# Scaled-up catalytic growth of ultra-long carbon nanotubes in the form of macro spools

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**Abstract:** A new process of continuous catalytic growth of centimeter-long carbon nanotubes is presented. A ferrocene-catalyzed growth proceeds at 1150$^0$C in the bottom part of a 5 m high suspended-bed reactor, while a top-seated rotating harvester collects a vertically grown nanotube stalk in a form of a large spool. This allows to translate the unique properties of carbon nanotubes into macro-material. Although there are several methods discovered for synthesizing millimeter- and centimeter-long nanotubes, this work is the first to provide the nanotubes in quantities necessary for producing macro-materials such as threads or re-enforced prepregs.
**Keywords:** Scale up, Carbon nanotubes, Suspended bed.

## 1. Introduction

Carbon nanotube-based materials exhibit properties far below theoretical predictions and even much lower than those for some conventional carbon materials. So it is one of the most challenging targets to translate outstanding properties of carbon nanotubes into macroscopic composite or fiber features. One can suggest rather obvious idea that the synthesis of longer nanotubes would help. Recently, some works like [1, 2] showed that it works and brings better properties. Although there are several methods discovered for synthesizing millimeter- and centimeter-long nanotubes, the macroscopic material requires the nanotubes in quantities, which are difficult or impossible to produce in laboratory bench scale. So it is necessary to scale up, which is a difficult chemical engineering problem for any process, and is especially difficult for such a delicate topochemical reaction. This work represents a scale up of our earlier catalytic process for growing double-walled nanotubes in bench scale [3].

## 2. Experimental

The scaled-up plant (Fig. 1) is based on a floating-catalyst process of decomposition of ethanol-based precursor at 1100-1200°C with hydrogen as a carrier gas. Ferrocene is used as a catalyst precursor. The plant consists of a reactor block, evacuation system, feedgas preparation system and product/waste collection block. All the technological parameters such as temperature, pressure, flowrate, etc. are under automatic control. The core of the reactor block is a 324 mm diameter bottle-shaped reactor made of high-temperature steel and clad with quartz glass. The reactor is heated by means of external electric furnace. The reactor is equipped with a harvesting device, which allows continuous spooling the product nanotubes into big spools of carbon cotton or spun proto-fiber. The spooling device can also grab and pull again any stuck nanotubes if they jam the outlet for some reason.

The produced material was characterized by various techniques including electron microscopy – see Fig. 2. It was shown that it is dominated by well-aligned double-walled carbon nanotubes.

**Figure 1.** Front view of the scaled-up plant for catalytic growth of carbon nanotube spools.

## 3. Results and discussion

Visual observation of carbon nanotube embryonation and growth showed the following pattern: first, the formation of embryonation zone was observed as a light-emitting nebula at the bottom of the reactor; then, in full accordance with mathematical model prediction, the evolution of fast-growing hollow stalk followed, - that took from several minutes; then a steady growth of that hollow stalk pulled by a spooling device. The rotation speed of the spool was controlled in a way to reach balance between the speed of natural growth and the speed of pulling. The growth speed varied from 3 m/min up to 10 m/min. Any attempt to accelerate pulling beyond the natural growth limit resulted in a break of the nanotube stalk and process jamming. The maximum duration of a continuous run was 9 hours.

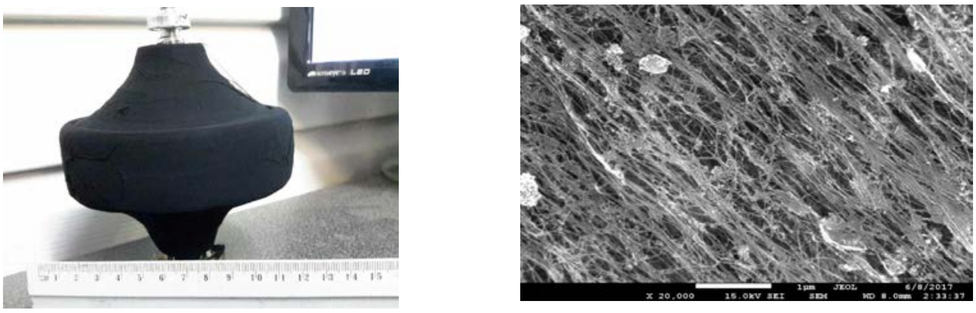


**Figure 2.** The carbon nanotube cotton. Left – a view of a spool; Right – a SEM picture of well-aligned nanotubes inside (residual catalyst is visible, too)

The plant has reached a capacity of 72 g of spooled CNT a day. Further increase of production capacity is planned. Variation of synthesis parameters leads to variation of CNT diameter (from 6-nm DWNT up to 40 nm MWNT) or residual catalyst concentration.

## 4. Conclusions

This successful scale-up development paves the way for intensification of the development in macroscopic carbon nanotube-based fibers and composite materials. The reported pilot plant allows producing long carbon nanotubes in the form of large spools or spreadsheets in kilogram amounts at annual scale from just one catalytic reaction tubes. The appearance of large-scale CNT technology will drive the nanotube prices down and make them easier available for novel composite materials.

## References

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