

**SYNTHESIZE AND CHARACTERIZATION OF
CELLULOSE NANOFIBRILS TO MITIGATE
RADON GAS EMANATIONS FROM
FABRICATED COMPOSITE BRICK FOR
HEALTHY ENVIRONMENT**

NURFARAH AINI BINTI MOCKTAR

UNIVERSITI SAINS MALAYSIA

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RADON GAS EMANATIONS FROM
FABRICATED COMPOSITE BRICK FOR
HEALTHY ENVIRONMENT**

by

NURFARAH AINI BINTI MOCKTAR

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LIST OF SYMBOLS

| | |
|--------------------|-------------|
| % | Percentage |
| g | Gram |
| M | Molar |
| α | Alpha |
| $^{\circ}\text{C}$ | Temperature |
| Θ | Theta |

LIST OF ABBREVIATIONS

| | |
|--------------------------|---|
| μm | Micrometer |
| BET | Brunauer-Emmett-Teller |
| Bq/m^3 | Becquerels per cubic meter |
| Ca | Calcium |
| Cl | Chlorine |
| CNC | Cellulose Nanocrystal |
| CNF | Cellulose Nanofibrils |
| EPA | United States Environmental Protection Agency |
| FESEM | Field Emission Scanning Electron Microscope |
| H_2O_2 | Hydrogen Peroxide |
| ml | Milliliter |
| mm | Millimeter |
| N | Newton |
| Na_2SO_3 | Sodium Sulphate |
| NaOH | Sodium Hydroxide |
| nm | Nanometer |
| OPEFB | Oil palm empty fruit bunch |
| Pb-206 | Lead |
| pCi/L | Picocuries per Liter |
| Pd | Palladium |
| Po-218 | Polonium |
| Ra-226 | Radium |
| Rh | Rhodium |

| | |
|--------|----------------------------------|
| Rn-222 | Radon |
| Sb | Antimony |
| TEM | Transmission Electron Microscopy |
| Th-230 | Thorium |
| U-238 | Uranium |
| UTM | Universal Testing Machine |
| WHO | World Health Organization |
| XRD | X-ray Diffraction |
| XRF | X-ray Fluorescent |

**SINTESIS DAN PENCIRIAN SELULOSA NANOFIBRIL UNTUK
MENGURANGKAN PANCARAN GAS RADON DARIPADA BATA
KOMPOSIT UNTUK PERSEKITARAN YANG SIHAT**

ABSTRAK

Radon adalah gas radioaktif semula jadi yang dihasilkan oleh siri pereputan U-238 dan kebanyakannya berasal dari batu, air, pasir dan tanah. Bata yang biasanya terdiri daripada batu, air dan pasir menjadi sumber utama radon kepada manusia dalam persekitaran bangunan yang tertutup. Gas radon berupaya meresap ke dalam sistem respirasi semasa proses pernafasan dan secara tidak langsung memberi kesan kepada tisu peparu oleh zarah α bertenaga yang berisiko menyebabkan kanser. Fibril nano selulosa (CNF) daripada kenaf dan kelapa sawit yang bertindak sebagai pengisi cecair di dalam bata dengan nisbah tertentu mampu mengurangkan pancaran gas radon. Hal ini kerana kerana pengurangan penggunaan sumber radon seperti batu, pasir dan air serta pengecilan saiz liang bata. Gabungan kaedah pulpa kimia dan ultrasonikasi telah digunakan untuk mensintesis CNF sebelum proses pencirian menggunakan FESEM, XRD, XRF, BET dan UTM. Morfologi permukaan menunjukkan julat diameter fibril dari 30-80 nm untuk kenaf dan 20-60 nm untuk kelapa sawit. Indeks penghabluran untuk kenaf dan kelapa sawit adalah masing-masing sebanyak 75.3% dan 77%. Sepuluh buah bata CNF komposit dan satu buah bata tanpa CNF (kawalan) telah difabrikasi mengikut protokol Malaysia Standard MS 7.6:1972. Lima jenis amaun CNF telah diaplikasi iaitu 40 ml, 80 ml, 120 ml, 160 ml dan 200 ml manakala agen pembusa digunakan untuk mengikat bancuhan. Semua bata dikeringkan dalam kebuk wasap pada suhu bilik selama 48 jam. Radon Sentinel Monitor model 1030 telah digunakan untuk menentukan kepekatan radon, kelembapan, tekanan dan suhu untuk

setiap bata selama 10 hari berturut-turut dalam bilik prototaip perspeks ruang tertutup. Bata kawalan menunjukkan kepekatan radon tertinggi sebanyak 3.77 pCi/L manakala 40 ml bata komposit CNF daripada kenaf dan kelapa sawit menunjukkan kepekatan radon sebanyak 1.4 pCi/L dan 0.93 pCi/L. 40 ml CNF daripada kelapa sawit adalah jumlah optimum untuk mengurangkan pancaran radon dengan pengurangan sebanyak ~75%. Bagi ujian fizikal, bata kawalan telah mengalami keretakan pada daya 39013 N manakala 50458 N dan 42160 N untuk bata komposit daripada 40 ml CNF kenaf dan kelapa sawit. Sementara itu, kawasan permukaan liang untuk bata kawalan ialah 3.4473 m²/g manakala 6.4449 m²/g dan 4.9025 m²/g untuk bata komposit daripada 40 ml CNF kenaf dan kelapa sawit dengan saiz liang 2.92 nm, 0.347nm dan 2.27 nm. Aplikasi amaun CNF yang lebih rendah untuk kedua - dua jenis tumbuhan juga mampu untuk mengurangkan pancaran radon dan turut meningkatkan kekuatan fizikal bata. Dalam kajian ini, 40 ml CNF kenaf dan kelapa sawit adalah nisbah optimum dalam mengurangkan pancaran radon, pengecilan saiz liang dan meningkatkan kekuatan fizikal bata komposit fabrikasi

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ABSTRACT

Radon is a natural radioactive gas produced by U-238 decay series and mostly originated from rock, water, sand and soil. Brick that commonly consist of rock, water and sand becomes the main radon resources to human in indoor building environment. Radon gas can diffuse into respiration system during inhalation process and indirectly affects lung tissues by its energetic α particles that risk to lung cancer. Incorporating kenaf and oil palm cellulose nanofibrils (CNF) as liquid fillers into brick with certain ratios enable to mitigate radon gas emanations. This is due to reduction of radon resources utilization such as stone, sand and water as well as miniaturization of brick porosity. CNF were synthesis using combination of chemical pulping and ultrasonication methods before characterized with FESEM, XRD, XRF, BET and UTM. Surface morphology showed fibril diameter ranges were from 30-80 nm for kenaf and 20-60 nm for oil palm. Crystallinity index were 75.3% and 77% for kenaf and oil palm, respectively. Ten CNF composite bricks and one without CNF (control) were fabricated according to Malaysia Standard MS 7.6:1972 protocol. Five different amounts of CNF had been applied which were 40 ml, 80 ml, 120 ml, 160 ml and 200 ml while foaming agent was used to bind the mixtures. All bricks were dried in fume hood at room temperature for 48 hours. Radon Sentinel Monitor model 1030 was used to determine the radon concentrations, humidity, pressure and temperature for each brick within 10 consecutive days in a close-space of prototype perspex room. Control brick gave highest radon concentration of 3.77 pCi/L whereas 40 ml CNF composite bricks from kenaf and oil palm gave 1.4 pCi/L and 0.93 pCi/L, respectively. 40 ml

CNF from oil palm was the optimum amount to mitigate radon emanation with ~75% reduction. For physical testing, control brick was cracked at force 39013 N whereas 50458 N for 40 ml kenaf and 42160 N for 40 ml oil palm CNF bricks. Meanwhile, surface area porosity for control brick was 3.4473 m²/g whereas 6.4449 m²/g for 40 ml kenaf and 4.9025 m²/g for oil palm CNF bricks with pore size 2.92 nm, 0.347 nm and 2.27 nm for each. Application of small amount CNF for both plants also reduced the radon emanations and increased the physical strength of the bricks. In this study, 40 ml CNF of both kenaf and oil palm were the optimum ratios in mitigating radon emanations, miniaturization the porosity and increased the physical strength of the fabricated composite bricks.

CHAPTER 1

INTRODUCTION

1.1 Background of study

Cellulose are commonly used in any research because it completes the natural fibre mechanical properties (Morán et al., 2008). Cellulose, hemicellulose and lignin are the main part that form the plant structure. In cell wall, there are existence of lignin between cellulose, hemicellulose and pectin component where it fills the spaces and it is an important structural materials in the support tissues (Srndovic, 2011).

On top of that, cellulose nanofibrils has been proven to provide us with many beneficial effects. Recently, many studies have been done to identify and evaluate the CNF of plants. It is commonly synthesis from organic material such as fibers from the wood within 1-100 nm in dimensions. With emerging technologies nowadays, it become the most highly demand from the industry. CNF is the type of cellulose material that has many advantages where it can be applied to most research or in industrial like biotechnology, composite, and also biomedical due to its unique structural features and impressive physical properties such as renewability, low density, adaptable surface chemistry, optical transparency, biocompatibility, biodegradability and improve mechanical properties (Trache et al., 2017). Due to the high crystallinity, CNF form a dense network held together by strong inter-fibrillar bonds and might act as a barrier material (Siro & Plackett, 2010).

From the natural growth, organic plant has a good mechanical properties based on its special structure. Example of organic plant is kenaf and oil palm as shown in Figure 1.1 and Figure 1.2 respectively. Kenaf with scientific name *Hibiscus Cannabinus L* is classified as an angiosperm as it produces seeds and flowers. Bast (bark) and cork (wood) are the main components of fibre production. It is close to jute and cotton because it is a warm season annual fibre crop and it only takes 150 days to harvest. Based on kenaf stalks, it consists of 35-40% bast fibre and 60-65% core fibre by weight. The structural kenaf fibre consists of 65.7% cellulose, 21.6% lignin and pectin and other composition (Lingenthiran, 2015) and kenaf bast was used in this research. Meanwhile, oil palm with scientific name *Elaeis Guineensis* is classified as an angiosperm because it produces seeds and oil palm empty fruit bunch was used to produce cellulose nanofibrils. Same structural component like kenaf, it also consists of 37.3%-46.5% cellulose, 25.3%-33.8% hemicellulose, lignin and others (Sudiyani et al., 2013).

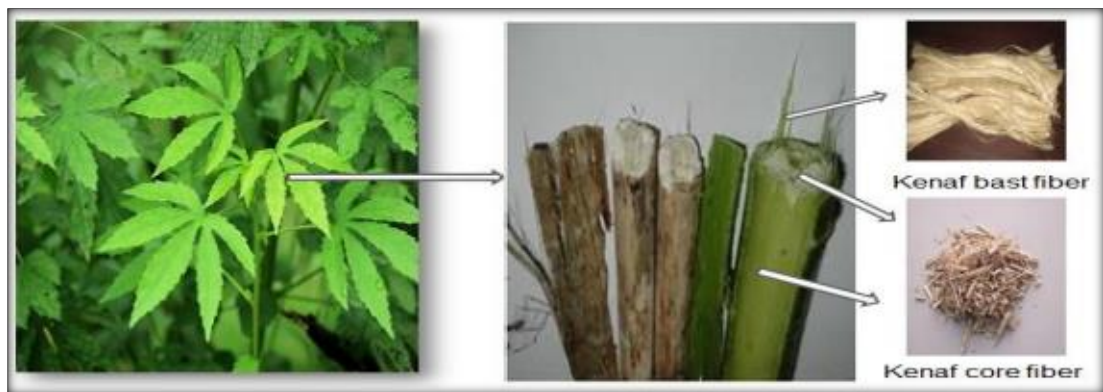


Figure 1.1: Kenaf plant (*Hibiscus Cannabinus L*)(Birnin-yauri et al., 2016)

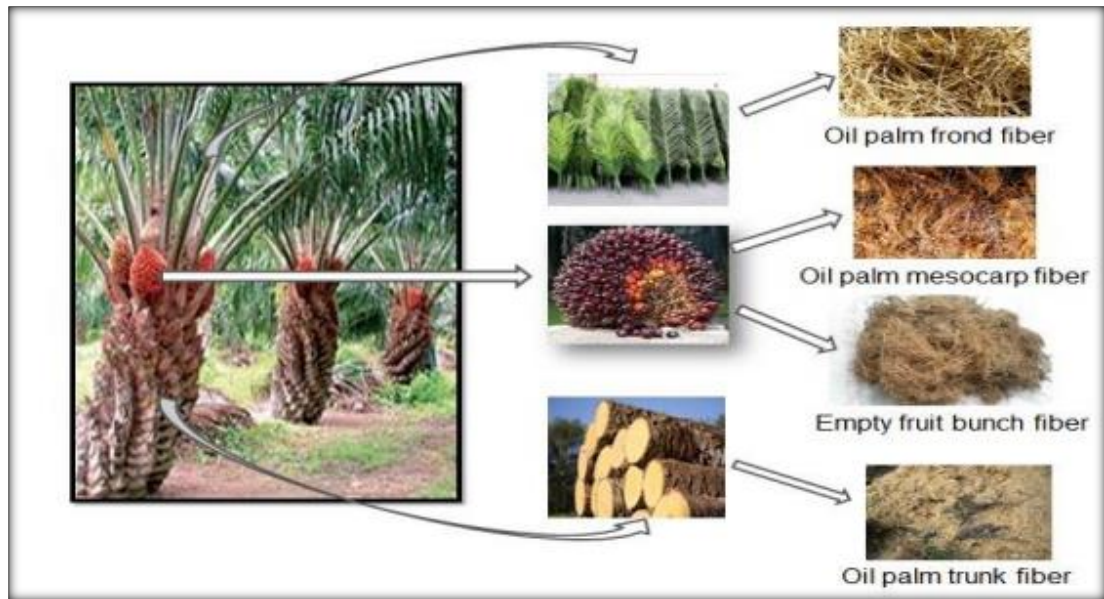


Figure 1.2: Oil palm (*Elaeis Guineensis*) (Birnin-yauri et al., 2016)

Year by year, CNF has become one of the most concern material that is used in all industries. Due to its nano size structure, it is suitable to be applied in brick as a nano filler. In Malaysia, commonly the building is made up from concrete which use stone, cement and sand as the major component to fabricate brick which contain higher radon concentration. The indoor and outdoor temperature has created different pressure and activate the soil air to flow into dwellings through leakage. According to Arvela (2001), the indoor radon concentration in single family houses is close to 150 Bq/m³ where it does not exceed the action level that has been stated by the Ministry for Social Affairs and Health in 1992 since the indoor radon concentration should not exceed 400 Bq/m³ in existing houses.

Rn-222 is already exist since the formation of the earth and it is known as a natural radioactive gas where it is a product from the radioactive decay of small amount of Uranium-238 that already occur in the rocks, soils and water (Iglesias & Taboada, 2014). U-238 breaks down into radium which then decays into Rn-222. It exist in the form of gas and easily move up through the soil into the atmosphere where

it is a colourless, odourless and tasteless radioactive gas (Hatif et al., 2016). Rn-222 and its progeny contribute more than 50% of background radiation in environment (Ramachandran, 2006).

After uranium, thorium and radium, Rn-222 that is known as noble gas moves easily through the ground and have 3.8 days half-life that will decay into alpha particle. Not all gaseous soluble in water but for Rn-222, it is highly soluble in non-polar solvents, soluble in water and also groundwater (Ali, 2013). Rn-222 increase the risk of lung cancer where the higher the Rn-222, the longer the exposure that will contribute to the greater risk. According to the research done, Rn-222 concentration become second leading of lung cancer and increase the environmental cancer mortality (Kim et al., 2016).

On the other hand, Rn-222 is the fourth element to be discovered with atomic mass 222. There are several parameters that influences Rn-222 concentration which are temperature, rainfall, pressure, humidity and wind speed. Countries with different winter and climate face different level of Rn-222 concentration. Rn-222 concentration in winter season is proven to be greater than in the summer season that have been stated by United States Environmental Protection Agency (EPA).

So in order to reduce the Rn-222 emanation in concrete brick, CNF from organic plant has been used as a filler by reducing the usage of sand, cement and stone which contain high U-238. Furthermore, CNF will also close the brick porosity and reduce the amount of Rn-222 concentration from diffused out to the surrounding.

1.2 Problem Statement

Cellulose nanofibrils is an excellent source of natural fibre and is abundantly available in Malaysia as it has gained growing interests owing to their attractive and excellent characteristics . Till date, a few studies have used CNF from different types of fibre. Moreover, the use of kenaf and oil palm CNF in fabricated composite brick has not been reported.

The brick that are used in making building eventually produce Rn-222 gas emanation because it is made from natural radioactive source such as sand, cement, stone and water. United States Environmental Protection Agency (US EPA) has set that the indicator for action level is 4.0 pCi/L. This action level is stated to avoid lung cancer but it is still post risks. In Malaysia, there are lack of community awareness about this Rn-222 gas where the action level is not stated. In order to reduce the Rn-222 emanation, this research will propose fabricated composite brick based on organic CNF that are synthesize from kenaf and oil palm. This organic CNF will reduce Rn-222 gas emanation since the reduction of Rn-222 resources such as stone and sand thus help to close up the brick porosity to prevent Rn-222 gas and improve the mechanical strength of composite brick respectively. Therefore, in the current study, CNF will be used to fabricate composite brick for radiation protection application, as they are a natural source and readily available as biomass waste.

1.3 Justification of study

Recently, CNF have gained much popularity due to its high young modulus, high tensile strength, lightweight and non-toxic (Chirayil et al., 2014). CNF is the major component in fabrication of composite brick in order to reduce Rn-222 concentration and increase the composite brick strength (Razab et al., 2019) . Since

Malaysia has an abundant waste of natural fibre, the fabrication of composite brick with the use of CNF derived from kenaf fibre and oil palm fibre will enable the production and commercialization of a new composite brick using CNF which will add one more commercial product for the Malaysian building industry.

1.4 Scope of study

Concerns regarding the natural fibre limitations, CNF is one of the unique structure to be used in new technologies where there is also a growing popularity and demand for building material fabrication. This demand opens opportunities to the researcher to find the method on how to synthesis CNF with lower cost and higher quality CNF within short time. CNF is produced from the chemical pulping, bleaching and ultrasonication while its morphology is observed through Transmission Electron Microscopy and Field Emission Scanning Electron Microscopy. This research will focus on fabricating the composite brick based on different CNF that is synthesized from kenaf and oil palm fibre. These CNF will be used to reduce the amount of Rn-222 sources as well as to reinforce the composite brick. The Rn-222 concentration reading will be taken using Radon Monitor Sentinel 1030 to measure the amount of the Rn-222 concentration produced by composite brick that use different ratio of sand, stone and cement with different ratio of CNF that act as a filler.

1.5 Objectives

1. To synthesis and characterize organic cellulose nanofibrils from Kenaf and Oil Palm fibers.
2. To fabricate composite brick based on cellulose nanofibrils and determine Rn-222 emanations, mechanical and physical properties of fabricated composite bricks.

1.6 Research Hypothesis

Fabricated composite brick is expected to reduce Rn-222 emanations significantly and enhance blocking mechanisms of α particles. The brick also has high quality in term of physical properties like low porosity, good strength, light weight, high tension and strain resistance compare to ordinary marketable building bricks in Malaysia.

CHAPTER 2

LITERATURE REVIEW

2.1 Natural Fibre

Natural fibre with scientific name lignocellulosic fiber consist of two types of fibre that were plant fibre and animal fibre. Recent research shows that natural fibre was being used in industrial purpose and does not only focus on making textile or fabrics. Nowadays, it mostly used in components of composite materials, medical implants and for agrotextiles. According to the research done by Wambua et al (2003), with increasing fibre weight fraction, the tensile modulus, impact strength and ultimate tensile stress of kenaf reinforced polypropylene composite were found to be increased. The composition of natural fibre bring an advantages to the fibre itself such as light weight, cheap, low density, renewable nature resources and easily available. Depend on the application, their mechanical and chemical properties can be modified where surface modification has been made to increase the properties of natural fiber where the modified natural fiber is able to perform well in advanced sector since there is a higher demand for superior material now (Cheung et al., 2009).

Different types of fibre have different chemical composition that consist of cellulose, hemicellulose, pectin and lignin with different percentage according to different types of fibre where 60-80% composition consist of cellulose, 5-20% lignin and 20% moisture (Saheb & Jog, 1999).

Table 2.0 shows different natural fiber with different chemical content where cellulose content of oil palm EFB and kenaf were 65% and 53.4% respectively.

Table 2.0: chemical content of natural fiber (Hao et al., 2018)

| Fiber | Cellulose | Hemicellulose | Lignin | Extract | Ash content | Water soluble |
|----------------|-----------|---------------|--------|----------|-------------|---------------|
| Cotton | 82.7 | 5.7 | – | 6.3 | – | 1.0 |
| Jute | 64.4 | 12.0 | 11.8 | 0.7 | – | 1.1 |
| Flax | 64.1 | 16.7 | 2.0 | 1.5–3.3 | – | 3.9 |
| Ramie | 68.6 | 13.1 | 0.6 | 1.9–2.2 | – | 5.5 |
| Sisal | 65.8 | 12.0 | 9.9 | 0.8–0.11 | – | 1.2 |
| Oil palm EFB | 65.0 | – | 19.0 | – | 2.0 | – |
| Oil palm Frond | 56.0 | 27.5 | 20.48 | 4.4 | 2.4 | – |
| Abaca | 56–63 | 20–25 | 7–9 | 3.0 | – | 1.4 |
| Hemp | 74.4 | 17.9 | 3.7 | 0.9–1.7 | – | – |
| Kenaf | 53.4 | 33.9 | 21.2 | – | 4.0 | – |
| Coir | 32–43 | 0.15–0.25 | 40–45 | – | – | – |
| Banana | 60–65 | 19 | 5–10 | 4.6 | – | – |
| Sun Hemp | 41–48 | 8.3–13 | 22.7 | – | – | – |
| Bamboo | 73.83 | 12.49 | 10.15 | 3.16 | – | – |
| Hardwood | 31–64 | 25–40 | 14–34 | 0.1–7.7 | <1 | – |
| Softwood | 30–60 | 20–30 | 21–37 | 0.2–8.5 | <1 | – |

2.1.1 Kenaf Fibre

Kenaf was known as *Hibiscus Cannabinus L* that was classify under kingdom plantae. It has economic and ecological advantages because of cellulosic source. It grow quickly as it takes only 150 days to harvest and has good mechanical properties (Samylingam, 2015).

The physical properties of the fiber depend on the area of the plant and the progressions that take place in 500 mm starting from the ground. It is found that kenaf was a suitable biological resources as a substitute for fossil fuels and wood pulps due to its extensive adaptation, strong resistance, large biomass and rich cellulose. (Ramesh et al., 2018).

The main factors which influence the properties of the fiber is its chemical composition. The strength of the composite is mostly influenced by the cellulose content of the fiber. Kenaf bast fiber has cellulose content as high as 60.8% while core fiber is approximately 50.6% (Joonobi et al., 2009). The mechanical properties of the composite increased with increasing the concentration of the alkali where the increase in the property can be achieved by surface modification of the fiber. From the Fourier transform infrared spectroscopy demonstrated that the content of lignin and hemicellulose decreased during the pulping process and lignin was almost removed during bleaching (Thiruchitrabalam & Narayanan, 2016).

2.1.2 Oil Palm Fibre

Oil palm was commonly known as *Elaeis Guineensis* where this plant will only harvest 30 months after field planting and have economic life about 25 years. It consists of 25% oil, 25% seeds, 13% mesocarp fibre, 7% shells and 23% empty fruit bunch (Basri, 2010). According to Isroi & Cifriadi (2017), there are 40.37% cellulose, 20.06% hemicellulose and 23.89% lignin that form the structure of oil palm.

Fiber strength is a crucial factor to choose fibre that is specific for certain usage. According to (Abdul et al., 2008), fibre length is a vital factor in determining bonding and stress distribution. The oil palm empty fruit bunch is a byproduct from the processing of crude palm oil (CPO) in a palm oil mill. It is obtained from the empty stalks of the fresh fruit bunches (FFB) after the fruits are separated from it. Apart from its abundance, OPEFB has high cellulose, making it a very promising feedstock for the extraction of nanocellulose and the production of various cellulose-based products. Compared to other agricultural wastes, OPEFB is a great option as a potential raw material since it is composed of high cellulose content in addition to its abundant availability.

2.2 Cellulose Nanofibrils

Recent research show that CNF have promoted the development of the application in biomedicine, biomaterial, pharmaceuticals, tissue engineering and others. In order of its mechanical and physical properties, CNF was a suitable structure to be apply on the composite brick since it can increase the compression strength which it can bear a higher load compare to commercial brick that has nowadays (Wicklein et al., 2015). For industrial applications, CNF was limited use due to the longer time taken to produce it. Table 2.1 shows the application of CNF in various fields such as in electronic devices, construction, food packaging, food products, paper products, composites, oil recovery, medical and tissue engineering, cosmetic and also automobile (Mishra et al., 2018).

Table 2. 1: Application of cellulose nanofibrils in various fields (Mishra et al., 2018)

| Area of application | Key application |
|--------------------------------|---|
| Electronic devices | Sensor, electronic displays and windows |
| Construction | Sensors to monitor stress levels in bridge |
| Food packaging | Packaging for oxygen barrier |
| Food products | Stabilizers for suspension and flavor carriers |
| Paper products | Greas-proof paper |
| Composites | Improve the mechanical properties of polymer matrices |
| Oil recovery | Fracturing fluid in oil recovery |
| Medical and tissue engineering | Water absorbent pads, antimicrobial films and tampons, sanitary napkins or wound dressing |
| Cosmetic | Composite coating agent for nails, hair, eyebrows or eyelashes |
| Automobile | Lightweight and high strength components such as bumpers, side panels and dashboards |

A prior to that discovery, a different method to synthesis organic plant was proposed in order to produce higher quality and quantity CNF. The method to synthesis

CNF was patented and become popular until most of the research nowadays used CNF to enhance the physical and mechanical properties for their research applications.

Generally, the CNF was found as naturally occurring compound, but it could be derived depends on their applications. Different elements that was found in different CNF structure might be from the nutrients that was absorb during their growth (Sharma et al., 2019).

Nano-structured cellulose or commonly known as cellulose nano-fibrils can be classify into three types that were cellulose nanocrystal, cellulose nanofibers or bacterial nanocellulose. Different types of CNF was used in different applications based on their physical properties. The length of the de-structuring that the raw material undergoes depend on the aspect ratio of the final CNF. The preparation of the CNF from mechanical or chemical method influence the properties and length of the CNF produced. As shown in Figure 2.1, Transmission Electron Microscope reveal that different organic plant have different micrograph of a microcrystalline cellulose from fodder grass, cellulose microfibril from sugar beet, cellulose nano-fibrils from banana peel, cellulose nanocrystal from ramie fiber and amorphous cellulose nano-fibrils from cotton (Kumar et al., 2018)

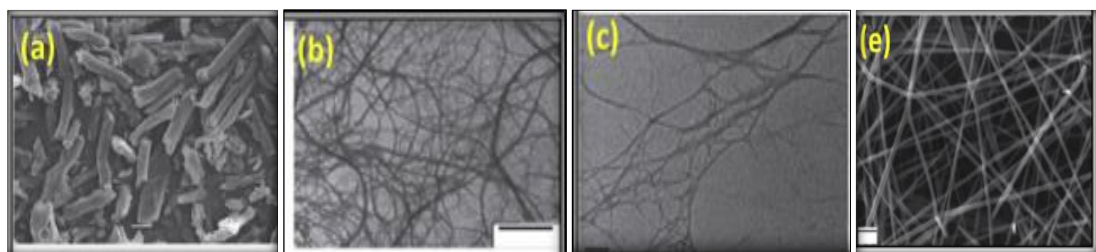


Figure 2. 1: TEM micrograph of (a) microcrystalline cellulose from fodder grass, (b) cellulose microfibril from sugar beet, (c) cellulose nanofibril from banana peel, (d) cellulose nanocrystal from ramie fiber, (e) amorphous cellulose nanofibril from cotton (Mishra et al., 2018)

2.2.1 Synthesis of Cellulose Nanofibrils

There are a few methods to synthesis CNF but the most common method used was chemical pulping because it was easy to handle and prepared. Moreover, smaller size nanofibrils can be obtained from chemical pulping treatment.

The production of CNF was firstly synthesis via chemical pulping method and most of the organic plant CNF have been synthesis using this method (Phanthong et al., 2018). However, in latter year many methods of synthesizing CNF have been well documented which involved various mechanism of reactions such as mechanical method. But, chemical pulping followed by bleaching process and sonication was one of the method that was popularly used.

Figure 2.2 shows Schematic representation of extraction of nanocellulose from lignocellulosic biomass by using different method such as mechanical process, acid hydrolysis and chemical treatment.

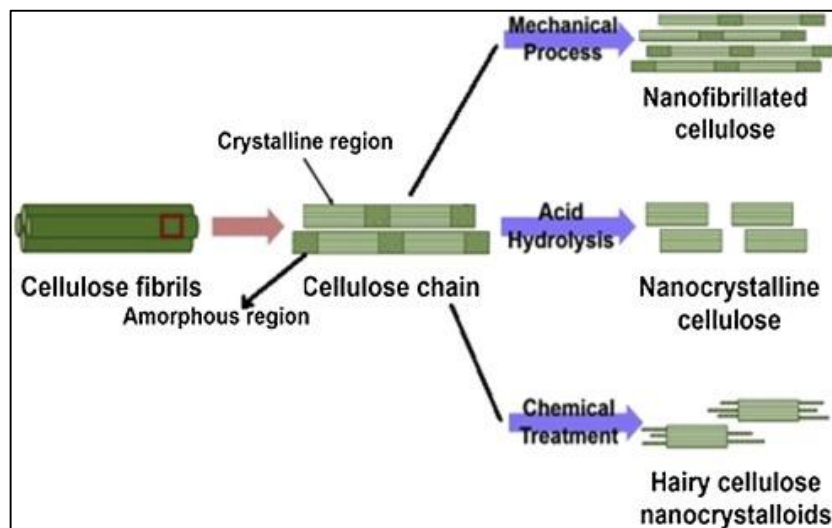


Figure 2. 2: Schematic representation of extraction of nanocellulose from lignocellulosic biomass (sharma et al., 2019)

Another method used is by using chemical-ultrasonication process. The chemical method involves pulping, bleaching and hydrolysis process using sulfuric acid, while the ultrasonic process to obtain cellulose nanofiber. Based on the research done by (Syafri et al., 2018), it shows with the increasing of ultrasonic time, diameter of cellulose ramie decreased due to the particle size distribution of cellulose ramie decreased. According to this research, different time taken for ultrasonication process produced different diameter of CNF that was confirmed by TEM images. Figures 2.3 shows the step involved in synthesis of ramie fibre.

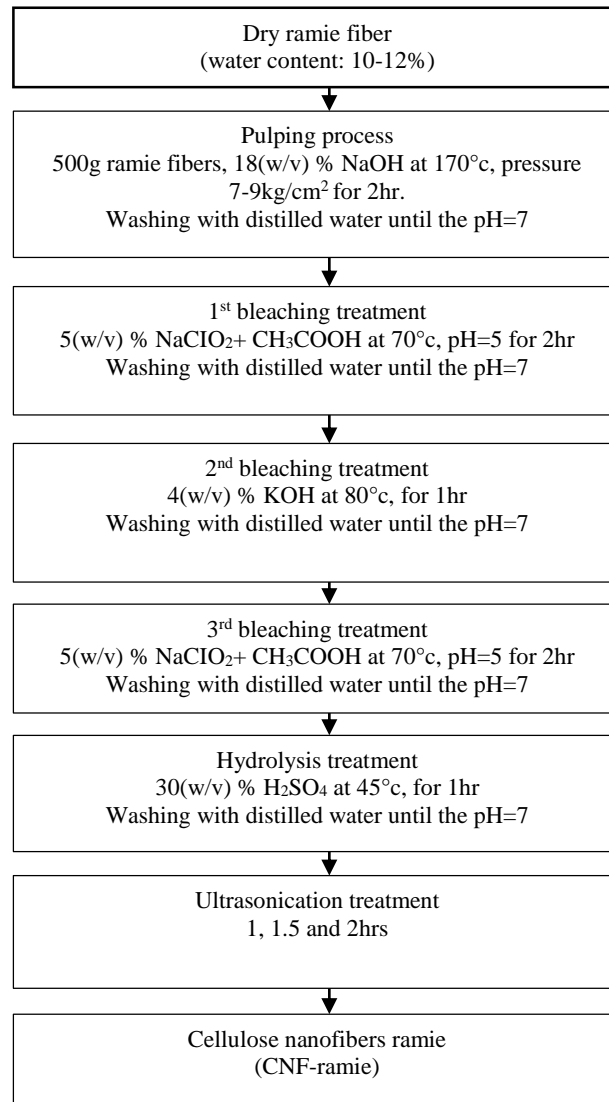


Figure 2.3: Steps involved in synthesis of cellulose nanofibers ramie.

2.3 Environmental Radiation

Radiation exist naturally in our environment since the earth formation and mostly the source that contribute to the radiation formation was come from soil, water and air that caused our exposure to ionizing radiation. There are two types of radiation that are ionizing and non-ionizing (Mubeen et al., 2008). Non ionizing radiation does not carry enough energy to break the molecular bonds and ionize atom that divided into two that are optical radiations comes from the ultraviolet, visible ray and infra-red whereas electromagnetic fields, for example is radiofrequency (Ng, 2003). Ionizing radiation is the type of radiation that carries enough energy to break the bonds between molecules and ionize atom for example were alpha, beta and gamma radiation (Parsa, 1998). Figure 2.4 shows the Source and average distribution of natural background radiation in the world. The presence of Rn-222 was 50% of the natural background that contribute to the radiation. Earth gamma radiation, water and food and also cosmic give the lowest percentage of radiation that were 20%, 12% and 18% respectively. Radioactivity is classified as artificial and natural where artificial radioactivity is when we manually destabilize certain element commonly for purpose of medical treatment and natural radioactivity occur naturally without human intervention such as Rn-222 formation.

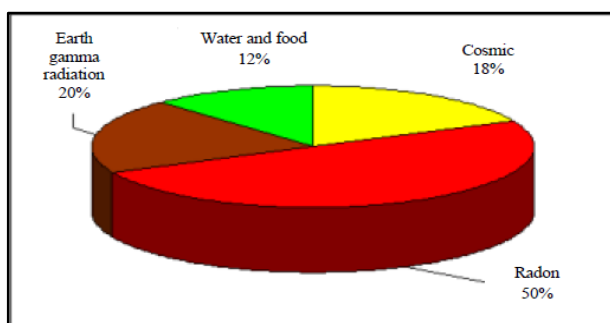


Figure 2. 4: Source and average distribution of natural background radiation for the world (Bowie & Bowie, 1991)

2.3.1 The Formation of Radon

Radon with atomic number 86 is the radioactive element which doesn't produce any smell, taste and cannot be seen naturally (US-EPA, 2001) where it was grouping in group 18 that was a noble gas (Mustonen et al., 1997). Half-life of Rn-222 is only 3.8 days that making it the most unique element that decay away so quickly. Rn-222 occurs from the decay chain of U-238 and Th-230 where most radioisotopes does not decay directly to achieve a stable state but will undergo a series of decay until a stable isotope is reached. Uranium and thorium exist in form of solid whereas radon exist in form of gaseous and easily to be inhaled by people. Figure 2. 5: Decay series of radium with daughter products and half-life, showing the formation of radon

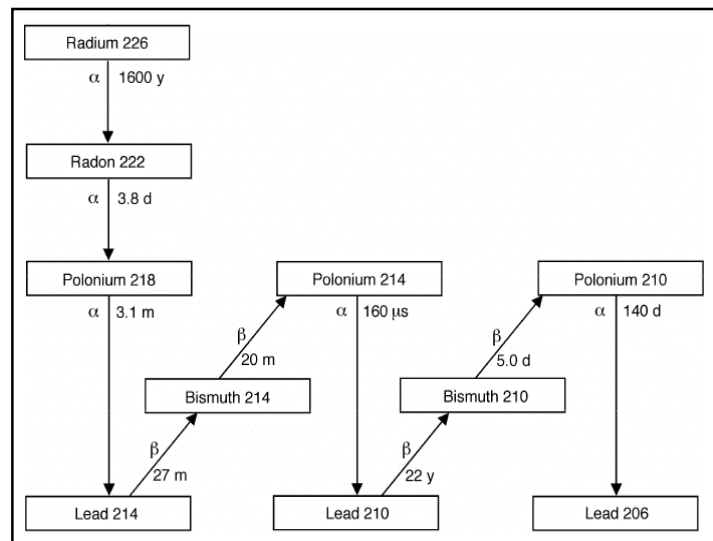


Figure 2. 5: Decay series of radium with daughter products and half-life, showing the formation of radon (Gillmore et al., 2010)

2.4 Source of Radon and How It Enter Building

According to Bruno (2012), the soil adjacent to the foundation, construction materials such as concrete and tap water are the sources of Rn-222 in building. High Rn-222 level occur in regions where the soil or rock that rich with U-238 in the ground

can enter the indoor air where it decays product accumulate in poorly ventilated areas. The main mechanisms of Rn-222 entry are diffusion and advection from the ground and building materials. The environmental temperature and pressure changes causes the Rn-222 emanation to change (Sharma et al., 2015). Figure 2.6 shows the routes by which Rn-222 enters through a dwelling system. Normally Rn-222 can enter the house in a very short time since it exists as a gaseous phase which was easily slip away from the ground. The cracks at the walls of the building generally become one of the factors that contribute to the Rn-222 increase in indoor environment.

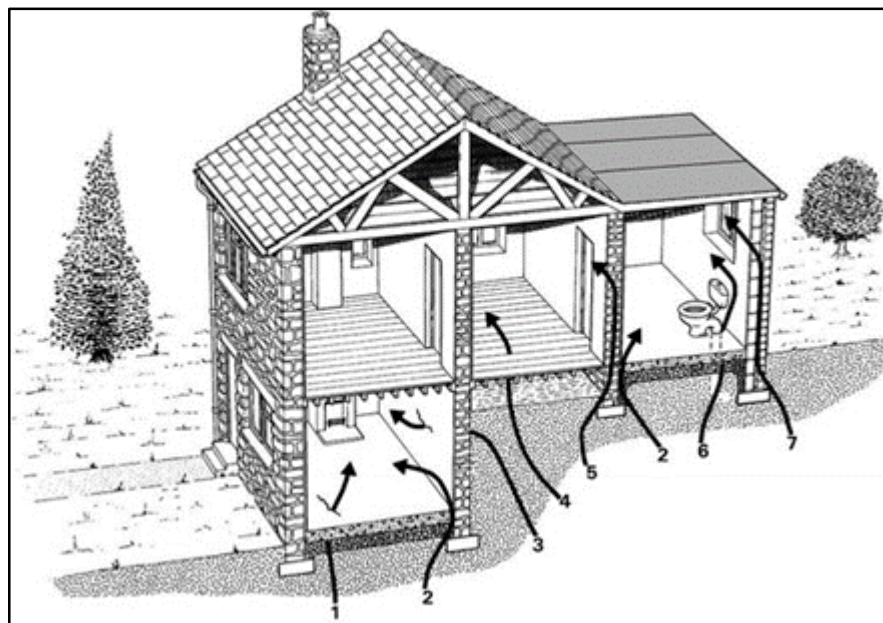


Figure 2. 6: Routes by which Rn-222 enters through a dwelling system (Yousefi et al., 2014)

2.4.1 Indoor and Outdoor Radon Concentration

According to the research from Habib et al (2018), Rn-222 gas concentration may be different due to the seasonal variations. Rn-222 concentration not only depend on seasonal variations but also the construction age of building, ventilation system and also geological materials (Park et al., 2016)

Naturally, Rn-222 exist as gaseous and may enter the building in a different way for example was soil under the building. Other than that, household also contribute to emanation of Rn-222 in the house (Bruno et al., 2012). The concentration of Rn-222 in ambient air varies with the type of soil and underlying bedrock of the area. These factors act as the pathway for Rn-222 to migrate into indoor which can lead to an increase in indoor Rn-222 concentration (Mustafa & Daniel, 2009)

Previous research has proven that outdoor Rn-222 exhibit a lower Rn-222 gas concentration compare to indoor environment (Pavia et al., 2003). To make the evidence stronger, Wu et al (2016) have conducted a survey on 33 provincial cities across China where the outdoor Rn-222 concentration level in China has not changing too much for 20 years compare to indoor Rn-222. However, the natural radiation level mainly from indoor Rn-222 concentration was increase almost 70% compared to previous data in 1990s, that commonly exist from the building materials and the alteration in ventilation (Wu et al., 2016).

2.4.2 Radon Levels in Malaysia and Other Countries

Different countries produce different Rn-222 concentration regarding to the different in geological and climate. In Malaysia, there was no specific laws or action levels on Rn-222 concentration. From the research done by Saat et al (2010), different location in peninsular Malaysia has different Rn-222 concentration due to the types of residential areas that are the city, housing complexes and traditional residential area. Table 2.2 show the comparison of the mean indoor Rn-222 concentration of the present study to the corresponding values of other countries. Rn-222 concentration in mostly tropical region was low because of opened window, so their weekly or monthly Rn-

222 concentration was almost same although there was slightly a bit different in daily average Rn-222 concentration (Mahat, 2007)

Table 2. 2: The comparison of the mean indoor Rn-222 concentration of the present study to the corresponding values of other countries (Mahat, 2007)

| Country | Mean indoor concentration (pCiL ⁻¹) | Reference |
|-----------------------|--|---------------------|
| Malaysia | 0.30-1.54 | Mahat, 2007 |
| Finland | 3.46 (128 Bqm ⁻³) | Enflo, 2002 |
| Norway | 1.97 (73 Bqm ⁻³) | Enflo, 2002 |
| France | 1.68 (62 Bqm ⁻³) | Enflo, 2002 |
| Denmark | 1.43 (53 Bqm ⁻³) | Enflo, 2002 |
| UK | 0.54 (20 Bqm ⁻³) | Enflo, 2002 |
| Canada | 0.92 (34 Bqm ⁻³) | Enflo, 2002 |
| Hungary | 1.14 (42 Bqm ⁻³) | Hamori et al., 2006 |
| USA | 1.14 (42 Bqm ⁻³) | Enflo, 2002 |
| Jordan | 1.41 (52 Bqm ⁻³) | Kullab, 2005 |
| Saudi Arabia (Riyadh) | 0.49 (18Bqm ⁻³) | Al-Saleh, 2007 |
| Japan | 0.43-5.14 (16-190 Bqm ⁻³) | Enflo, 2002 |
| Indoor Global Mean | 1.08 (40 Bqm ⁻³) | UNSCEAR, 2000 |

2.4.3 Environmental Factors Influence Rn-222

There are a few environmental factors that influence Rn-222 emanation in the environment that were humidity, temperature and pressure. Although these factors were not a major contribution to the Rn-222 amount but it slightly increases the amount of Rn-222 emanated from brick material. However from the research done by Al-Sharif & Abdelrahma (2001), Rn-222 in the house decrease when near the window but increase when far from the window.

2.4.3 (a) Humidity

Humidity was the attention of water vapour found in air. Water vapour, the gaseous state of water, was usually invisible to the human eye. Humidity indicates the probability for precipitation, dew, or fog to be present. The quantity of water vapour needed to acquire saturation will increase because the temperature increases. The temperature of a parcel of air decreases where it'll ultimately attain the saturation point without adding or dropping water mass (Groves-Kirkby et al., 2015) . From the research by Singh et al (2011), the higher the humidity in the area, the higher the Rn-222 concentration.

2.4.3 (b) Pressure

Radon gas was a naturally-occurring radioactive gas found in the soil. As it was a gas, then it continuously looking for a lower air pressure area by moving from the soil to the air. Rn-222 will find the easiest path out of the soil and into the air where it moves faster when there was a bigger difference in pressure between the high pressure soil and the low pressure air. This principle of pressure differences was the main driving force that causes Rn-222 to enter the home and also causes Rn-222 levels to change (Nazaroff, 1992).

2.4.3 (c) Temperature

Weather was one in every of the foremost common factors affecting Rn-222 levels. Changes in weather significantly extreme weather events will change the pressure variations and so change however Rn-222 enters the home. Some weather events, like storms, generally cause Rn-222 levels in an exceedingly home to rise. Primarily, weather events will cause Rn-222 levels to either rise or fall. Typically, light

weather events sort of a light downfall, low winds, gentle downfall don't dramatically have an effect on Rn-222 levels. But, serious weather events like severe storms, high winds, usually do have an effect on Rn-222 levels enormously (Iskandar et al., 2004).

2.5 Health Hazards

Radon is an odourless, tasteless and colourless natural radioactive gas that cannot be detected without special equipment. Because of that unique properties, people easily inhaled it without knowing the effect of Rn-222 gas to our lung due to the alpha particle but the possible effect was depend on exposure level. After breathed in Rn-222 gas, it was rapidly exhaled but Rn-222 progeny which is α particle combines with other molecules. In the air that contain dust, smokes will readily deposit in the airways of the lungs. While stick there, α particles emits ionizing radiation that damage the cell lining during the airways. Table 2.3 shows the number of Rn-222 attribute lung cancer deaths per year according to smoking status and gender with approximately 90% of all lung cancers occurs among smokers but never smokers are effected more by Rn-222 exposure than smokes but absolute risk was higher for smokers than for non-smokers (Kim et al., 2016).

Table 2. 3: Number of Rn-222 attribute to lung cancer deaths per year according to smoking status and gender (Kim et al., 2016)

| Country (reference) | Mean indoor radon (Bq/m ³) | Model used in risk estimation | Ever-smokers | | | Never-smokers | | | Ever- and never-smokers | | |
|---------------------|--|-----------------------------------|--------------|--------|-------|---------------|--------|-------|-------------------------|--------|-------|
| | | | Male | Female | Total | Male | Female | Total | Male | Female | Total |
| United States | | | | | | | | | | | |
| ([2], 1999) | 46 | BEIR VI, EAC | 12.5 | 13.7 | 12.9 | 25.8 | 26.9 | 26.4 | 14.1 | 15.3 | 13.9 |
| | | BEIR VI, EAD | 8.7 | 9.6 | 9.1 | 18.9 | 19.7 | 19.1 | 9.9 | 10.8 | 9.8 |
| Netherlands | | | | | | | | | | | |
| ([18], 2001) | 23 | Two-mutation carcinogenesis model | - | - | - | - | - | - | 2 | 6 | 4 |
| Sweden | | | | | | | | | | | |
| ([18], 2001) | 110 | Two-mutation carcinogenesis model | - | - | - | - | - | - | 17 | 24 | 20 |
| Canada | | | | | | | | | | | |
| ([15], 2005) | 28 | BEIR VI, EAC | - | - | 7.3 | - | - | 13.5 | - | - | 7.8 |
| ([1], 2012) | 42 | EPA model | 15.3 | 14.3 | 14.8 | 29.5 | 27.8 | 28.4 | 16 | 16 | 16 |
| ([17], 2013) | 43 | BEIR VI, EAC | - | - | 12.3 | - | - | 21.9 | - | - | 13.6 |
| France | | | | | | | | | | | |
| ([16], 2006) | 89 | BEIR VI, EAC | - | - | 11 | - | - | 50 | - | - | 13 |
| | | BEIR VI, EAD | - | - | 8 | - | - | 36 | - | - | 9 |
| | | European pooling study | - | - | - | - | - | - | - | - | 5 |
| Germany | | | | | | | | | | | |
| ([14], 2008) | 49 | European pooling study | 5.0 | 5.2 | - | 5.2 | 5.2 | - | - | - | 5.0 |
| Switzerland | | | | | | | | | | | |
| ([14], 2008) | 78 | European pooling study | 8.2 | 8.6 | - | 8.8 | 8.8 | - | - | - | 8.3 |
| United Kingdom | | | | | | | | | | | |
| ([20], 2009) | 21 | BEIR VI, EAC | - | - | - | - | - | - | - | - | 6.0 |
| | | European pooling study | - | - | - | - | - | - | - | - | 3.3 |
| Portugal | | | | | | | | | | | |
| ([21], 2012) | 81 | BEIR VI, EAC | 25 | 23 | - | 40 | 38 | - | 27 | 34 | - |
| | | BEIR VI, EAD | 18 | 17 | - | 31 | 29 | - | 20 | 27 | - |
| South Korea | | | | | | | | | | | |
| ([19], 2015) | 62 | BEIR VI, EAC | 18.6 | 18.5 | - | 33.2 | 32.8 | - | 19.5 | 28.2 | - |
| | | BEIR VI, EAD | - | - | - | - | - | - | 13.5 | 20.4 | - |
| | | European pooling study | - | - | - | - | - | - | 8.3 | 8.3 | - |

The values not presented in papers were left blank

EAC exposure-age-concentration model, EAD exposure-age-duration model, EPA Environmental Protection Agency

2.5.1 Alpha Particle

Alpha particle is a radioactive particle that is Helium atom which give out α particle when they go through radioactive decay that is known as α decay. Due to the short range of absorption and inability to penetrate the outer layers of skin, it commonly dangerous to life. Linear energy transfers mass of electron in micrometer length of cell. Rn-222 progenies were produced during Rn-222 decays. 3.8 days is too short for mass production of α particles. From the Figure 2.7, it shows that α particle breaks the “backbone” of the DNA, the beta particle breaks hydrogen bonds, and X-

rays damage bases when in fact all three types of radiation can cause all three types of direct damage. However, heavy charged particles such as α particles have a greater probability of causing direct damage compared to low charged particles such as X-rays (Desouky et al., 2015).

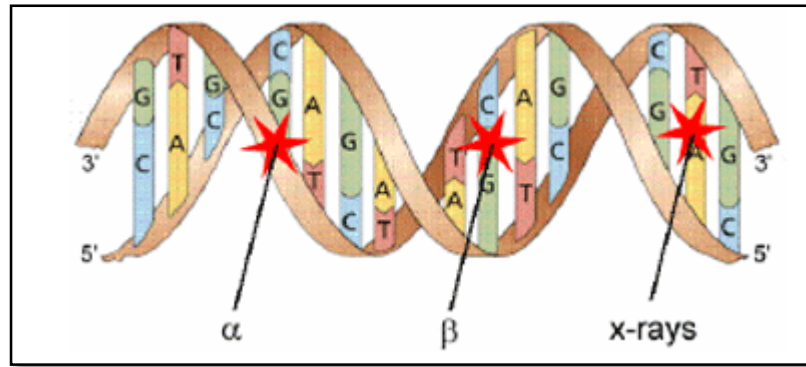


Figure 2.7: Direct action of ionizing radiation on DNA (Desouky et al., 2015)

2.5.2 Stochastic Effects

Stochastic effects are those that occur by chance and consist primarily of cancer and genetic effects. Stochastic effects often show up years after exposure. As the lung cancer is increase with increase dose, probabilities of cancer occurrence is random (Kamiya et al., 2015). Since they have no apparent threshold, the risk from the exposure increases with increasing the dose, but the severity of the effect is independent of the dose as shown in Figure 2.8. In stochastic effect, the effect rate increases with increase in the dose rate.

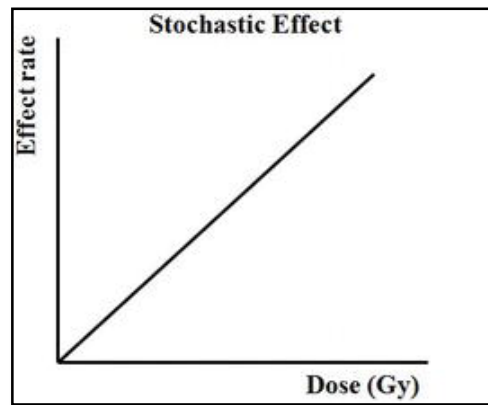


Figure 2.8: Stochastic effect graph

2.6 Building Bricks

Most of the buildings in Malaysia are made up from concrete brick. There were a few type of bricks that are used according to their purposes such as common burnt clays brick used in general work, soda lime bricks offer excellent load bearing capacity, concrete brick provide excellent aesthetic presence and fly ash clay bricks may expand when they come in contact with moisture. Cement, sand and gravel are commonly the main raw materials in producing a brick but when there was another materials replace in the concrete, the physical and mechanical properties of the bricks will be enhance such as it density, drying shrinkage, water absorption characteristic, compressive and also it flexural strength (Ismail & Yaacob, 2010). Conventional brick that used cement as a concrete may contain high embodied energy and large carbon footprint. According to Zhang (2013), there are three methods to produce bricks from waste materials include firing, cementing and geopolymerization. The cement content, water content, aggregate grading and other physical characteristics was generally influenced the workability of concrete. Moreover brick aggregate replacement exhibit many benefit such as reduction in weight, improve absorbent properties and reduced damage compared to normal brick (Sasah & Kankam, 2017). However, there was an