# Investigation on Electron Absorbed Dose in a Mixture of Natural Rubber Latex and Cross-linking Agents

P Nanthanasit<sup>1,2</sup>, P Jaikaew<sup>1</sup>, E Kongmon<sup>1,3</sup> and S Rimjaem<sup>1,4,5,\*</sup>

<sup>1</sup>Plasma and Beam Physics Research Facility, Department of Physics and Materials Science, Faculty of Science, Chiang Mai University, Chiang Mai 50200, Thailand <sup>2</sup>Master Degree Program in Physics, Department of Physics and Materials Science, Faculty of Science, Chiang Mai University, Chiang Mai 50200, Thailand <sup>3</sup>Master Degree Program in Applied Physics, Department of Physics and Materials Science, Faculty of Science, Chiang Mai University, Chiang Mai 50200, Thailand <sup>4</sup>Thailand Center of Excellence in Physics, Commission on Higher Education, Bangkok 10400, Thailand

<sup>5</sup>Research Center in Physics and Astronomy, Chiang Mai University, Chiang Mai 50200, Thailand

\*E-mail: sakhorn.rimjaem@cmu.ac.th, panat\_14@hotmail.com

Abstract. Natural rubber vulcanization using electron beam is one of interesting applications of electron accelerators with an energy in a range of a few hundred keV to a few MeV. At the Plasma and Beam Physics (PBP) Research Facility, Chiang Mai University, a 4-MeV radiofrequency (RF) linear accelerator is developed for utilizing in natural rubber vulcanization. To accelerate the vulcanization process, cross-linking agents are added in a natural rubber latex to increase the bonding probability between the chains of rubber molecules. This research used a Monte Carlo based program GEANT4 to simulate the electron absorbed dose in a mixture of natural rubber latex and cross-linking agents. The initial electron energy distributions were obtained from the beam dynamic simulation in the accelerator, which were done with program ASTRA. The initial electron beams have average energy in the range of 0.70 to 4.02 MeV at the position 6 cm prior the 50-µm titanium vacuum window foil. The air gap between the vacuum window to the natural rubber latex was set at 18 cm. The electron beam transportation in the titanium foil, air gap and a mixture of natural rubber latex were simulated. The cross-linking agents that are added in the natural rubber latex are ethylene glycol dimethacrylate (EDMA) and 1,6-hexanediol-diacrylate (HDDA). Finally, electron absorbed dose distributions in both transverse direction and along the depth of the natural rubber latex mixed with each cross-linking agent were calculated and analyzed. The results from this work can be used for experimental planning of electron beam irradiation on the natural rubber latex with appropriate cross-linking agents and electron absorbed dose.

#### **1. Introduction**

Natural rubber is one of the most important export products of Thailand. It is originally derived from the milky colloid latex produced by a rubber tree with the scientific name "Hevea brasiliensis". The natural rubber consists mainly of polymer chains of isoprene, which are called polyisoprene. The important reaction for the natural rubber is vulcanization. This reaction is a chemical process to transform the natural rubber latex into a durable material. Generally, sulfur is widely used for vulcanizing the natural rubber latex. However, heating process is necessary during the vulcanization with sulfur. This condition can lead to degradation problem of the rubber product when it is in high temperature environment again after the forming process [1]. This differs from vulcanization with electron beam, which can happen at atmospheric pressure and room temperature. Moreover, this process can break molecule chains of protein that may cause allergy and there is no any toxic waste produced during the process.

When the electron beam passing through the natural rubber latex, the accelerated electrons with enough kinetic energy will break the chemical bonds between carbon and hydrogen in the polyisoprene. This results in the generation of free radicals and bonding between the chains of the radical polyisoprene [2]. Furthermore, cross-linking agents are added in the rubber latex prior the electron beam irradiation. Molecules of the crosslinking agents can be bonded between the radical polyisoprene that can increase the probability of vulcanization. This research focuses on simulation of electron beam in a mixture of natural rubber latex and cross-linking agents. The varied electron beams are produced by a 4-MeV RF linear accelerator, which is developed at the PBP-CMU Electron Linac Laboratory of the Plasma and Beam Physics Research Facility, Chiang Mai University.

### 2. Simulation setup

A Monte Carlo based program GEANT4 [3] was used to simulate the electron beam in the irradiation system. Initial electron beam distributions at the position 6 cm prior the 50-um titanium vacuum window foil were obtained from the beam dynamic simulation with the program ASTRA [4]. They have mean energies from 0.70 to 4.02 MeV with energy step of about 0.5 MeV. Our accelerator can produce electron beam with 4-µs macro-pulses, which contain 2998 micro-bunches per microsecond. Hence, there are about 12,000 micro-bunches in one macro-pulse. The beam dynamic simulation result suggested that each micro-bunch has  $2.061 \times 10^8$  electrons. It is noted that the number of electron macropulses per second can be varied from 10 to 200 pulses. Thus, the number of electrons can be varied from  $2.061 \times 10^9$  to  $4.122 \times 10^{10}$  electrons. The large number of electrons needs an enormous computer memory and time for simulation. Optimization of number of electrons for reliable simulation results with reasonable simulation load and time was done [5]. It was found that the simulations with number of electrons equal or greater than 10<sup>5</sup> particles provided good resolution of simulation results. The simulated values such as total kinetic energy, deposited energy and absorbed dose for more electron number can then be scaled to the results from simulation with  $10^5$  electrons. Due to limitation of our computer capability, we used this optimal number of electrons (10<sup>5</sup> particles) in all GEANT4 simulations. Examples of energy spectrum for initial electron beam distributions with low energy (1.51 MeV) and high energy (4.02 MeV) are shown in fig. 1.



**Figure 1.** Examples of energy spectra for initial electron beam distributions with average energy of (a) 1.51 MeV and (b) 4.02 MeV.



**Figure 2.** The geometry layout of the electron beam irradiation system generated by program GEANT4.

The geometry layout of this simulated system, which was generated by the program GEANT4, is illustrated in fig. 2. The air gap between the titanium window and the natural rubber latex was set at 18 cm and the volume of the natural rubber latex was 5 cm  $\times$  5 cm  $\times$  3 cm.

#### 3. Simulation of electron beam total energy from the accelerator to the natural rubber latex

The total kinetic energy of electron beams along the longitudinal axis (*z*-axis) from the linac to the natural rubber latex are calculated and two examples' results are shown in fig. 3. The energy presented in fig. 3 is already scaled to the total kinetic energy of electrons in a macro-pulse. From these graphs, the total kinetic energies are quite constant in the vacuum (z = 0 to z = 6 cm) and they are suddenly decreased when the electron beams pass through the titanium vacuum window (z = 6 cm). Then, the total kinetic energies are decreased continuously in the air gap between the titanium foil and the natural rubber latex (z = 6 cm to z = 24 cm). The total kinetic energies at the natural rubber latex for the initial electron beams with average energies of 1.51 MeV and 4.02 MeV are 97.3% and 98.8% of their initial values, respectively. In addition, percentages of the total kinetic energy are presented in Table 1.



**Figure 3.** Total kinetic energies of electrons per macro-pulse along the *z*-axis from the accelerator to the natural rubber latex for the initial electron beams with average energy of (a) 1.51 MeV and (b) 4.02 MeV.

Average kinetic energy of initial electron beam (MeV)	Percentage of total kinetic energy at the natural rubber latex surface compared to the total kinetic energy of initial electron beam (%)
0.70	93.8
1.03	92.6
1.51	97.3
1.96	97.8
2.50	98.2
3.03	98.5
3.51	98.7
4.02	98.8

**Table 1.** Percentages of the total kinetic energy of electron beam at the natural rubber latex surface compared to the total kinetic energy of initial electron beam for each average energy.

## 4. Simulation of electron dose in a mixture of natural rubber latex and cross-linking agents

Absorbed dose is the quantity of radiation energy deposited in absorber material per mass. Results of electron absorbed doses per macro-pulse along the depth of the natural rubber latex for different electron beam average energies are presented in fig. 4. The results show that the higher electron beam energies, the larger electron absorbed dose and the deeper beam penetration.



**Figure 4.** Absorbed doses per macro-pulse in natural rubber latex for initial electron beams with different average energies from 0.70 to 4.02 MeV.



**Figure 5.** Absorbed doses per macro-pulse in a mixture of natural rubber latex and crosslinking agents for electron beam with the average kinetic energy of 4.02 MeV.

Furthermore, cross-linking agents, which are ethylene glycol dimethacrylate (EDMA) [6] and 1,6hexanediol-diacrylate (HDDA) [7], are added in the natural rubber latex (NR) with an amount of 12 phr (parts per hundred rubber). Simulation results for electron beam with maximum average kinetic energy of 4.02 MeV in fig. 5 show that electron absorbed doses in the mixture of natural rubber latex and each cross-linking agent are not significantly different compared to that in the pure natural rubber latex. This is because the Monte Carlo simulation in GEANT4 is only based on the composition and density of material. Addition of small amount of the cross-linking agents does not significantly change the density of the rubber latex. Their influences on natural rubber cross-linking efficiency are only based on the bonding between the radical polyisoprene and their molecules. Examples of transverse distributions of the electron absorbed dose for the pure natural rubber latex and the mixtures with cross-linking agents are presented in fig. 6. The results show that the maximum absorbed dose at the depth of 1.2 cm is about 32% higher than that at the rubber surface.



**Figure 6.** Transverse distributions of absorbed doses per macro-pulse in (a) NR, (b) NR+EDMA and (c) NR+HDDA for 4.02 MeV electron beam at the rubber surface and at the depth of 1.2 cm.

# 5. Conclusion

Monte Carlo simulations with program GEANT4 were performed to study transportation of electron beams from the accelerator to the natural rubber latex. The results suggest that the total kinetic energy at the natural rubber latex surface is 98.8% of the initial kinetic energy for the electron beam with the maximum average energy of 4.02 MeV. The electron absorbed dose distributions in both transverse directions and along the depth of natural rubber latex were investigated. Even though the cross-linking agents EDMA and HDDA are added in the natural rubber latex, the absorbed doses are not significantly changed. In addition, the transverse dose distributions are not uniform, which can cause the non-uniform cross-linking rate in the rubber latex. Therefore, the flattening filter prior the rubber latex surface is necessary to enlarge and modify the transverse distribution of electron absorbed dose to be more uniform.

### 6. Acknowledgements

We would like to acknowledge the support from Chiang Mai University, the Thailand Center of Excellence in Physics, the Science and Technology Park Chiang Mai University (CMU STeP) and the Development and Promotion of Science and Technology Talents projects (DPST).

## References

- [1] Manaila E et al 2012 Aspects Regarding Radiation Crosslinking of Elastomers *INTECH Open Access Publisher*
- [2] Lapin S C 2015 Modification of Polymer Substrates Using Electron Beam Induced Graft Copolymerization UV+EB Technology 1 pp 44-49
- [3] Agostinelli S et al 2003 Geant4-a simulation toolkit Nucl. Instrum. Methods Phys. Res. A 506 250
- [4] Flottmann K ASTRA Particle Tracking Code < http://www.desy.de/~mpyflo >
- [5] Kosaentor K 2018 Simulation and Design of Electron Beam Irradiation System for Natural Rubber Vulcanization *Master thesis* Chiang Mai University
- [6] Manaila E et al 2014 Radiation Vulcanization of Natural Rubber with Polyfunctional Monomers *Polymer Bulletin* **71** pp 57-82
- [7] Senna M M H and Monem Y K A 2010 Effect of Beam Irradiation and Reactive Compatibilizers on Some Properties of Polypropylene and Epoxidized Natural Rubber Blends *Journal of Elastomers and Plastics* 42 pp 275-295