a Si wafer by a vapour transport process.