

A Note of Technology for Developing New Carbon-based Devices to Realize a Low-Carbon Society

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Abstract

The increase in the amount of global CO₂ emissions is evidenced by the fact that the globally averaged temperature of any decade over the past 30 years has been higher than those of all previous decades since 1850. It is therefore imperative to prevent global warming through the stabilization of the concentration of CO₂ by means of a low-carbon society or carbon-positive state. Researchers and scientists hold responsibility for contributing to global environmental conservation through the suppression of carbon use and the energy consumption of newly-developed electronic devices. Huge amounts of energy are consumed by each solid-state device, from production to implementation to operation. To reduce all related energy from technological development to use in our daily lives and to attain a low-carbon society, we have been studying the development of devices that effectively use carbon-based nanomaterials. The methods for reducing the energy consumed in industrial processes and running costs propose the use of carbon nanomaterials in new devices. Accordingly, we have fabricated a planar lighting panel with low power consumption employing highly crystalline single-walled carbon nanotubes as a field emitter and succeeded in achieving a high brightness efficiency of over 80 lm/W. We have further realized process technology and device development technology that maximally utilize CNTs.

INTRODUCTION

In 2017, numerous typhoons affected many Asian countries, particularly Japan, a likely result of recent climate changes. A total of six typhoons struck Japan in 2016 (four in August and two in September), the most since 2004, when ten were recorded [1]. Furthermore, the intensity of these typhoons as well as others that passed close to Japan was more severe than previously. These storms wreaked considerable damage across the country. One of the reasons for this phenomenon is the increase in the seawater temperature around Japan, which means that typhoons approach Japan without weakening. There are several explanations for the increase in seawater temperature, among which is global warming. In 2007, the Fourth Assessment Report of the Intergovernmental Panel on Climate Change (IPCC) concluded that the emission of anthropogenic greenhouse gases causes global warming at a probability of >90%. In addition, the Fifth Assessment Report of the IPCC (2013) reported that the globally averaged temperature data show a warming of 0.85 °C over the period from 1880 to 2012 [2], which is higher than the 0.6 °C of the 100 years from 1901 to 2000 as reported in the Third Assessment Report of the IPCC (2001). In particular, the globally averaged temperature in any decade of the most recent 30 years is higher than that of all previous decades since 1850. One theory holds that an increase in seawater temperature is a phenomenon associated with climate change over a long cycle of approximately one hundred thousand years of cold and warm cycles as a result of changes in the amount of solar radiation reaching the Earth (Milankovitch cycle [3]). However, the increase in average atmospheric temperature observed in the latter half of the twentieth century is too significant to be explained only by a change in solar radiation. The anthropogenic increase in the concentration of greenhouse gases in the atmosphere is considered to be the main cause of the recent global warming. In view of these findings, in 1992, the United Nations adopted the United Nations Framework Convention on Climate Change with the ultimate goal of achieving a stabilized greenhouse gas concentration in the atmosphere, and participating countries agreed to address the global warming problem on a worldwide scale. On the basis of this Framework Convention, at the 3rd Session of the Conference of the Parties to the United Nations Framework Convention on Climate Change (COP3) held in Kyoto in 1997, the participating countries agreed to uphold the Kyoto Protocol, which specifies legally binding obligations for developed countries to reduce their greenhouse gas emissions over the period 2008-2012 (Japan, the US, and EU countries were to achieve 6, 7, and 8% reductions relative to those in

1990). Countries around the world have been taking major steps toward the reduction of greenhouse gas emissions. In 2015, the 21st Session of the Conference of the Parties to the United Nations Framework Convention on Climate Change (COP21) was held in Paris; a multinational and international agreement on climate change mitigation was adopted and concrete guidelines for countermeasures against global warming after 2020 were developed. These guidelines constrain us to make efforts to reduce greenhouse gas emissions. One article in the Paris Agreement specifies that the world should achieve a balance between anthropogenic emissions and removal in the second half of this century (in practical terms, zero emission) and pursue efforts to limit the increase in atmospheric temperature to <1.5 °C (currently, 0.9–1 °C) above preindustrial levels. Greenhouse gases refer to water vapor, CO₂, and methane in the atmosphere [4, 5] which serve as a heat insulator that prevents thermal radiation from the ground surface heated by solar energy from escaping into space. In recent years, emissions of chlorofluorocarbons, CO₂, and methane have rapidly multiplied as a result of human economic activities, and the concentration of greenhouse gases in the atmosphere has undeniably increased. In particular, CO₂ has a significant effect on global warming: the concentration of CO₂ over Antarctica, where CO₂ concentration is considered to be the lowest, has been reported to exceed 400 ppm [6]. The Kyoto Protocol and the Paris Agreement are international treaties to either reduce CO₂ emissions or not increase the amount of CO₂ in the atmosphere any further. What are the concrete guidelines for suppressing and reducing CO₂ emissions?

Background: Reduction of CO₂ emissions from electronic devices to realize a low-carbon society

A huge amount of greenhouse gas including CO₂ is emitted as a result of the consumption of electric energy to process materials or operate machines for processing during the manufacture of any product. The entire process from the procurement of materials, manufacturing, disposal, and recycling involves tasks that emit greenhouse gases. The carbon footprint of products (CFP) refers to a scheme to visualize the emissions of greenhouse gases by converting them into an equivalent amount of CO₂. For example, in the case of a light-emitting diode (LED) ceiling light (30×30 cm², four LEDs), the total CO₂ emission is approximately 2.15 kg-CO₂ as calculated from the sum of the power consumption required for the manufacturing of four LEDs, a diffuser, a reflector, and a light-guiding panel [3.9 kWh (the sum of power consumption required to manufacture one ceiling light) $\approx$ 0.004 kWh (LED) +

1.3 kWh (diffuser) + 1.3 kWh (reflector) + 1.3 kWh (light-guiding panel); CO₂ emission factor, 0.55 kg-CO₂/kWh;[ref] the diffuser, reflector, and light-guiding panel are assumed to be processed by sputtering and transfer]. When the amount of CO₂ emission due to turning on the ceiling light is included, the total amount of CO₂ emission of a ceiling light from manufacture to disposal is approximately 16 kg-CO₂. This is a rough estimate of the amount of CO₂ emission due to an ordinary industrial product. The total amount of CO₂ emission used for illumination all over Japan is ~100,000,000 t-CO₂ [7-10]. As is obvious from this figure, a huge amount of CO₂ is emitted just for the purpose of illumination.

The chemical formation of CO₂ is known obviously by



Carbon, an element in CO₂, exists everywhere in our environment on Earth. Recently, efforts have been made to suppress CO₂ emission by understanding and controlling the amount of carbon circulating in the environment, i.e., how carbon is cycled in the natural environment and finally emitted as CO₂.

Regarding CO₂ emitted as a result of human economic activities, the state in which the amount of emission is equal to that of removal is referred to as carbon neutral. Figure 1 shows an illustration of the carbon cycle as a whole, where CO₂ is removed through photosynthesis and is generated by forests, the ocean, and factories. Currently, carbon neutrality obtains only in limited cycle systems. When the amount of carbon cycled over the entire earth is determined macroscopically, the amount of CO₂ emission is much larger than the amount being removed. The phenomenon in which the amount of CO₂ emission is larger than the amount removed by photosynthesis is referred to as carbon negative. We have tried to directly or indirectly remove CO₂--the amount of which is calculated by subtracting the amount of removal from that of emission--through activities such as forestation, the preservation of forests, and clean energy projects (purchase of greenhouse gas emission rights) at sites apart from the emission sites. The Ministry of the Environment of Japan has launched a carbon offsetting scheme and built a support system for CO₂ emission reduction and removal projects to further promote the efforts of CO₂ emission reduction [11].

A society (assembly) that promotes the reduction of the amount of CO₂ emission to address the carbon-negative state is collectively called a low-carbon society. A low-carbon society refers to a social system with full environmental

considerations--such as the use of low-carbon energy sources--to reduce the emissions of greenhouse gases--such as CO_2 --to a level at which they can be removed by nature. As a method of introducing low-carbon energy, we have been searching for methods to realize a low-carbon society through the development of applications using carbon-based materials, in particular, carbon nanotubes (CNTs). The purpose of this study is to contribute to the reduction of carbon use and CO_2 emissions through active utilization of carbon-based electronic components, which contribute to a reduction in energy consumption.

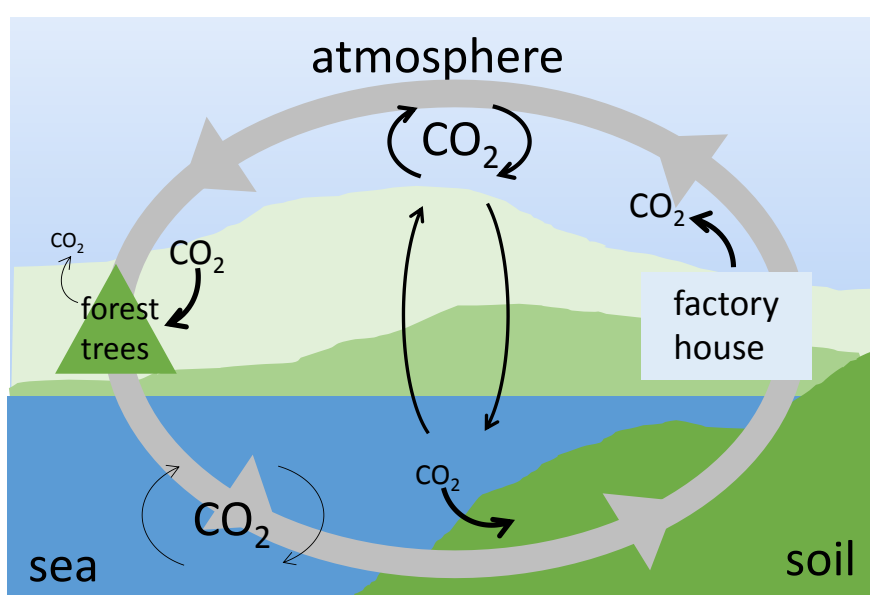


Fig. 1. Illustration of the carbon cycle.

REVIEW: DEVELOPMENT OF PLANAR LIGHTING DEVICES USING CNTS

We have been promoting research on and the development of energy-saving planar lighting devices using CNTs. Since CNTs were first reported by Iijima [12], applied research on one-dimensional nanoscale carbon-based materials such as CNTs and carbon nanofibers (CNFs) has been carried out. In particular, CNTs have a nanosized tubular shape along with physical properties such as high chemical stability, thermal conductivity, rigidity, and electrical conductivity that are considered to be effective for use in electronic devices [13-20].

However, CNTs have many crystal defects in the carbon network constituting the CNTs, regardless of the synthesization method, rendering it problematical to use them

in electronic devices requiring high reliability because of their unstable physicochemical properties. Therefore, we have been focusing on techniques for controlling the crystallinity of CNTs. Tohji et al. improved the purity and crystallinity of single-walled carbon nanotubes (SWCNTs) synthesized by arc discharge and succeeded in obtaining highly pure SWCNTs [21]. Jointly with Iwata and coworkers, he established a method of analyzing the crystallinity of highly crystalline SWCNTs with high resolution [22]. On the basis of these achievements, a process for controlling the crystallinity of CNTs has been established; however, no efficient handling technique for highly crystalline CNTs has yet been established. In addition to the development of a process to control crystallinity, applied research on field emission (FE) sources industrially using CNTs and CNFs has been actively carried out since 2000. Our research group has integrated these two achievements, carried out basic research to develop FE sources using highly crystalline SWCNTs [23], and succeeded in employing the SWCNT FE source as the cathode in a planar lighting device [24]. In addition, we found that highly crystalline SWCNT FE sources are effective in saving energy in FE, increasing the device lifetime, and improving the in-plane emission homogeneity of planar lighting devices [23].

As an example of such SWCNTs, Fig. 2 shows an overview of purified SWCNTs powder by arcing together with a solution with SWCNTs dispersed into an organic $\text{In}_2\text{O}_3\text{-SnO}_2$ precursor solution and a non-ionic surfactant. We were able to obtain a solution dispersing SWCNTs homogeneously and control the amount of SWCNTs into the solution from μg to mg per square-meter according to a thin film coated by a wet process. Fig. 3 shows transmission electron microscopy (TEM, HR-3000, Hitachi High-Technologies Corporation, Japan) images of SWCNT bundles dispersed with surfactant after annealing at 1400 K and 10^{-1} Pa.

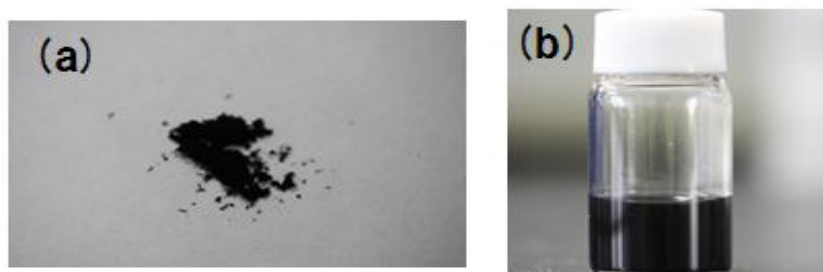


Fig. 2 (a) Overview of SWCNTs nano-powder.
(b) A solution with SWCNTs dispersed into ITO solution.

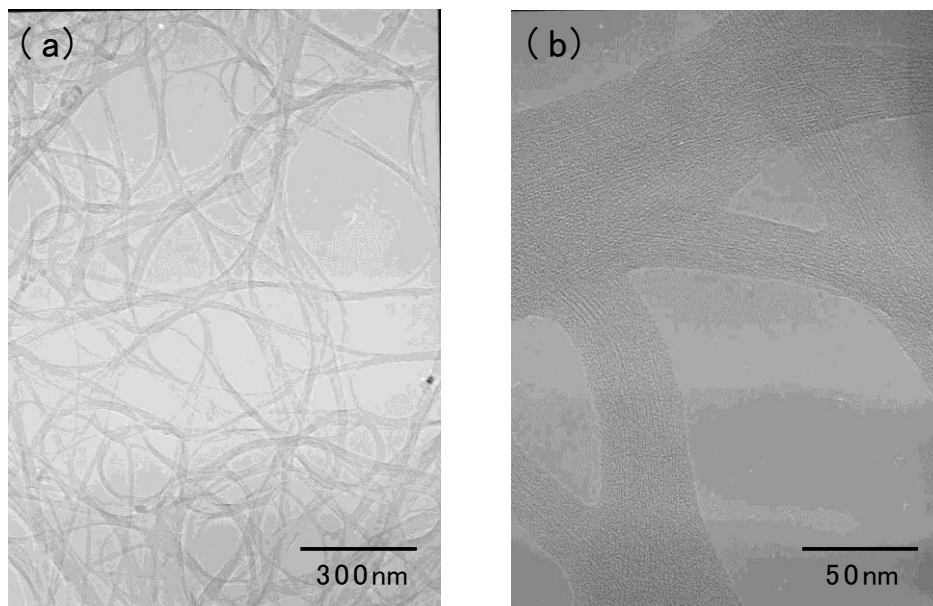


Fig. 3. TEM images of SWCNTs bundles after annealing dispersed into ITO solution. **(a)** Dispersed highly crystalline SWCNTs bundles. **(b)** Enlarged image of SWCNTs bundles covered with surfactants.

As shown in Fig. 3, each bundles were gathered with a few SWCNTs homogeneously. We developed a coating in which the annealed SWCNTs are uniformly dispersed by a wet process and then fabricated a planar thin film FE source using the coating. Figure 4 shows (a) a schematic of the planar FE source and (b) a photograph of planar light emission. Note that the homogeneity of the planar light emission depends on the amount of SWCNTs added. Nine points were selected from the light-emitting surface and the deviation of luminance at the nine points is evaluated in Fig. 5. Figure 6 shows the relationship between the deviation of luminance at each point normalized by the average luminance of the entire surface and brightness efficiency for the diode lighting device with the cathode employing dispersed SWCNTs against the amount of SWCNTs added.

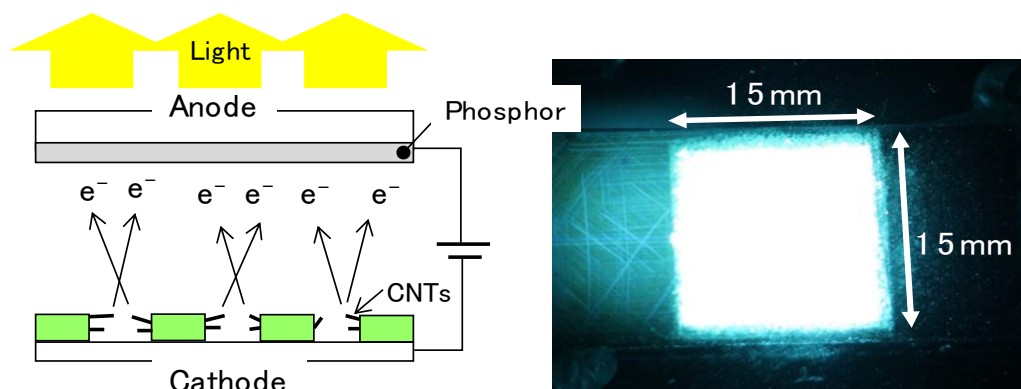


Fig. 4 (a) Schematic of planar FE source and (b) photograph of light-emitting surface.

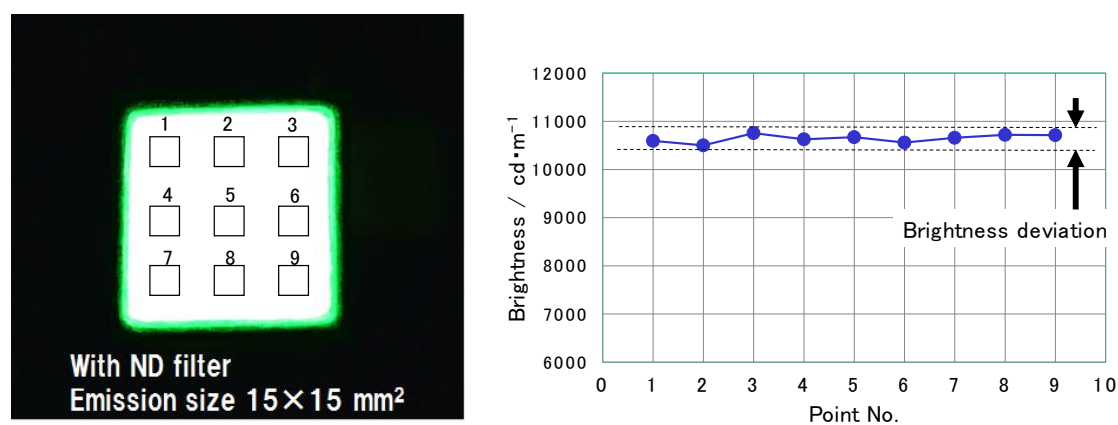


Fig. 5. Deviation of luminance at nine points on a lighting-emitting surface.

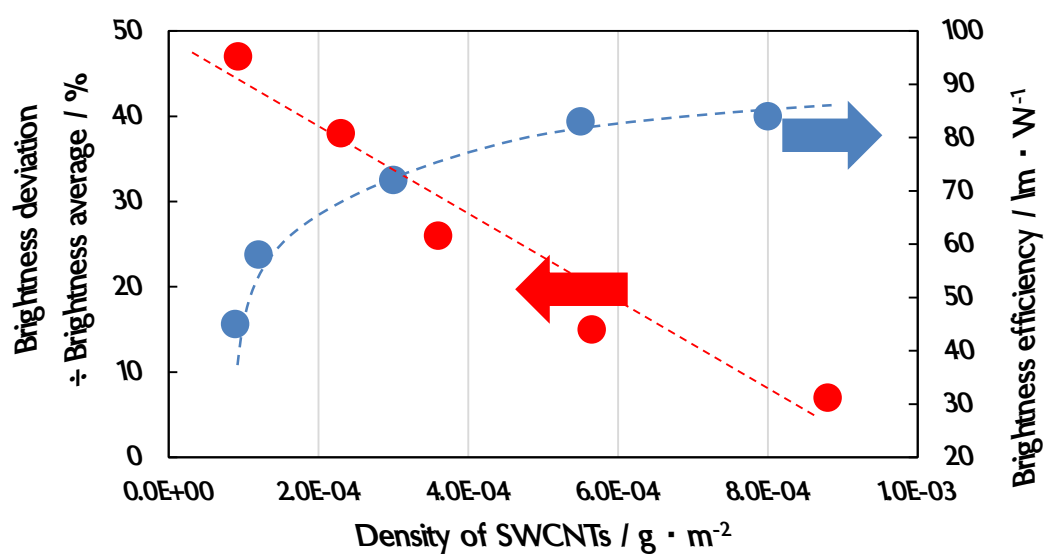


Fig. 6 Dependence of deviation in luminance on a surface density of SWCNTs.

The surface density on the abscissa in Fig. 6 represents the amount of highly crystalline SWCNTs added per unit area of coating. We have succeeded in obtaining a planar thin film with homogenous light emission by adding SWCNTs at a density of 200–300 $\mu\text{g}/\text{m}^2$. We have also reached an energy conversion efficiency of ~ 80 lm/W (in luminance) for a planar lighting device using this thin film, although details are not included here [25]. Using highly crystalline SWCNTs, the amount of carbon (or SWCNTs) used in the illumination devices can be significantly reduced; at the same time, energy consumption by these devices can be successfully lowered. This is equivalent to reductions in both CO_2 emission and the amount of carbon used, showing the possibility of realizing a low-carbon society by suppressing the amount of carbon entering the cycle.

Next, the amount of CO_2 emitted from various processes associated with lighting devices shall be examined. The amounts of CO_2 emitted during the fabrication process per lumen of an LED [26] and our planar lighting device per year are shown in Fig. 7. The vertical axis was calculated by the following formula [27],

$$\begin{aligned} & (\text{Power consumption for process of a planar panel (kWh)} \times 0.55 \text{ (kg-}\text{CO}_2\text{/kWh)} + \\ & \text{organic additive in a planar panel (mg)}) / (\text{Brightness output of a planar panel (cd/m}^2\text{)} \\ & \times \text{A planar panel size (m}^2\text{)} \times 3.14) \end{aligned} \quad (2)$$

We developed a planar FE source by spraying the coating in which the SWCNTs were dispersed by a wet process. Without a high-vacuum, electron emission by a FE planar lighting device with SWCNTs cannot be realized. Therefore, the data in Fig. 7 include the amount of CO_2 emitted during the fabrication of vacuum valves. Power consumption when fabricating a planar panel of 6 inch spent an average of 0.00048 kWh in our manufacturing laboratory in 2016; we were able to obtain a high brightness of 14,000 cd/m^2 with power consumption of 4.2 W for lighting. This finding indicates that a marked reduction in the amount of CO_2 emitted from device processes can be realized by optimizing the amount of SWCNTs added and improving the efficiency of the luminance.

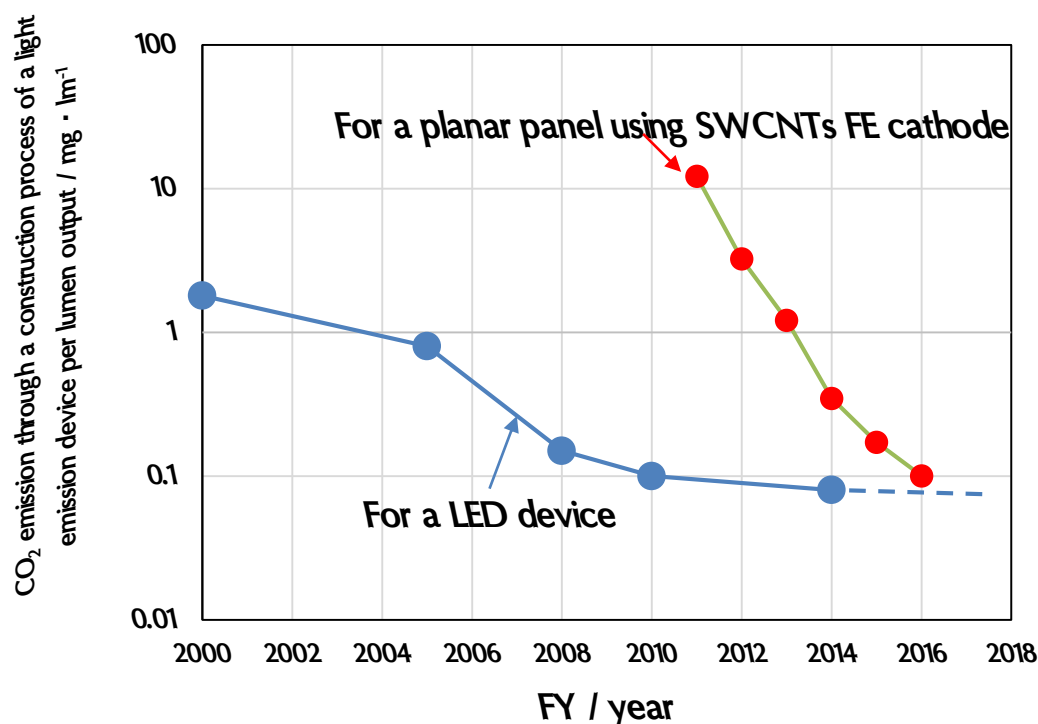


Fig. 7. Amount of CO₂ emitted during fabrication processes per lumen of LED and planar lighting device.

Smalley et al. focused on the advantages of carbon-based materials with high physicochemical properties (e.g., CNTs, CNFs) and predicted that these carbon-based materials will promote the development of a low-carbon society [28]. Effective use of highly crystalline SWCNTs will lead to controlling the amount of carbon used in devices and a reduction in CO₂ emissions through energy-saving light emission. In the near future, we anticipate development of a technology that can realize a change from a carbon-neutral state, in which the amount of CO₂ emission is equal to that of removal, to a carbon-positive state, in which the amount of removal exceeds that of emission. The carbon-based materials used in electronic components, including the FE source mentioned herein, are expected to contribute to reductions in the amount of carbon cycled and energy consumption (i.e., reduction in the amount of CO₂ emission).

CONCLUSIONS

The amount of global CO₂ emission continues to increase. If this trend progresses into the future, it will be very difficult to suppress the concentration of CO₂ over

Antarctica to below 400 ppm in the next 20–30 years. To prevent global warming through the stabilization of the concentration of CO₂ at a low level, a low-carbon society or carbon-positive state should be realized. We are responsible for contributing to global environmental conservation through the suppression of carbon use and energy consumption through the development of new electronic devices. All the electric equipment and electronic devices that support our lives consist of modular solid-state devices. A huge amount of energy is consumed for each solid-state device, from production to implementation to operation. To reduce all related energy from development to use in our daily lives and to develop a low-carbon society, we have been studying the development of devices that effectively use carbon-based nanomaterials. In this paper, we explained the methods of reducing the energy consumed in industrial processes and running costs. We also presented some of the process and device development technologies that maximally utilize CNTs. It is our hope that these technologies will promote the realization of a low-carbon society.

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