Recent progress in diamond radiation detectors

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ABSTRACT
This paper reviews recent progress in diamond radiation detectors. Diamond is an ultra-wide gap (5.5 eV) semiconducting material which has several ideal properties for radiation detectors, such as solar blindness, high temperature operation, and fast response. Furthermore, diamond has near tissue-equivalence due to its low atomic number ($Z = 6$) and chemical stability due to its strong covalent bonds. Because of these features, diamond has long been used as a radiation detector in the fields of nuclear engineering, nuclear fusion, high energy physics and medical therapy. Until the 1990s, most of the research was conducted using selected high purity natural diamonds. Since the 2000s, the detector characteristics of synthetic diamond detectors have been greatly improved by achieving high purity diamond by microwave plasma enhanced chemical vapor deposition (CVD). Single-crystal CVD diamonds present best characteristics for spectroscopy in diamond radiation detectors. For applications requiring large sensitive areas, polycrystalline CVD diamond is mostly used. Heteroepitaxial diamond detectors are a promising alternative to increase the area of spectroscopic diamond radiation detectors. For applications in extreme environments, high radiation flux which leads to polarization effects is a crucial issue. Even with diamond, which has excellent radiation hardness, degradation of detector characteristics due to irradiation is inevitable. Detectors designed with small carrier travel distances, such as membrane diamond detectors and three-dimensional diamond radiation detectors, are effective ways to mitigate the degradation.

1. Introduction

Diamond radiation detectors have excellent properties, such as high temperature operation [1,2] and solar blindness due to a wide band gap of 5.5 eV, fast response due to high electron and hole mobility and saturation drift velocity [3], radiation hardness and tissue equivalence [4] due to low atomic number ($Z = 6$) and large displacement energy ($E_d = 34$ eV). Because of these excellent characteristics, they have been widely used in the fields of high-energy physics [5], nuclear engineering, nuclear fusion [6], and medical physics [4].

Diamond was originally shown to be sensitive to radiation in the 1940’s [7–9]. In the 1970’s, selected high quality natural diamonds were used for neutron measurements in the field of nuclear fusion, etc [10]. At that time, screening by UV light irradiation was performed to select the high purity type IIa natural diamonds. Since the 1980’s, the success of chemical vapor deposition has greatly expanded the applications of synthetic diamond [11]. Initially, polycrystalline diamond on Si substrates were used for radiation detectors in 1990s [12], followed in the 2000s by reports from several research groups on the growth of high-purity single-crystalline diamond by microwave plasma CVD, which greatly improved the spectroscopic properties of diamond radiation detectors [13,14].

This paper reviews the recent progress in diamond radiation detectors. Firstly, the operation principle of diamond radiation detectors and the parameters which describe the detector characteristics are introduced. Then, different types of diamond radiation detectors and their characteristics are summarized. Finally, examples of applications are presented. In recent years, the demand for harsh environment electronics has been increasing due to the decommissioning of nuclear power plants and the increasing beam intensity and flux in high energy experiments. Based on this background, this paper discusses the environmental resistance of diamond, especially its high temperature operation and radiation hardness. Applications of detectors and related technologies, such as diamond FETs and diamond nuclear batteries, are also presented.
2. Physical parameters to determine detector properties

In this section, we will introduce the characteristics of diamond radiation detectors and the parameters which determine their performance. Diamond radiation detectors have a structure consisting of high-purity diamond sandwiched between two electrodes. This structure is called MSM (Metal-Semiconductor-Metal) or MIM (Metal-Insulator-Metal) structure. In the field of radiation measurements, it is also called a solid-state ionization chamber because it operates on the same principle as gas ionization chambers. Figure 1 shows the schematic of the detector. When a charged particle is injected into the detector, it generates electron-hole pairs. The generated charge carriers move along the electric field. This movement of electrons/holes induces current following Shockley-Ramo theorem [15]. By detecting this induced current/charge, incidence of radiation can be obtained. The timing of the arrival of the radiation can be determined by the current pulse, and the energy of the radiation can be determined from the amount of charge. Since the amount of charge is linear to that of incident energy in the detector and energy spectrum is unique to nuclide, energy spectroscopy is a powerful tool to identify nuclide. The following is a list of performance specifications for diamond radiation detectors.

2.1. Charge collection efficiency (CCE)

CCE is one of the performance indicators of semiconductor radiation detectors for spectroscopy applications. It is defined as the amount of collected charge relative to the amount of generated charge inside the detector induced by ionization.

\[ \text{CCE} = \frac{Q_{\text{collected}}}{Q_{\text{induced}}} \times 100(\%) \quad (1) \]

The amount of induced charge \( Q_{\text{induced}} \) is defined by the following equation

\[ Q_{\text{induced}} = qN = \frac{qE_{\text{deposit}}}{\epsilon} \quad (2) \]

where \( N \) is the number of charge carriers and \( \epsilon \) is the average energy required to produce an e-h pair. \( \epsilon \) increases with the increase of the band gap. In general, it is known to be about three times of the band gap by Klein’s equation [16]. It is in good agreement with narrow bandgap semiconductor materials. In wide bandgap materials, experimentally obtained values are often lower than the values derived by Klein’s equation. For diamond, experimentally obtained values of around 13 eV are commonly used [10,17,18].

With inadequate CCE, charge trapping in detectors causes a polarization effect, where the electric field inside the detector causes a field that cancels the applied field. This results in an electric field that cancels out the applied field by bias voltage, thus degrading the CCE, energy resolution and counting capability of the detector with operation time. Therefore, for the spectroscopy application at high counting rate, it is desirable that the amount of charge trapping in the detector is as low as possible.

2.2. Energy resolution

Energy resolution is an important performance parameter for spectroscopy. It is defined as the full width at half maximum relative to the energy to be detected. For spectroscopy, high quality single crystal diamond is required. Figure 2 shows examples of spectra obtained with single crystal diamond and polycrystalline diamond [19]. It is difficult to obtain energy spectrum using polycrystalline diamond due to the charge trapping at grain boundaries. For this reason, polycrystalline diamond radiation is used for timing and position detection. Figure 3 shows the energy spectrum using high quality single crystal diamonds. Excellent energy resolution
of about 0.3% is obtained for alpha-particles from $^{241}$Am [17]. The theoretical limit of the energy resolution is determined by the number of charge carriers produced in the detector. The theoretical limit is expressed by the following equation.

$$\Delta E / E = 2.355 \sqrt{F / N} = 2.355 \sqrt{F / E_{\text{dep}}}$$

(3)

The energy distribution measured by spectroscopy is approximated by a Gaussian function, and the half-width is determined by the number of carriers produced in the detector. In other words, it is determined by the $\epsilon$ value and the energy given by the incident radiation. The energy resolution of wide bandgap materials is not as high as that of Si and Ge detectors, which have smaller $\epsilon$ values because diamond radiation detectors produce a smaller number of carriers by ionization.

Compared with the theory, high quality single crystal CVD diamond radiation detector presents ideal energy resolution under irradiation of monoenergetic $\alpha$-particles from $^{241}$Am under low counting rate. Here, $F$ in the equation is called the Fano factor and is expressed as a constant smaller than 1. A value of $F = 0.4$ was obtained for diamond [17]. These results indicate that the detector characteristics of high quality single crystal CVD diamond radiation detectors have achieved nearly ideal properties as semiconductor radiation detectors.

2.3. Schwalts (mean free path), CCD

The CCE mentioned in section A depends on the film thickness. The distance until the CCE reaches $1/e$ is called Schwalts or mean free paths (MFP). The MFP is described as follows

$$\lambda = \mu \tau E$$

(4)

where, the carrier lifetime is $\tau$, the mobility is $\mu$, and the electric field strength is $E$. MFPs of hundreds of $\mu$m to $\sim$ mm have been reported for high purity single crystal CVD diamonds.

CCD (Charge collection distance) is the CCE multiplied by the detector thickness $w$. It is often used as a performance indicator for polycrystalline diamond detectors. The values range from several tens of micrometers to several hundred micrometers [20].

$$CCD = CCE \times w$$

(5)

2.4. $\mu$τ(mobility- lifetime) products

The CCE depends on the applied voltage (electric field strength) and film thickness. This is because the distance that charge carriers can drift in the semiconductor increases as the electric field strength increases. The field strength dependence of CCE is described by Hecht's equation[21], which is widely used in the characterization of detector performance of compound semiconductor radiation detectors, as follows:

$$\frac{CCE}{100} = \frac{Q_{\text{collected}}}{Q_{\text{induced}}} = \frac{\mu \tau E}{w} \left(1 - \exp \left(-\frac{w}{\mu \tau E}\right)\right)$$

(6)

By evaluating the field strength dependence of CCE, the product of carrier mobility ($\mu$) and lifetime ($\tau$) can be obtained. The product $\mu \tau$ of diamond has been reported to be $10^{-6} \sim 10^{-3}$ cm$^2$/Vs at room temperature [2,14,22]. The carrier mobility and lifetime are temperature dependent due to the effect of phonon scattering. At cryogenic temperatures, extremely high $\mu \tau$ of 0.2 cm$^2$/Vs have been reported [23]. On the other hand, at high temperatures, the CCE tends to decrease as the mobility decreases [3,24,25]. The mobility can be evaluated by the measurements of pulse width of current pulses. For diamond, saturation drift velocity of about $10^5$ cm/s, mobility of 1600–2000 cm$^2$/Vs, and carrier lifetime of several ns to several tens of ns have been reported using ultra-high purity diamonds by ToF (Time of Flight) measurements using a particle and UV laser.

2.5. Factors degrading detector characteristics

The characteristics of the detector, described in Section 3, depend on the crystal quality such as impurities and crystal defects. It is known that the CCE of diamond detectors decreases with an increase in the amount of impurities such as boron [26] and nitrogen [27]. It is also shown that CCE decreases in textured diamond detector. CCE, however, improves with decrease of dislocation density as shown by mapping using IBIC (Ion Beam Induced Charge) in Figure 4 [28].
Table 1 summarizes the impurities, defects and capture cross sections, and evaluation methods. Except for the typical dopants of diamond semiconductors such as boron and nitrogen, it is not clearly understood which defects and impurities affect the capture of charge carriers. In addition, defect formation due to irradiation also significantly degrades the detector characteristics. This will be discussed in detail in Section 6.

3. Classification of diamond detectors by crystal and device structure

In this section, we will review the classification of diamond detectors by crystal and detector structure.

3.1. Classification by crystal

3.1.1. Single crystal diamond

3.1.1.1. CVD diamond. Diamond grown by microwave plasma enhanced chemical vapor deposition (CVD) is used for energy spectroscopy applications because of its excellent CCE and energy resolution. The disadvantage of single crystals is the detector size: the diameter per one device is limited to a few mm square. This is due to the non-availability of high-quality, large-area diamond single crystals in the market. Most of the single crystal diamond substrates for research applications are a few mm square to up to 10 mm square. However, mosaic single crystal diamond wafer is a promising way to increase area of the single crystal diamond detector. This is because single crystal mosaic diamond wafers up to 2 inches in diameter have been reported using bonding of tiled clone single crystal diamonds. Dislocation density of the mosaic diamond wafers is comparable to that of HPHT type Ib diamond substrate except for the bonding boundary [34]. This method could offer large area and multi-channel single crystal diamond radiation detectors.

3.1.1.2. HPHT diamond. There were several reports on detector properties using high purity HPHT type
Ila diamond in the early 2000s to 2010s. Though type Ila diamond presents good spectroscopic properties for hole drift operation. However, electron trapping is particularly prominent in the transport properties.

3.1.1.3. **Natural diamond.** There were many studies using natural diamond detectors from the 1970s to 2000s [10,18,35]. However, it is not used now due to the progress in synthetic diamond radiation detectors.

3.1.2. **Heteroepitaxial CVD diamond**

Advantage of heteroepitaxial diamond is its large wafer size. Diameters up to 3.5 inches have been reported for heteroepitaxial CVD diamond substrate using Iridium/YSZ [36], and 2 inches using sapphire crystals [37]. It is possible to provide both large detection area and good spectroscopic properties comparable to single crystal diamond radiation detector for hole drift operation. The disadvantage of heteroepitaxial diamond is the high dislocation density of $10^6$ cm$^{-2}$. Several methods have been shown to reduce the dislocation density of heteroepitaxial diamond [38–40]. Reduction of the dislocation density may further improve the carrier transport characteristics of electron and could lead to even closer detector properties to single crystals diamonds [29,41].

3.1.3. **Polycrystalline CVD diamond**

Advantage of polycrystalline CVD diamond detectors is that they enable a larger sensitive area than single crystal diamond detectors. Polycrystalline CVD diamond is formed by CVD growth on Si substrates or metals. They are mainly used for timing measurement for beam accelerators. It is difficult to obtain good spectroscopic properties due to charge trapping at grain boundaries and non-uniformity of electric fields.

3.2. **Detector configurations**

Figure 5 summarizes the configurations used in diamond detectors.

3.2.1. **MIM (metal-insulator-metal) structure**

This is the most widely used detector structure. Self-standing intrinsic diamond plate is sandwiched between two electrodes. Most of the research reports on diamond semiconductor detectors are based on planar electrode structures, but the operation of detectors using pixel or strip electrodes for position detection has also been reported.

3.2.2. **pn and pin structures**

Since the realization of n-type diamond [42] and diamond pn junctions in the 2000s [43], some research groups have reported diamond pn and pin structure radiation detectors. The pn junction can have a larger built-in potential than the conventional MIM structure, and due to the stability of the junction interface, it may operate more stably than the Schottky type at high temperature. The detector characteristics of pin diodes grown on both [111] and [100] have been reported [44]. It is reported that the pin-diamond detectors on [100] saturate the CCE at lower applied voltages and show better energy resolution than that of grown on [111] [45,46].

3.2.3. **Stacked structure**

The stacked detector consists of a highly boron-doped diamond layer for contact and high-purity diamond for drift layer. After deposition, the boron-doped layer is exposed by polishing or etching [47,48]. The advantage of this structure is that the sensitive layer can be thin, which makes it difficult to fabricate a self-standing film.

**Figure 5.** Device structures used in diamond radiation detectors. (a) Planar type MIM detector, (b) pn/pin structure, (c) stacked layer (pseudo vertical) structure, (d) three-dimensional structure.
For thermal neutron measurement, the charged particles emitted from neutron converters (LiF and B$_4$C) travel a few tens of micrometers in diamond. By reducing the thickness of the film, it is possible to reduce the sensitivity to γ rays, which are background noise in neutron measurements.

### 3.2.4. Membrane detectors
Membrane diamond radiation detectors have an extremely thin drift layer using etching of self-standing diamond plates [49]. Figure 6 presents a picture of the membrane diamond fabricated by Ar/O$_2$ plasma etching. The central thin film area is supported by the surrounding thick film. Since the travel distance of charge carriers generated by radiation can be reduced, good CCE characteristics have been reported even for medium grade CVD diamonds that contain several ppm of nitrogen impurity. The energy loss of an ion beam is small because the sensitive layer is a thin freestanding film. For this reason, membrane type diamond detectors can be used for beam extraction windows at accelerators for irradiation of biological samples. In addition, because the carrier collection distance can be smaller, it is less susceptible to the degradation of characteristics caused by radiation than conventional thick diamond radiation detectors [50].

### 3.2.5. Three-dimensional structure
The concept of three dimensional radiation detector has already been proposed in Si and GaAs detector technology for high energy physics experiments to enhance radiation hardness of the detector [51]. Diamond radiation detectors with a three-dimensional structure are made by forming graphite electrodes by laser irradiation [52–54]. Figure 7a shows 3 D diamond electrode arrays and Figure 7b presents charge collection property of 3 D detector and planar configuration. As with membrane diamond radiation detectors, good charge collection characteristics have been reported at low voltages by reducing the carrier collection distance. This also improves radiation hardness of the detector.

The detectors were fabricated on the same single-crystal diamond, a reference conventional planar detector, where the electrodes are fabricated with two arrays of superficial graphitic traces, and a 3 D-fs sensor in the interdigitated combs configuration made with fs-laser columns, connected with superficial graphitic combs on a same side of the sample. Irradiation was performed using $^{90}$Sr beta source.

### 4. Hardness against environment
Diamond radiation detector is promising for application in harsh environments because of its excellent chemical stability, operation at high temperatures, and radiation hardness. Historically, diamond has been used to measure neutrons and neutral particles generated from

![](image_url1)

**Figure 6.** Defect-less bare 2 × 2 mm$^2$ and 3 μm thick membrane etched in 40 μm thick 3 × 3 mm scCVD diamond. Published in Ref [49]: Copyright © 2013 AIP Publishing LLC.

![](image_url2)

**Figure 7.** (a) 3D diamond electrode arrays formed by femto-sec laser (b) Mean signal of planar and 3 D sensors as a function of bias voltage. Published in Ref [52] Copyright © 2013 AIP Publishing LLC.
nuclear fusion reactors. In recent years, due to the accident at the Fukushima Daiichi Nuclear Power Plant, there has been a trend to use diamond for decommissioning and radiation monitoring in harsh environments. In this section, high temperature operation and radiation resistance will be discussed.

**4.1. High temperature operation**

In a semiconductor, the intrinsic carrier density decreases as the bandgap increases, resulting in a smaller leakage current. This makes it possible to operate the diodes at high temperatures. Diamond, an ultra-wide gap semiconductor, is expected to operate at even higher temperatures than conventional wide gap semiconductor materials such as SiC. Diamond Schottky barrier diodes have been reported to operate stably at 400 °C for 1500 h [55].

Diamond radiation detectors have been confirmed to operate at temperatures between 150 °C and 500 °C [1,2,56,57]. Figure 8 shows alpha-particle induced charge measurements of diamond radiation detectors from 298 to 573 K. Though the detector presents good CCE at 300 °C, the degradation of CCE and energy resolution were observed due to the decrease in mobility and carrier lifetime with increasing temperature. SiC radiation detector has also been reported to operate at high temperatures, however, degradation of energy resolution due to increased leakage current has been reported at 300 °C [58].

CCE measurements induced by incident 5.486 MeV alpha-particles of $^{241}$Am up to 573 K for the detector of Diamond Detector Ltd., (DDL). The CCE and the resolution of DDL diamond indicated 98.2% and 1.6% of holes and 97.9% and 1.1% of electrons at room temperature. At 523 K, the CCE and the resolution of DDL diamond indicated 98.2% and 2.1% of holes and 96.7% and 2.2% of electrons. At temperatures higher than 573 K, only electron spectra were detected. The values were CCE 93.8%, with 2.6% resolution. At 623 K, no detection was possible.

**4.2. Radiation hardness**

The primary radiation damage that can affect semiconductor radiation detectors is atomic displacement. Atomic displacement occurs due to non-ionizing loss (NIEL) processes [59,60] such as Rutherford scattering. The NIEL is described as a function of scattering cross section, and displacement energy. These depend on the atomic number and mass number, and atomic displacement energy ($E_d$) of semiconducting materials. Table 2 summarizes the displacement energies of Si, SiC, GaN, and diamond. These wide gap materials have higher binding energies than Si [61].

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**Table 2. Displacement energy of semiconductor materials.**

| Material | Displacement energy (eV) |
|----------|--------------------------|
| Silicon  | 13 [61]                  |
| Diamond  | 35 [62]                  |
| SiC      | 38(Si), 19(C) [63]       |
| GaN      | 39(Ga), 17(N) [64]       |

Figure 9 shows the NIEL of Si and diamond [65,66]. It is expected that diamond is several times more radiation tolerant against protons up to several MeV, and one order of magnitude more resistant above several hundred MeV. Furthermore, the NIEL for neutrons is expected to be more than an order of magnitude smaller because of the smaller damage by recoil atoms and fission fragments due to the light element composition. This is an advantage compared to other wide bandgap materials such as GaN, SiC, which have the equivalent displacement energy as diamond. On the other hand, it is important to note that NIEL can only take into account the amount of defect formation. In order to understand how irradiation affects degradation of the detector properties, it is important to investigate the energy level of each defects and the capture cross section that reduces carrier mobility and lifetime.
It has been experimentally confirmed in high energy physics and nuclear fusion experiments that diamond radiation detectors have higher radiation hardness against protons and neutrons than silicon detectors by beam irradiation experiment using accelerators. Diamond is also radiation tolerant to photons (X-rays and γ-rays). The interaction of photons with matter is given by the product of density and atomic number to the 3-5 order of magnitude. (ρZ^{3-5}) The small atomic number of diamonds is less likely to be damaged by the same photon flux compared with other semiconducting materials.

The degradation of detector properties can be evaluated as a function of radiation fluence, with the following relationship between mean free path (λ) and irradiance [67]

\[
\frac{1}{\lambda} = \frac{1}{\lambda_0} + k\Phi
\]  

(7)

where \(\lambda_0\) is the initial value of MFPs before irradiation, \(\Phi\) is the irradiation fluence and \(k\) he damage factor. Figure 10a shows inverse of MFPs as a function of 800MeV protons fluence for single-crystal CVD and polycrystalline CVD detectors. MFPs of the detector are similarly reduced in both single and polycrystalline diamonds. Figure 10b presents measured mean free path (MFP) as a function of fluence. Dashed line shows the damage model expressed in eq. 7. The model is in good agreement with experimental data.

Equation 7 can also be used for parameters such as CCD, CCE and carrier lifetime. There are attempts to associate the above equation with the physical properties of the material, such as mobility, carrier lifetime, and capture cross section [68]

Detectors with radiation damage have a reduced carrier lifetime. In addition to the amount of defect formation, capture level and capture cross section are directly related to the detector characteristics. In particular, the CCE and energy resolution are likely to be degraded due to the polarization effect under irradiation of high radiation flux. The decrease in CCE due to polarization is temporary and can be recovered by reversing bias voltage or increasing the applied voltage. To prevent polarization effect, it is effective 1) to use high purity single crystal diamond to achieve ideal charge collection. 2) to neutralize the polarization effect using periodic reversing bias [69] 3) reduce the carrier travel distance by using a thinner detector structure or a three-dimensional structure referred to in Section 3.2. These structures mitigate degradation of CCE and increase longevity of detectors.

5. Applications

5.1. Nuclear engineering

In the field of nuclear engineering, since the accident at the TEPCO's Fukushima Daiichi Nuclear Power Plant on March 11, 2011, there has been a growing demand for monitors that can withstand severe accidents. As one example, devices are necessary for operations in reactor containment vessels at high temperatures of 300 °C with integrated doses of 5MGy of gamma-ray irradiation.

The author of this review, Kaneko et al., have been working on the development of diamond γ-ray detectors and diamond FETs for the development of diamond radiation measurement systems for use in reactor containment vessels [70]. The high temperature operation of 500 °C has been achieved by growing high purity diamond single crystal and reducing surface leakage current by guard ring. For radiation resistance of the diamond radiation detector, we have confirmed that the characteristics of the detector do not degrade even after γ-ray irradiation with an integrated dose of > 3 MGy [71].
In such a harsh environment, the entire radiation measurement system is also required to have radiation hardness and heat resistance. In order to satisfy these requirements, diamond FETs are under development. For radiation hardness and high temperature operation of the FETs, diamond MESFETs have been shown to operate stably at 10 MGy and 500 °C [72]. Radiation hardened Hydrogen-terminated Diamond metal–oxide–semiconductor FET (RADDFET), which enables higher transconductance than diamond MESFET and nearly temperature independent operations, were also stably operated after x-ray irradiation up to 1 MGy [73]. Figure 11 shows drain current and drain voltage characteristics of diamond FETs after X-ray irradiation. Clear pinch-off characteristics were maintained after 1 MGy irradiation.

**Figure 10.** (a) inverse of MFP as a function of 800 MeV protons fluence for scCVD and pCVD samples. (b) Measured mean free path (MFP) as a function of 24 GeV protons equivalent fluence compared to the radiation damage model (dotted curve). The figure is from Ref [67] Copyright © 2018 Elsevier.

**Figure 11.** Drain current and drain voltage characteristics of RADDFET (a) before and (b) after 10 kGy, (c) 100 kGy, and (d) 1 MGy. After the MGy of X-ray irradiation, diamond FETs presents stable drain current and drain voltage characteristics. The figure is from Ref [72] Copyright © 2020 AIP publishing.
Other applications in the nuclear engineering is the detection of alpha particles derived from nuclear fuel in nuclear fuel reprocessing plants [74]. In the plants, the reprocessing process of nuclear fuel containing uranium melts under strong acid, which requires the measurement of alpha particles at temperatures above 100 °C. Since diamond is chemically stable, it is suitable for such applications because of its excellent corrosion resistance. Diamond radiation detectors also offer high temperature operation as mentioned above. It has been reported that polycrystalline diamond radiation detectors present long stability for this application.

5.2. Nuclear fusion

In the field of nuclear fusion, diamond radiation detectors have been used in plasma diagnostics. Radiation measurements are the probe to obtain conditions of plasma such as ion temperature. The detector is used to measure the energy of neutrons produced by DT reactions, and the flux of neutral particles produced by plasma recombination [75,76]. The use of diamond radiation detectors is expected to reduce the frequency of replacement of the detector due to its superior radiation hardness. It also allows high temperature operation of the detector. Diamond has a low sensitivity to γ-rays due to its low atomic number. Since γ-rays and neutron radiation are mixed in many neutron fields, semiconducting materials which have low atomic numbers are suitable for measuring the energy of charged particles from nuclear reactions, discriminating them from the background of γ-rays.

The method of neutron measurements depends on the energy of the neutron. For fast neutron measurements, diamond radiation detectors use a $^{12}$C(n, α) $^{9}$Be reaction of carbon in diamond. Figure 12 shows the spectrum of a single-crystal diamond detector for DT neutrons, where the peak around 8.4 MeV corresponds to the $^{12}$C(n, α) $^{9}$Be reaction in diamond. A high energy resolution of 1.4% has been achieved using high quality single crystal diamond. Prior to 2000s, selected natural diamond was used for such applications [35,78]. With the improvement of crystal quality, CVD diamond detectors which have reproducibility have been used for the measurements [77]. For the measurement of thermal neutrons, a stacked structure of neutron converters such as LiF and B$_4$C is used. These nuclides react with neutrons. Neutrons are measured by detecting the charged particles [79].

In the ITER project, a diamond radiation detector is installed as a Radial Neutron camera (RNC). In the In-Port RNC, the detector should operate at ~100 °C and survive 250 °C of the baking process. 14 MeV neutron fluence at the single crystal diamond detector location is expected to be of the order of $10^{16}$ n/cm$^2$ over the whole ITER lifetime [6]. Diamond radiation detector is experimentally confirmed to have a neutron irradiation tolerance of $10^{14} – 10^{15}$ n/cm$^2$. As mentioned in Section 5.A, radiation hardness is required for both the detector and FETs. It is reported that Hydrogen-terminated diamond FETs stably operate after $10^{12}$ cm$^{-2}$ of 14 MeV neutrons generated from DT reactions [80].

5.3. High energy physics

Diamond radiation detectors have been applied as beam monitors in accelerators and synchrotron radiation facilities because of their fast response time and radiation hardness [81]. In recent years, luminosity (beam intensity and flux) has been increasing in the field of high energy physics, and there is an urgent need to improve radiation tolerance of detectors and related electronics [82]. In an accelerator, diamond radiation detectors are used as beam stop monitors for safety purposes, and used as detectors to detect trajectories in high energy experiments [83]. Polycrystalline diamond is used in this application due to its large area. The types of radiation to be detected vary widely by the facility, including protons, electrons, neutrons, and pions.

The RD42 project at CERN has been the most pioneering in the development of diamond detectors in this field [5,84]. The development of diamond radiation detectors for high energy physics has been actively pursued. There have been reports on radiation hardness of single and polycrystalline diamond beam monitors as mentioned in Section 4.2. More recently, they focus on the improvement of radiation tolerance of detectors using three-dimensional structures as mentioned in Section 3.2.

5.4. Medical applications

Radiotherapy is an important tool in the treatment of cancers. It requires accurate dosimetry to measure high gradients of dose over an area of few square mm to ensure that dose is accurately delivered to the target area surrounded by healthy tissue. Dosemeters for
radiotherapy should have tissue-equivalence ($Z = 7.6$) to avoid difference in energy or type of radiation response, and high spatial resolution. Ionization chambers and Si semiconductor radiation detectors have been used in this field. Compare with the conventional detectors, diamond radiation detector is suitable for medical dosimetry because it is near tissue equivalent ($Z = 6$) than Si detector ($Z = 14$) and can measure dose with a high spatial resolution and high sensitivity than conventional gas ionization chambers [4]. Historically, natural diamond radiation detectors have been tested as a dosimeter for X-ray radiotherapy. Progress on growth of detector grade CVD diamond has led to producing diamond detectors with excellent reproducibility and it is currently being commercialized [85,86]. Figure 13 shows single crystal diamond detector for medical therapy. Stacked structure configuration (Pseudo vertical diamond SBD) is used to fabricate thin sensitive layers. The detector is encapsulated in a waterproof mount and connected to coaxial cable.

In recent years, proton beam and heavy ion beam radiotherapies have become increasingly popular [87–89]. Compared to X-ray radiotherapy, these radiotherapies can deliver dose locally, thus minimizing damage to healthy tissues. Diamond detectors are used for beam profile calibration to determine the irradiation range of radiation. Localized irradiation of biological tissues requires consideration of the biological effects of secondary electrons ($\delta$-rays), it is necessary to evaluate the distribution of the energies transferred in a small volume. Detectors with a high spatial resolution of 100 μm square and several μm in depth have been developed using diamond detectors for this purpose [4,88].

5.5. Nuclear battery

In this section, we will introduce a nuclear battery as a technology similar to radiation detectors. Nuclear battery is a device that converts energy from radiation into electricity. The cells consist of radioisotopes and the diodes. It has similar operating principle as a solar cell and uses a semiconductor junction for the energy conversion [90]. The longevity of the battery depends on the half-life of the radioisotope and radiation tolerance of the cell (diode). For long operation, low energy beta emitting radioisotopes are suitable to mitigate irradiation induced damage for diodes. Using $^3$H ($T_{1/2} = 12\text{yrs}$), $^{63}$Ni ($T_{1/2} = 100\text{yrs}$) as the beta-source, a long operating time from several years to decades can be realized. X-ray voltaic batteries are also used for this purpose. Alpha voltaic batteries offer higher energy density than these two types of batteries, however irradiation by heavy ion induces damage for diodes, and it degrades energy conversion efficiency [91–93].

Nuclear batteries are expected to be applied to supply power to the devices in remote environments such as space and underground. The power density of a nuclear battery is the order of several nW/cm$^2$ to µW/cm$^2$, which limits the application to powering ultra-low power sensors and electronics. Since there is a trade-off between lifetime of the device and output power density, it is necessary to select a proper isotope according to the application.

Recently, the use of wide bandgap materials has been intensively studied to improve the energy conversion efficiency of nuclear batteries [94,95]. The reason for this is that the built-in potential of the diode increases with the increase in the band gap of the semiconductor material. As a result, the open-circuit voltage and energy conversion efficiency of the cell are improved. By using diamond pn junction, which is an ultra-wide gap material, a conversion efficiency is expected to be approaching 30% [96].

For diamond, several characterizations using Schottky diodes (SBDs) have been reported. A Russian research group has reported a prototype of a diamond nuclear battery [97,98]. Figure 14 shows a view of the prototype of the diamond micro battery. By combining of 130 single cells based on diamond Schottky barrier diodes, the total active area of about 15 cm$^2$ was obtained.

Figure 13 Single crystal diamond detector for medical therapy from Ref [86]. (a) Longitudinal cross section of the encapsulated diamond detector, (b) x-ray image of the device, and (c) photo of the waterproof housing and of the triaxial connector. The figure is from Ref. [85] © 2012 WILEY-VCH Verlag GmbH & Co.
They reported open circuit voltage \(V_{oc}\) of 1-1.85 V, and output power of 45 nW to 36 μW using several radioisotopes.

Although it is a tiny device, characteristics of the nuclear battery using a diamond pn junction have also been reported. Since the diamond pn junction has an built-in potential of about 4.5 V, which can further improve the energy conversion efficiency of the cell. Here, we will introduce the improvement of energy conversion efficiency using diamond pn junctions. Figure 15a shows schematic of the experimental setup. Instead of beta emitting radioisotopes, the electron beam of SEM is used for characterization. The diameter of the diode is 240 μm. Figure 15b shows the P-V and I-V characteristics of the diode under 15 keV electron beam irradiation. The open-circuit voltage \(V_{oc}\) was 4.26 V. The short-circuit current \(I_{sc} (V = 0\) V\) was 10.7 nA, and the maximum value of \(P (V \times I)\), \(P_{MP}\) was 38.6 nW. From the obtained values, Equations (1) and (2)

\[
FF = \frac{P_{MP}}{I_{sc} V_{oc}} \quad (8)
\]

\[
\eta_s = \frac{V_{oc} FF}{\epsilon} \times 100 \% \quad (9)
\]

Using the equation (8) and (9), the fill factor (FF) and the conversion efficiency \(\eta_s\) are calculated from the current-voltage characteristics of the semiconductor: \(FF = 0.85\) and \(\eta_s = 28\%\). Here, the average electron-hole pair formation energy \(\epsilon\) was set to 13 eV. For \(\eta_s\) obtained from the I-V and P-V characteristics of semiconductors, the reported value for diamond SBDs using electron beam irradiation is 11% [99]. Diamond pn junctions have achieved a conversion efficiency that is more than twice that of diamond SBDs and the highest for a nuclear battery [100].

6. Summary and outlook

In this paper, we briefly reviewed the physics, classifications and recent research progress in diamond radiation detectors as well as related research topics. The properties of diamond radiation detectors are described by mobility and carrier lifetime. These characteristics are affected by temperature, defects, and impurities. In particular, impurities strongly affect the carrier transport characteristics in the detectors. Improvements in microwave enhanced CVD diamond growth technology have led to ultra-high purity diamond crystals. These single crystal diamond radiation detectors showed ideal charge collection efficiency and energy resolution under irradiation conditions with low count rates at room temperature.

Different types of crystals and detector structures required depend on purposes. For energy spectroscopy applications with small area, single crystal diamond is used. Polycrystalline diamond can cover a large area and is used for timing and position detection. Heteroepitaxial diamond has been shown to have characteristics similar to those of single crystal for hole drift mode. It may offer a larger detection area for spectroscopy than single-crystal diamond. Moreover, the size of the single-crystal diamond wafer has also been increasing in recent years due to the realization of mosaic diamond wafer and progress of HPHT diamond growth technology. Though the most common detector is the planar structure, concepts such as thin-film type and three-dimensional structure have been proposed, which presents operation with lower bias voltages than conventional planar-type detectors and improves radiation hardness by reducing charge travel distance in the detector.

One of the promising fields for use of diamond radiation detectors is in harsh environments. Compared with other wide-gap materials, diamond has a particular advantage in high-temperature operation. Operation at temperatures close to 500 °C has been confirmed for diamond radiation detectors and MESFETs. In addition to detectors and FETs, it is also required to develop discrete devices which withstands harsh environments.

The characteristics of diamond radiation detectors degrade by radiation damage. Even with diamond, which has excellent radiation hardness, degradation of detector characteristics due to irradiation is inevitable. The details of the mechanism of degradation due to irradiation are still unclear. Detailed investigation of the charge trapping levels and capture cross sections of defects formed by irradiation will be required. Regarding the enhancement of radiation hardness, in silicon technology, improvements in radiation hardness have been reported through microfabrication of semiconductor
devices. Damage-less micro and nano processing technology will be required to further enhance the advantages of diamond semiconducting devices.

Disclosure statement

No potential conflict of interest was reported by the authors.

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