

MONTE CARLO SIMULATION OF ELECTRON BEAM IRRADIATION SYSTEM FOR NATURAL RUBBER VULCANIZATION

K. Kosaentor^{1,2}, E. Kongmon^{1,2}, J.Saisut¹, C. Thongbai¹, S. Rimjaem^{1,†}

¹Plasma and Beam Physics Research Facility, Department of Physics and Materials Science, Faculty of Science, Chiang Mai University, Chiang Mai 50200, Thailand

²The Graduate School, Chiang Mai University, Chiang Mai 50200, Thailand

Abstract

This paper presents the results of Monte Carlo simulation of electron beam irradiation system for natural rubber vulcanization, which is underway at Chiang Mai University in Thailand. The accelerator system can produce electron beams with adjustable energy and current in the ranges of 0.5-4 MeV and 10-100 mA, respectively. The electron beam exits from vacuum environment in the accelerator to the atmospheric air through a titanium (Ti) window. The electron dose absorption in Ti window and air was calculated by using the program GEANT4. The simulation results show that 50 μm Ti foil causes the energy loss of 1 and 18% for the beam of 4.0 and 0.5 MeV, respectively. The air gap between vacuum window and rubber surface is adjustable from 180 mm to 540 mm. The total beam energy loss of around 8-17% and 1-3% from the initial energies of 0.5 and 4 MeV, respectively. The proper depth of the natural rubber for the vulcanization process is 0.13 to 1.68 cm with the surface dose of 5.32 kGy for 0.5 MeV electron beam and 3.34 kGy for 4.0 MeV electron beam at the pulse repetition rate of 200 Hz. Accordingly, the treatment time of around 10-15 second per irradiated point is required.

INTRODUCTION

Natural rubber is an importance export product of Thailand and Southeast Asia countries. It is used in many applications and products. However, it becomes sticky and deforms easily at high temperature and brittle as cold. In order to improve the natural rubber properties, the suitable vulcanization process is needed. Vulcanization is a chemical process in which individual rubber molecules are linked into three dimensional polymer chains. The conventional method to vulcanize the natural rubber is adding chemical substance, i.e. sulfur, at high temperature. This may cause some problems e.g. toxicity, low aging as processing in high temperature. The accelerated electron beam irradiation is another option for natural rubber vulcanization, which does not need to add toxic chemical compounds. This will increase the commercial value and reduce unwanted by-products. In addition, electron beam vulcanization process can be occurred at room temperature, which promotes lower degradation rate and higher quality rubber product [1]. This method is suitable to be used in natural rubber for producing of medical and sensitive products.

† sakhorn.rimjaem@cmu.ac.th

ELECTRON BEAM VULCANIZATION

The vulcanization by using an accelerated electron beam is based on the energy transfer from electrons to natural rubber. Free radicals are formed along the molecule chain. These free radicals are significant in the cross-linking process of polymer [2]. The throughput of this technique depends greatly on the electron beam energy and dose.

An electron beam irradiation system for natural rubber vulcanization is developed at the PBP-CMU Linac Laboratory of the Plasma and Beam Physics Research Facility (PBP), Chiang Mai University. The project aims to find the optimum processing conditions for proper natural rubber vulcanization. The overview of the accelerator and electron beam processing system was described in Ref. [3]. The expected specifications of the accelerator and electron beam are listed in Table 1.

Table 1: Expected specifications of the accelerator and beam for vulcanization of natural rubber latex.

Specification / Beam parameter	Value	Unit
Maximum RF power from magnetron	2	MW
Resonant frequency of linac at 35°C	2996.395	MHz
Beam energy	0.5-4	MeV
Pulse repetition rate of electron beam	20-385	Hz
Electron pulse duration (FWHM)	4	μs
Electron pulse current	10-100	mA
Average current at 385 pulses/s	40-154	μA
Maximum beam power	620	W

An important parameter, which is used to determine the occurrence of the cross-linking process in the rubber vulcanization, is an electron absorbed dose. It is defined as an absorbed energy (E) per mass (m) of the material, which can be written as

$$Dose [Gy] = \frac{dE [J]}{dm [kg]} \quad (1)$$

The energy deposition, D_e , is defined as the energy per unit area density per incident electron as

$$D_e [MeV \cdot cm^2 / g] = \frac{dE [MeV]}{dz [g / cm^2]} \quad (2)$$

The energy deposition and the absorbed dose can be calculated using a computer program based on the Monte Carlo simulation to find the probability of each situation that can be randomly occurred [4]. In irradiation process, absorbed dose can be calculated by using Eq. (3), where η is the efficiency factor that the materials receive energy from the initial electron beam, I is the electron beam current, s is the depth in material and v is the speed of the conveyor. This equation is appropriate for the one dimensional problem [5] and the electron dose is according written as

$$Dose [Gy] = \frac{D_e [MeV \cdot cm^2 / g] \cdot \eta \cdot I [mA]}{10 \cdot s [m] \cdot v [m / s]} \quad (3)$$

According to Ref. [6], the absorbed dose required in the crosslinking of polymer is 50-150 kGy. Characteristics of an absorbed dose distribution are shown in Figure 1. It presents the relationship between percentage absorbed dose and depth in target material. As shown in Fig. 1, D_s is the dose at the irradiated material surface, R_{opt} is the depth that electron beam dose is equal to the surface dose, R_{50} is the depth of dose at a half of maximum beam dose, R_{50e} is the depth of dose at a half of surface beam dose and R_q is the depth of the dose at the steepest tangential plotted. These distances are used to determine the irradiated thickness in material. The optimum distance for irradiation is normally selected from the distance R_{opt} , which gives better uniformity of dose along the penetration depth.

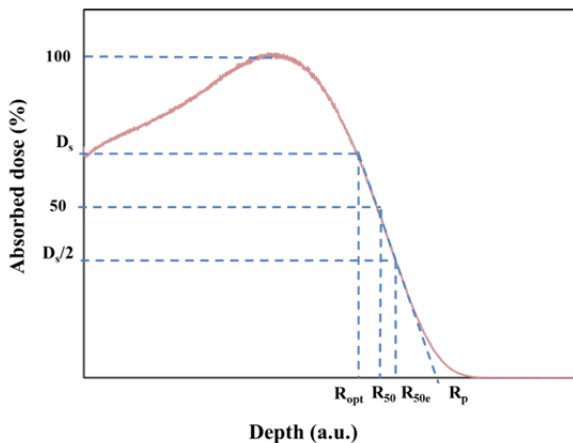


Figure 1: Absorbed dose distribution along the depth of material.

GEANT4 SIMULATION

Monte Carlo method is a reliable technique for simulation of particle transportation and interactions in matter with random particle trajectories [4]. In this research, we use the GEometry ANd Tracking (GEANT4) toolkit to study the depth of electron beam deposition in the beam exit window, in air and in the natural rubber. In GEANT4 simulations, the user needs to define their own equipment setup and input parameters by using C++ programming language [7].

A simple accelerator tube with 3.25 cm aperture and the irradiation system including titanium window, air and

natural rubber were built. To find the optimum condition of the irradiation system, the energy counting is simulated by using the scoring mesh UI command and dumps a scoring quantity in each mesh to the output file for the analysis. The scoring mesh dimensions are $100 \times 100 \times 0.004 \text{ mm}^3$ for 50 μm titanium foil, $100 \times 100 \times 2 \text{ mm}^3$ for 180 mm and 540 mm air, and $100 \times 100 \times 0.01 \text{ mm}^3$ for 30 mm natural rubber latex. The incident electron is monoenergetic point source beam with the energy varied from 0.5 to 4 MeV. The considered quantities are energy and dose deposition in the material. These data were used to study the optimum depth of electron beam in each medium. Then, the proper natural rubber thickness for the vulcanization process is defined for each beam energy.

RESULTS AND DISSCUSSION

In our simulation, the electron beam has a pulse train structure with a macropulse duration of 4 μs (FWHM). The number of macropulses per second (pulse repetition rate) can be varied from 20 to 385 pulses. Each pulse has 2996 macrobunches with a bunch charge of 300 pC. The total initial energies per macropulse are $1.12 \times 10^{13} \text{ MeV}$ and $8.99 \times 10^{13} \text{ MeV}$ for the beam energy of 0.5 MeV and 4 MeV, respectively. The simulations of electron beam energy deposition discussed in this paper are defined into 3 parts, which are titanium window, air and natural rubber latex.

Titanium Foil Vacuum Window

The vacuum window is used to separate the vacuum inside the accelerator and the air outside. In order to minimize the energy loss from this part, low atomic number material has to be used. However, it should have high strength and tolerant to work with high pressure and high temperature. Accordingly, the titanium (Ti) foil is chosen. Figure 2 shows the total electron beam energy that remains after passing through 50 μm Ti foil, 180 mm and 540 mm air gaps for different electron beam energies. After exiting the 50 μm Ti foil, the total beam energy respectively reduces by about 1% and 18% for the initial electron beam energies of 0.5 and 4 MeV.

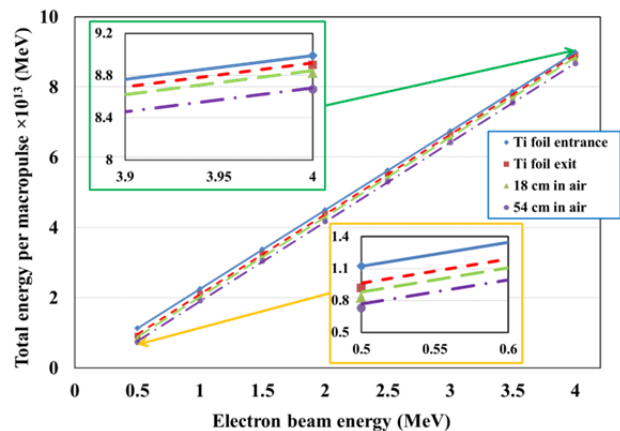


Figure 2: Total electron beam energy per macropulse at the Ti foil entrance, Ti foil exit, 18 cm and 54 cm air gaps.

Air Gap

The electron beam energies that remain after exiting the Ti window passes through the air gap before entering the rubber surface. The air gap distance depends on the adjustable height of the conveyor, which can be varied from 18 cm to 54 cm. The influence of the air gap to the electron beam energy is also shown in Fig. 2. The electron beams of 0.5 and 4 MeV loss their total energies of around 8-17% and 1-3%, respectively.

Accordingly, the 0.5 and 4 MeV beams will lose 26.3% and 1.8% of their initial energies after penetration through the Ti foil and the air gap of 18 cm. As a result, the electron beam that remains to interact with the natural rubber have a total energy of $8.28 \cdot 10^{12}$ MeV for the 0.5 MeV beam and $8.82 \cdot 10^{13}$ MeV for the 4 MeV beam.

Natural Rubber Latex

The optimization of the natural rubber thickness in this study was conducted by considering the distance R_{opt} . Figure 3 shows the electron absorbed dose per macropulse in natural rubber for different initial beam energies. The relationship of the absorbed dose and the pulse repetition rate is shown in Fig. 4. The linearity of the graph in Figure 4 will be used to calculate the total absorbed dose in the natural rubber for different pulse repetition rates.

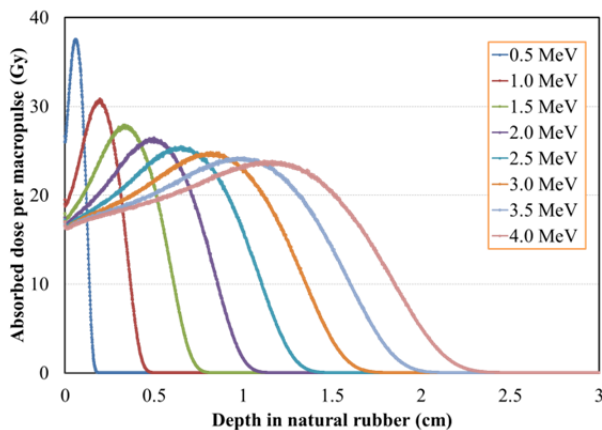


Figure 3: Simulated absorbed doses of natural rubber latex per macropulse for electron beam with energy varied from 0.5 to 4 MeV.

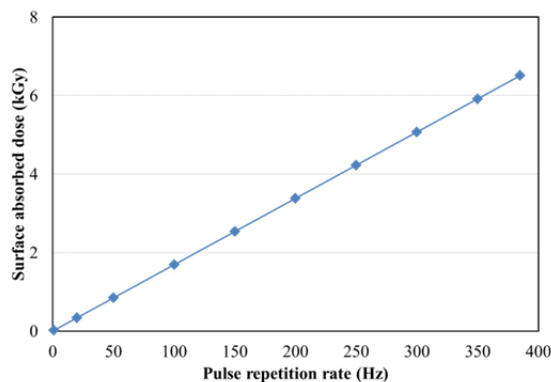


Figure 4: Simulated surface absorbed dose of 4 MeV electron beam as a function of the pulse repetition rate.

The results in Figs. 3 and 4 can be used to determine the chamber volume and the natural rubber thickness. It can be concluded that the thickness of the rubber latex should be 0.13-1.68 cm based on the electron beam initial energy. The surface absorbed dose is varied from 26.6-16.9 Gy per macropulse for the beam energy of 0.5-4 MeV. The dose value increases linearly with the pulse repetition rate of the beam. Accordingly, the irradiation with 200 pulses per second increase the surface dose to 5.3-3.4 kGy. These values lead to the processing time of about 10-15 seconds per irradiated point.

CONCLUSION

The results of GEANT4 simulation with monoenergetic initial electron beam show that before interacting with the natural rubber latex, the beam will lose their energies of 26.3% and 1.8% for the beam with initial total energy of 0.5 and 4 MeV, respectively. The optimal natural rubber thickness which is suitable for the vulcanization process is 0.13-1.68 cm depending on the electron beam energy. The maximum surface absorbed dose is 5.32 kGy for the beam irradiation with 200 pulses per second. The estimated minimum processing time is about 10 seconds per irradiated point. More study by using the beam with practical properties including the energy spread and the transverse beam size will be continued.

ACKNOWLEDGEMENTS

The authors would like to acknowledge the supports by the Science Achievement Scholarship of Thailand, the Thailand Center of Excellence in Physics (ThEP Center), the Graduate School of Chiang Mai University and the Science and Technology Park Chiang Mai University (CMU STeP).

REFERENCES

- [1] E. Manaila, G. Cracium, M.D. Stelescy, "Aspects regarding radiation crosslinking of elastomers", INTECH Open Access Publisher, 2012.
- [2] A.J. Berejka, M.R. Cleland, "Industrial electron beam processing", IAEA International atomic energy agency, 1 December 2010.
- [3] S. Rimjaem *et al.*, "Electron linear accelerator system for natural rubber vulcanization", *Nuclear Instruments and Methods in Physics Research B*, Articles in Press. Available online: <http://dx.doi.org/10.1016/j.nimb.2016.11.016>.
- [4] K. Prarodi, "Monte Carlo methods for dose calculation", *Ion beam therapy fundamentals, technology, clinical application*, U Linz, NY, USA: Springer, 2011, pp. 97-116.
- [5] International atomic energy agency (IAEA), Use of mathematical modeling in electron beam processing, 2010, ISBN 978-92-0-112010-6.
- [6] M.R. Cleland, "Industrial applications of electron accelerators", CERN Accelerator School, Zeegse, Netherlands 24 May to 2 June, 2005.
- [7] Geant4 Collaboration, "GEANT4 user's guide for application developers", Geant4 version 10.2, 4 December 2015.