Supplementary Figure 1. Comparison between sapphire ($\alpha$-Al$_2$O$_3$) and hematite ($\alpha$-Fe$_2$O$_3$).

The two-dimensional projection of the lattice structure of sapphire (a) and hematite (b). In Supplementary Fig. S1a, red balls refer to O$^{2-}$ and white balls refer to Al$^{3+}$; In Supplementary Fig. S1a, red balls refer to O$^{2-}$ and Green balls refer to Fe$^{3+}$. The structures of $\alpha$-Al$_2$O$_3$ and $\alpha$-Fe$_2$O$_3$ are compared firstly. They have high similarity that they are all trigonal crystal system, oxygen atoms are arranged in a hexagonal close-packed and Al$^{3+}$ and Fe$^{3+}$ are filled in the octahedral void respectively. In the structure of $\alpha$-Al$_2$O$_3$ or $\alpha$-Fe$_2$O$_3$, only 2/3 octahedral void are occupied. On the other hand, Al$^{3+}$ and Fe$^{3+}$ share the nearly same radius, 53.5 pm and 55.0 pm respectively. It provides the possibility that the Fe$^{3+}$ enters in the $\alpha$-Al$_2$O$_3$, in that Fe$^{3+}$ can be arranged in the left octahedral void or substituted the Al$^{3+}$, when $\alpha$-Al$_2$O$_3$ and $\alpha$-Fe$_2$O$_3$ are annealed together in the air at high temperature.
Supplementary Figure 2. Entropy analysis of the “dissolve” process. (a) The schematic illustration of the interinfiltration process, purple balls refer to Al$^{3+}$ while white balls refer to Fe$^{3+}$; (b) the entropy change during the interinfiltration process. $P$ refer to the proportion of doping. With the same valence and similar diameter, we believe that the enthalpy change of the dissolve process approach to 0, which means the process is not driven by enthalpy. As shown in Supplementary Fig. S2, we suppose that this process is driven by entropy, for the dissolve process increase the chaos of the system. It is easy to proof that the entropy maximize when the two layers have the same content of Fe and Al.
Supplementary Figure 3. XPS depth analysis of the substrates without annealing loaded with catalysts. (a) Depth analysis data. The vertical axis is the ratio of atom number between Fe and Al, in which the number of Al atom is considered to be a constant; (b) XPS spectra of each point.
Supplementary Figure 4. XPS depth analysis of the substrates after annealing loaded with catalysts. The red line is the fitting result of the XPS depth analysis of Fe after annealing. Due to the first point contains more surface information, it is not considered in the fitting. Suppose the content of Fe has an exponential decay from the first layer to the inner layers, let \( k \) to be the proportional constant, and \( \rho^{(N)} \) to be the content of Fe in the \( N \text{th} \) layer. Then we can get the relationship between \( \rho^{(N)} \) and \( \rho^{(1)} \):

\[
\rho^{(N)} = \rho^{(1)} k^N
\]  

(1)

And the average content \( c \) of a certain depth \( d \) should be:

\[
c = \frac{1}{n} \sum_{i=1}^{n} \rho^{(i)} = \frac{\rho^{(1)}}{n} \frac{1 - k^n}{1 - k}, \quad n = \frac{d}{\delta}
\]

(2)

In which \( \delta \) is the distance between two adjacent layers.

From Supplementary equation 2 we can see that \( c \) is only relates to content the first layer if \( d, \delta \) and \( k \) are constant.

In sapphire, \( \delta = 0.2165 \text{ nm} \), and \( n = 14 \) layers.
According to the fitting result, we can get k and $\rho^{(1)}$

$$14 \ln k = -0.131, \quad k = 0.972 \quad (3)$$

$$\rho^{(1)} = \frac{0.013 \times (1 - q) \times 14}{1 - q^{14}} = 0.0154 \quad (4)$$

As mentioned above, to realize the greatest entropy change, k should be close to 1. Our result is close to the ideal value.
Supplementary Figure 5. Analyze the requirement of Fe amounts to grow SWNTs with high
density. (a) two-dimensional projection of sapphire crystal structure, white balls refer to Al$^{3+}$ while
red balls refer to O$^{2-}$; (b) an SWNT with the diameter of 1.5 nm and the length of 1000 nm.
Supplementary Fig. S5a shows a two-dimensional projection of sapphire crystal structure. Atoms in
black circles are oxygen atoms in the same plane. We can calculate the areal density of Al in a
single layer:

$$S = a^2 2 \sin \frac{1}{3} \pi = 0.20 \text{nm}^2, \quad \rho_{Al} = \frac{N}{S} = 10 \text{nm}^2$$  \hspace{1cm} (5)

In which a is the cell parameter of sapphire.

According to Supplementary Fig. 4, the content of Fe in the first layer is 0.0154, so

$$\rho_{Fe} = \rho_{Al} \times k = 0.154 \text{nm}^{-2}$$  \hspace{1cm} (6)

Supplementary Fig. S5b shows an SWNT with the diameter of 1.5 nm and the length of 1000 nm.
This SWNT can cover the area of 7000 nm$^2$, which contains about 270 atoms. The amount of Fe
atom can form a nanoparticle with the diameter over 2 nm, which is big enough for such an SWNT.
In fact, our high density (130 tubes/µm) is 5 times smaller than the ideal density (670 tubes/µm) if
the SWNTs are closely packed on the lattice of sapphire, and the length of the SWNT is more than
200 times larger than 1 µm. So if 0.1% Fe(III) in the first layer are reduced to Fe as catalyst, it is
enough to for us to get an high-density arrays.
Supplementary Figure 6. XPS depth analysis in 3 nm depth with different metal ion to ensure whether they have dissolved into sapphire substrates. (a) Mn$^{3+}$; (b) Co$^{3+}$; (c) Cu$^{2+}$; (d) Mo$^{6+}$. 0.05 mmol/L ethanol solutions with different metal ion (Mn, Co, Mo and Cu) are spin coated on sapphire substrates with the same rotate speed, and the substrates are annealed under 1100 °C in the air for 8 h. Then elementary compositions are analyzed by XPS depth analysis to ensure if metal oxides have dissolves. From Supplementary Fig. S6, we can see that Mn and Co can dissolve into sapphire substrate, while others cannot. These results accord with the conditions we mentioned before.
Supplementary Figure 7. Catalyst particles diameters and counts analysis. (a-b) AFM images of substrate after H\textsubscript{2} treatment for 5 min and 10 min. (c-f) Catalyst particles diameters and counts analysis after different treatment conditions: before annealing, H\textsubscript{2} treatment for 5 min, 10min and 30 min, respectively. The diameters of catalyst particles statistics revealed that catalyst particles gathered and became larger. Meanwhile, more and more catalyst particles were released with the reducing time extension. These evidences meant catalysts are released continuously in reducing atmosphere.
Supplementary Figure 8. Detail data of XPS depth analysis to confirm the dissolve and release processes of Fe catalyst. (a) XPS depth analysis of substrate with loaded catalysts after annealing in meffle furnace. (b) XPS depth analysis of the substrate after SWNTs growth. The peaks marked by * are satellite peaks. Depending on XPS depth analysis, it clearly indicated that Fe catalyst is indeed dissolved into sapphire substrate. The intensity of Fe peak would get weaker with the depth increase while the intensity of Al peak is nearly not changed. Furthermore, the shift of Fe peaks on surface and in bulk phase further indicated that some Fe catalyst is on the surface and some under the surface.
Supplementary Figure 9. SWNT growth with other metals. (a-b) SWNTs growth with Co without annealing process (a) and with annealing process (b); (c-d) SWNTs growth with Mn without annealing process (c) and with annealing process (d).
Supplementary Figure 10. Synthesis of high density aligned SWNT arrays combining with ion implantation technology. (a) AFM image of sapphire substrate after the ion implantation process. (b) AFM image of sapphire substrate after another annealing process. (c) XPS depth analysis about the content distribution of iron catalysts in the substrate. (d) SEM images of the as-grown SWNT array by the ion implantation method (inset high-magnification SEM image).
Supplementary Figure 11. The surface morphology of sapphire substrate before and after annealing. AFM image of sapphire substrate before annealing (a) and after annealing (b). After cleaning, it is very clear that many lattice steps on the surface of sapphire substrate, which is brought from mis-cut in substrate producing process. The sapphire substrates must undergo a necessary annealing process (at 1100°C in air for 8 h) for a better crystallization. After annealing, the surface is reconstructed and could develop a comb-like morphology made of parallel strips along the [1-100] direction.
Supplementary Table 1. Structure parameters of $\alpha$-Al$_2$O$_3$ and $\alpha$-Fe$_2$O$_3$. The structure of sapphire ($\alpha$-Al$_2$O$_3$) and hematite ($\alpha$-Fe$_2$O$_3$) are very similar structure, as well as the diameter.

<table>
<thead>
<tr>
<th>Syngony</th>
<th>$\alpha$-Al$_2$O$_3$ (sapphire)</th>
<th>$\alpha$-Fe$_2$O$_3$ (hematite)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Trigonal system</td>
<td>Trigonal system</td>
</tr>
<tr>
<td>$a_0$</td>
<td>476 pm</td>
<td>504 pm</td>
</tr>
<tr>
<td>Ion Radii</td>
<td>53.5 pm</td>
<td>55.0 pm (LS)</td>
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Supplementary Table 2. The stable valence of different metal oxides under 1100 °C in the air and the diameter of corresponding metal ion. To realize the “dissolve” process, the metal oxide should satisfy two conditions: 1) the metal oxide should be stable under 1100 °C in the air; 2) The diameter and the valence of the metal ion should be close enough to the ion radius of Al$^{3+}$. From Supplementary Table S2 we can see that Fe, Co and Mn satisfies the two conditions we mentioned above, which are expected to realize the dissolve process; Mo(VI) has the proper ion radius, but the valence is much higher than Al(III); Cu is not qualified in both valence and radius.

<table>
<thead>
<tr>
<th>Metal</th>
<th>Stable valence</th>
<th>Radius of the ion / nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al</td>
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</tr>
<tr>
<td>Fe</td>
<td>III</td>
<td>0.0550</td>
</tr>
<tr>
<td>Co</td>
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</tr>
<tr>
<td>Mo</td>
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</tr>
<tr>
<td>Mn</td>
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</tr>
<tr>
<td>Cu</td>
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</tr>
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</table>