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# Nuclear fuel materials and its sustainability for low carbon energy system: A review

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**Abstract.** World energy generation for electricity is still dependent on fossil fuels since it is more reliable and secure than the current intermittent renewable energy systems. Although the integration of renewable energy as an energy mix is in progress, still it could not be able to replace fossil fuels. Dependency on fossil fuels will not only contribute to severe climate change but will also degrade future generation quality of life. Hence, the solution to quandary is by integrating nuclear power plants with those of renewable energy such as solar and wind to meet the energy demand and to ensure sustainability of energy source. The current operating nuclear power plants in the world use the concept of water-cooled reactors. It was designed so that the fast neutrons born from fission reactions were slowed down in the moderator to allow other fission reactions events in sustainable chain reactions. Besides, the slow neutrons with low energy is a favourable reactor feature for safe and efficient operation. The common types of nuclear fuel materials in water-cooled reactors are enriched uranium dioxide and natural uranium contained in nuclear fuel elements. After it has been used, the fuel elements will be stored as spent fuel. Prolonged storage of used nuclear fuels will make the volume of nuclear waste high and become hard to manage after a long period of storage. An effort to reprocess the spent fuel as to extract fissile and fertile material to be used in nuclear fuels usually was undertaken to reduce the waste volume. However, this process may lead to an undesirable proliferation of nuclear material. In this review article, research on the advancement of nuclear fuel materials will be discussed based on the reduction method of the nuclear spent fuel volume and radiotoxicity, as well as to study its sustainability for the future low carbon energy system.

## 1. Introduction

Climate change has pushed the world to harness a low carbon energy system. In 2019, 84.3% of the global energy consumption was produced by fossil fuels, 4.3% from nuclear and the rest of it from various renewable energy sources [1]. According to Our World in Data (2021), the trend of carbon dioxide (CO<sub>2</sub>) emission was increasing since 1900 but reported a slow declining trend in 2019 due to the inclusion of 11.40% of the renewable energy in the energy-mix system. However, the Earth's global surface temperature as stated in the Global Climate Summary is increasing up in July 2021 to 0.93°C above the average of 15.8°C where this is the highest temperature recorded [2]. This will lead to catastrophic climate issues if the world doesn't act now and depending too much on fossil fuel as the major energy production and consumption. The IPCC Report (2021) stated that climate change has been affecting every region on Earth in multiple ways and the changes will increase with additional warming



[3]. For instance, the highest temperature recorded this year has led to the largest wildfires in California, Colorado and the “black summer” of fires in eastern Australia [4]. Hence, immediate implementation of the low carbon energy system is necessary. This can be done by shifting the energy production from fossil fuels that releases an enormous amount of the CO<sub>2</sub> and other greenhouse gases to the low carbon and sustainable energy system to meet the needs of current generation without compromising the needs of the future generation.

In 2015, the treaty on climate change which is The Paris Agreement has been adopted internationally and, almost 200 countries have pledged to reach the main goal such that “to limit the global warming to well below 2°C, preferably to 1.5°C compared to pre-industrial level” [5][6]. In addition to that, the 2030 Agenda for Sustainable Development was introduced in the same year and focused on providing 17 Sustainable Development Goals (SDGs) plan of action for humanity, planet, and prosperity [7]. One of the SDGs is to develop affordable and clean energy (SDGs 7). According to IAEA, by harnessing nuclear energy in the future sustainable energy mix, SDGs 7 could be achieved and would help to reach another 16 goals indirectly as shown in Figure 1 [8].

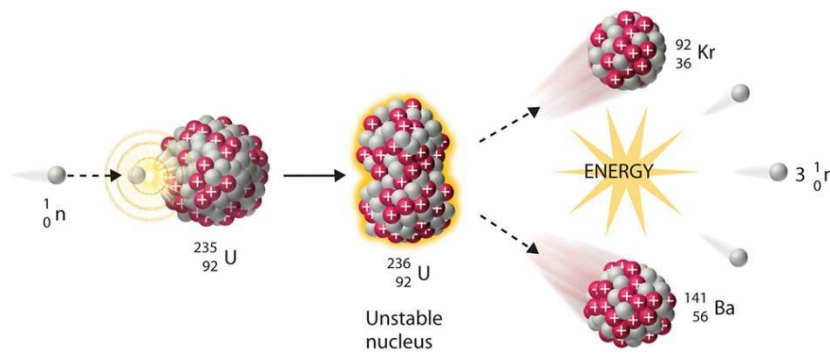


**Figure 1.** Nuclear energy is the key to the rest of SDGs [22].

In this review article, the focus area for the low carbon energy system discussed will be concentrating on the nuclear energy, specifically in the nuclear fuel materials subjected to its sustainability. The implementation of nuclear energy could help in reducing the CO<sub>2</sub> emission and greenhouse gases. However, issues in nuclear waste management is among the concern factors that limit users from harnessing the nuclear energy, due to the long-lived radioactive waste and lack of public acceptance [9]. Thus, this paper aims to review and discuss the fuel materials used in the conventional and advanced reactor technology related to its waste as well as radiotoxicity management, which can be elaborated in the following sections.

## 2. Nuclear fuel materials

Nuclear energy is generated via a fission process. In general, when a neutron is bombarded onto a heavy fissile nucleus, it will split into two daughter nuclei and release a huge amount of kinetic energy together with 2-3 fission neutrons as depicted in Figure 2. The fission process can be sustained by allowing another fission process to occur when the fission neutrons meet with another heavy fissile nuclei. This scenario is known as chain fission reaction and is used in current operating nuclear fission reactors to produce clean energy. The common types of heavy fissile nuclei for generating nuclear fission are Uranium (U) atom, Plutonium (Pu) atom and Thorium (Th) atom. These atoms can be divided into two isotope categories which are fissile isotopes and fertile isotopes as the fuel materials in the nuclear reactors. Fissile isotopes such as  $^{235}\text{U}$ ,  $^{233}\text{U}$ ,  $^{239}\text{Pu}$  and  $^{241}\text{Pu}$  required the thermal neutron to create the fission process. While  $^{238}\text{U}$  and  $^{232}\text{Th}$  are the example of fertile isotopes that required the fast neutron to convert the nucleus into fissile isotopes ( $^{239}\text{Pu}$  and  $^{233}\text{U}$ ) for fission process.



**Figure 2.** Fission reaction [10].

According to IAEA, the number of light water reactors (LWRs) in operation in the world, as on 31 Dec 2019, are 413 reactors, with 52 units are under construction while 73 reactors are planned to be implemented in future [11]. The current operating LWRs utilize the conventional enriched uranium oxide ( $\text{UO}_2$ ) as the nuclear fuel materials. The composition of fissile materials in  $\text{UO}_2$  is less than 5% which is commonly known as low enriched uranium (LEU). Besides LEU, mixed oxide (MOX) fuels were also used as the fuel materials in LWRs. The composition of MOX consists of 94-97% of  $\text{UO}_2$  blended with 3-5% of the  $\text{PuO}_2$  that has been extracted via reprocessing from the used nuclear fuels [12]. The extension on the reprocessing methods can be obtained in the next section.

The advancement of nuclear fuel materials has been conducted extensively to develop future nuclear energy with sustainable concepts. The Fukushima accidents has led to the improvement of the fuel materials in LWRs for advanced reactor applications via Accident Tolerance Fuels (ATF). The ATF should be designed to have the capability of performing better during normal operation as well as responding well when loss of coolant accident and reactivity-initiated accident to prevent reactor from meltdown as had happened in Fukushima plant [13]. For instance, fully ceramic microencapsulated fuel (FCM) with tri-structural isotropic (TRISO) particles, uranium carbide (UC), uranium nitride (UN), uranium dicarbide ( $\text{UC}_2$ ) are among the potential candidates of the advanced nuclear fuel materials for advanced reactor technology besides LWRs types [14][15]. In addition, more researchers are also interested in investigating the higher composition of uranium enrichment between 5% to 19.75% called high-assay low enriched uranium (HALEU) as the advanced nuclear fuel materials. A study done by [16] suggested that the reactor performances and safety analysis of LWRs when using HALEU composition in the fuel concluded that no significant neutronic or reactor safety interruption could be observed, however further exploring the potential of HALEU in LWRs was recommended. Besides, the study also claimed that the waste volume per unit energy generated can be reduced but required

additional natural resources due to higher enrichment of the  $^{235}\text{U}$  isotopes. HALEU also may be needed in future advanced reactor technology for improving the fuel utilization and helps in plant economics [17]. Furthermore, the advancement of nuclear reactor technology such as long-term reactor operations, advances nuclear fuel materials, improvement on the reactor design as well as minimizing the waste volume and cost-effectiveness makes the nuclear energy are more feasible and sustainable to the environment, economic and social [18].

### 3. Nuclear spent fuel volume

Besides the radiation hazard and public acceptance issues, the volume of the used nuclear fuel and waste produced in the nuclear power plant causing this type of low carbon energy system is not reliable for sustainability. In addition, the long-lived radioactive and the high heat load in the spent fuels contributes to the development of an intensive nuclear waste management, deep geological repository and poses a great danger to humankind and the environment. Current operating nuclear reactors are of the water-cooled reactors, commonly known as LWR. The spent fuel produced from these reactors consists of major actinides, minor actinides, and short-lived fission products. Approximately 0.4% of these cumulative inventories are classified as high-level waste (HLW) with half-life more than  $2.0 \times 10^5$  years [19]. According to [20], around 150 tonnes of the HLW and fission products were stored permanently in the underground nuclear waste facilities worldwide in 2006. And the trend of the spent fuel being unloaded from the reactors will increase by 11,500 tHM in the year of 2010 annually as depicted in Figure 3. Thus, the volume of the HLW waste also will keep on increasing and eventually, making nuclear energy unsustainable, although this energy is clean and produces zero  $\text{CO}_2$  emission during energy generation.

Hence, to overcome this issues, various studies have been conducted with the aim to reduce the waste volume by extracting and reprocessing the useful actinides from the spent fuels for the peaceful use rather than treating it as HLW directly and disposing it permanently. The extraction of useful actinides such as Pu could lead to the proliferation threat. Due to this issue, in 1953, the Atoms for Peace program for regulating the uses of nuclear technology for peaceful purposes was announced [21]. In 1968, the Treaty on the Non-Proliferation of Nuclear Weapons (NPT) was introduced to all countries with its objective to prevent the spread of nuclear weapons and weapons technology as well as to promote the cooperation in the nuclear energy for peaceful uses [22]. The efforts made by almost 191 States that joined to follow the NPT has promoted the advancement of nuclear technology to emerge with other's fields without worrying about the proliferation threat.

As stated in IAEA report, the cumulative of the spent fuel generated around the world in the beginning of 2004 was 268 000 tHM, where 90 000 tHM had been reprocessed [23]. Based on Figure 3, by conducting the reprocessing methods, the amount of the spent fuels that needed to be stored will decrease annually and helps this clean energy production to be among the sustainable energy for achieving the sustainable development goals (SDGs).

The current strategy to reduce the waste volume can be done by partitioning and transmutation (P&T) process. This process could help in reducing the waste volume for peaceful uses. For instance, the process works in two parts. The first part is partitioning where it is carried out by separating the actinides, including Pu and U in the spent fuels using the pyrochemical technologies to produce the new nuclear fuel from the waste.

Next, the separated actinides will be burned and transmuted into the short-lived radioactive materials in the transmutation process where it can be carried out in any types of nuclear reactor theoretically, but preferable to be conducted in fast breeder reactors (FBR) and accelerator driven system (ADS) [24-26]. The pyrochemical technologies has been conducted extensively in many studies worldwide as reported by OECD in their status report [27]. In addition, the study conducted by [28] to investigate the significant capability of P&T methods for decreasing the waste volume was proven where the number of vitrified waste packages can be reduced to 1/4 which also lessens the waste facility footprint to 1/100 effectively. In [29] work, the comparison of waste produces with the P&T and non-P&T methods showed substantial

waste package and footprint reduction by 54% incorporating the proposed ‘high-waste-loading glass and Cold Crucible Induction Melter (CCIM)’ waste case as stated in the study. Since this paper is only focusing on reviewing the suitability of nuclear fuel materials for sustainability, thus the details on the technical parts of the P&T and the pyrochemical technologies will not be discussed.

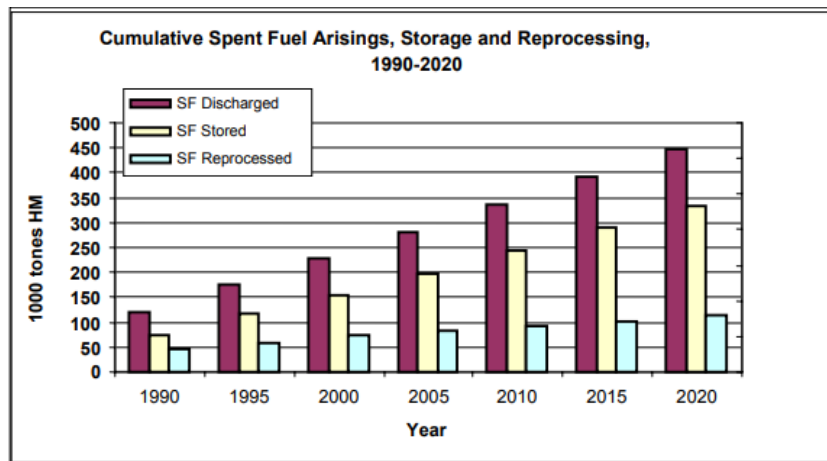


Figure 3. Total amount of spent fuels to be stored, discharged, and reprocessed [20].

#### 4. Radiotoxicity

The parameter to determine the toxicity released by the nuclear spent fuel is known as radiotoxicity. This parameter is an essential information required in managing the waste disposal facilities either in temporary storage facilities or geological repositories for long-term disposal. It is well noted that radiotoxicity is the result of the decay actinides and fission products which mainly depends on the fuel materials such as  $UO_2$ , MOX or Th fuel. Figure 4 shows the trend of the radiotoxicity over the years after reactor's shutdowns based on U and Th fuels. For the radioactivity during and after the reactor operation, in comparison with radioactivity of the spent fuel during its subsequent handling, several things must be taken into consideration. Even during the operation, it is very difficult to determine the exact number of different radionuclides within the fuel. This is because many radionuclides have very short lives, with behaviour that instantly disappears after the reactor shutdown.

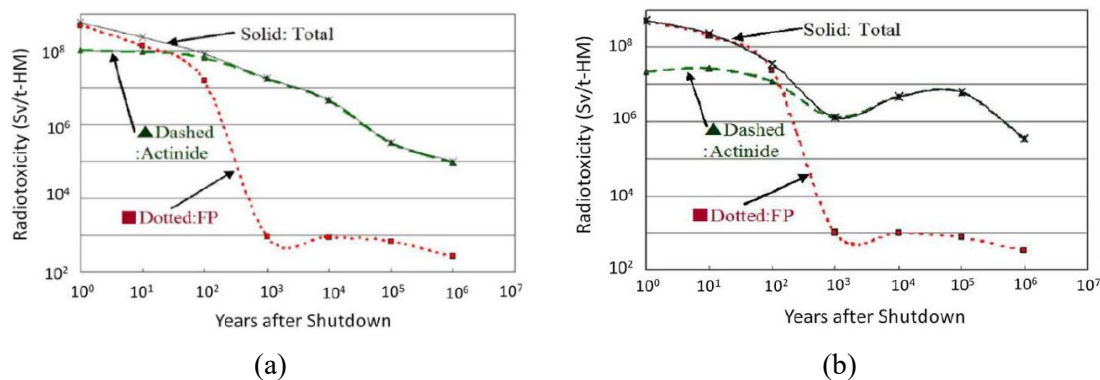


Figure 4. Component of radiotoxicity for the (a) U fuels and (b) Th fuels [30].

Radiotoxicity is mainly influenced by short-lived nuclides, including iodine. Majority very short half-lives that only contribute to a small percentage of the overall radiotoxicity after one month. Some comparison studies have been conducted to investigate the radiotoxicity among the fuel material types. For example, reactors with Th fuels produce less Pu than the U fuels which result in lesser radiotoxicity, but a daughter nuclide of  $^{233}\text{U}$  in the Th reactors causes higher radiotoxicity during long-term storage than the U core [30]. For the following Pressurized Water Reactor (PWR) cores, the ORIGEN2 code [31] was used to estimate the amount of radiotoxicity. For fuels irradiated to 45 GWd/t, a constant rated power density of 39.2 MW/t was used, and radiotoxicity was analysed for time periods up to 1 million years.

- U-PWR-core: 4%  $^{235}\text{U}$  + 96%  $^{238}\text{U}$ ,  $^{220}\text{U}$ -Library.
- Th-PWR-core: 4%  $^{233}\text{U}$  + 96%  $^{232}\text{Th}$ ,  $^{214}\text{Th}$ -Library.

Equation (1) was used to translate the radioactivity calculated by ORIGEN2 to radiotoxicity. Note that ORIGEN2 gives the radioactivity of a tonne of HM (initial Heavy Metal) in 'Curie' units, which must be converted to 'Becquerel' units.

$$\text{Radiotoxicity [Bq/t - HM]} = \text{Radioactivity [Bq/t - HM]} \times \text{DCF [Sv/Bq]} \quad (1)$$

When a nuclide is injected into a human body, it generates internal radiation exposure, which is measured by radiotoxicity. It is determined by the type of radiation, its energy, and its influence on the human body, which is described as a dose conversion factor (DCF). The majority of DCFs may be found in numerous references, such as [32] and [33]. DCFs for several other nuclides can also be found in [34] and [35], or authors can make assumptions for them. A comparison of radiotoxicities calculated for the U and Th cores in PWR is shown in Figure 5. Up to 1000 years, the Th core has a 10–90 percent lower radiotoxicity than the U core; beyond that, the tendency reverses. Around 0.1 million years, there is a peak in radiotoxicity. The decay heat curves are found to be similar to these curves. Table 1 shows the numerical values of the data in Figure 5.

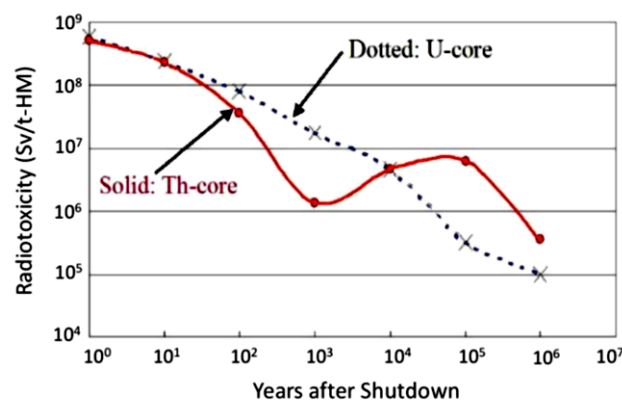


Figure 5. Radiotoxicity for U core and Th core.

**Table 1.** Radiotoxicity for U core and Th core (Sv/t-HM).

<i>Year</i>	<i>1</i>	<i>10</i>	<i>100</i>	<i>1.00E+03</i>	<i>1.00E+04</i>	<i>1.00E+05</i>	<i>1.00E+06</i>
U Core	5.88E+08	2.36E+08	8.18E+07	1.77E+07	4.59E+06	3.30E+05	9.80E+04
Th Core	5.18E+08	2.32E+08	3.57E+07	1.32E+06	4.65E+06	6.16E+06	3.51E+05
%	-12.0	-1.9	-56.4	-92.5	1.3	1766.9	258.5

$^{239}\text{Pu}$ , which contributes about half of the overall radiotoxicity in the U core during long-term storage (e.g., 10 000 to 100 000 years), is the most important contributor of radiotoxicity during long-term storage (e.g., 10 000 to 100 000 years).  $^{229}\text{Th}$  is the largest contributor of radiotoxicity in the Th core during long-term storage (e.g., from 10,000 to 100,000 years), accounting for over half of the total radiotoxicity.  $^{233}\text{U}$  has a daughter nuclide,  $^{229}\text{Th}$  (half-life: 159 200 year). Thus, the Th core produces less Pu than the U core, resulting in lesser radiotoxicity from Pu, but a daughter nuclide of  $^{233}\text{U}$  in the Th core causes higher radiotoxicity during long-term storage than the U core.

In [36] stated that the cumulative radiotoxicity of the actinides in the U-Pu fuels and Th-U fuels were 2.5 times higher and 3.5 times lower than the U fuels respectively. Effort has been made to reduce the toxicity of the spent fuel which can be carried out by recycling the actinides and fission products in the spent fuel to decrease the nuclear waste volume via P&T process. The separation and transmutation of the minor actinides may contribute to shortening the long-term decay heat and radiotoxicity inventories. However, in [28] works claimed that only  $^{90}\text{Sr}$  and  $^{137}\text{Cs}$  of the minor actinides produced significant depletion on the radiotoxicity using P&T process in the deuteron accelerator. In [37] stated that the transmutation of Pu by irradiating all the MOX fuels in current operating LWRs in the next 50 years would require a long period of time for radiotoxicity reduction.

Chemical separation of Plutonium, Americium, and Curium before long-term controlled storage is recommended for Uranium and Plutonium spent fuels. For significant conversion of  $^{241}\text{Pu}$  into  $^{241}\text{Am}$ , Americium should be separated after 50-70 years of storage.  $^{244}\text{Cm}$  decays almost completely after 100 years. Extracted americium should be utilised for transmutation and plutonium should be reused. Separation of actinides is also effective in lowering the heat of decay.

$^{232}\text{U}$  determines the vast majority of radiotoxicity in Thorium spent fuel. It is self-evident that repeated usage of Thorium fuel will result in an increase in radiotoxicity. Additional deep burn-out (transmutation) of Uranium fraction comprising both  $^{233}\text{U}$  and  $^{232}\text{U}$  is required for one-fold utilization of thorium fuel with deep  $^{233}\text{U}$  burnup. The extraction and transmutation of Plutonium fractions can reduce radiotoxicity by several orders of magnitude ( $^{238}\text{Pu}$ ). Because  $^{228}\text{Th}$  decays virtually completely in 10 years with its short-lived daughter nuclides, transmutation of  $^{228}\text{Th}$  - daughter nuclide of  $^{232}\text{U}$  is not required.

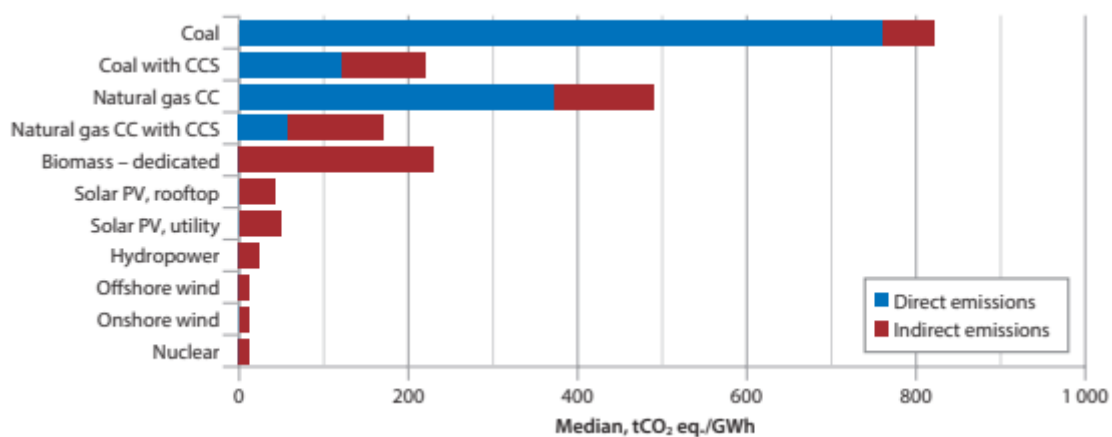
## 5. Discussion

From the literature review, the nuclear fuel materials improvement in advancing the reactor technology to make it more feasible and sustainable has been conducted extensively. Align with the Paris Agreement and the 2030 Agenda on Sustainable Development, the application of nuclear energy could help in reducing the  $\text{CO}_2$  and greenhouse gases emission. The current contribution of nuclear energy for electricity generation has been reported to avoid nearly 20% of the global  $\text{CO}_2$  emission from 1971-2018 [38]. Whereas the greenhouse gases emissions from nuclear energy are among the lowest compared to other energy sources as depicted in Figure 6 [39].

However, to fully utilize nuclear energy by replacing the current fossil fuel-fired power plant is not feasible as it still has the risk potential from the waste and large releases of radioactive materials. The improvement in fossil fuel-fired power plant also has been made to reduce the  $\text{CO}_2$  emission and greenhouse gases via Carbon Capture and Storage (CCS). In [40] report, the usage of CCS has captured



almost 40 Mt of CO<sub>2</sub> in 2020 and expected to capture more in 2050 around 5,635 Mtpa if the application of CCS is speed up globally. In addition, the net installed renewable energy is expected to grow. As reported in [41], the addition of wind and hydropower are increasing drastically, accounting for almost 90% of the total global power capacity, while the application of the solar photovoltaic remain stable and on demand. Hence, the authority especially the policymakers need to come out with the solution on the future energy mix strategies by including all these three types of energy generation from nuclear energy (advanced reactor technology), fossil fuel-fired power plant with CCS and renewable energy (wind, hydropower, solar) to solve the climate issues as well as saving the environmental and human from the consequences of insecure energy source. In addition, by installing the CCS facilities powered by nuclear energy for capturing the CO<sub>2</sub> also could resolve the global warming issues to achieve sustainable development.



**Figure 6.** Greenhouse gases emissions comparison [35].

## 6. Conclusion

This paper outlined the advantages of having nuclear energy as the key to overcome climate change issues by focusing on sustainability aspects of the nuclear fuel materials. The conventional fuel types for LWRs and advanced fuel materials were discussed as well as the P&T and the pyrochemical technologies in reducing the radiotoxicity, waste volume and heat load of spent fuels. It can be concluded that by having the advanced nuclear fuel materials with advanced reactor technology in the energy mix strategies can help in reducing the CO<sub>2</sub> gases emission and greenhouse effects and, providing the world with effective solutions for energy security besides depending on the fossil-fuel fired power plants for continuous energy production.

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