e-Jurnal Sains Nuklear Malaysia, 2022, Vol. 34: 22 – 28
eISSN: 2232-0946

EFFECT OF POST-GAMMA RADIATION ON MECHANICAL PROPERTIES OF HYBRID GAMMA-PEROXIDE VULCANIZED NATURAL RUBBER LATEX FILMS

Sofian Ibrahim[1], Hasan Sham[1], Chantara Thevy Ratnam[1],
Khairiah Badri[2] and Noor Hasni M. Ali[1]

[1]Malaysian Nuclear Agency, 43000 Kajang, Selangor, Malaysia
[2]School of Chemical Science and Food Technology, Faculty of Science and Technology,
Universiti Kebangsaan Malaysia, 43600 UKM Bangi, Selangor, Malaysia
[*]Corresponding author: sofian_ibrahim@nm.gov.my

ABSTRACT

One of the common sterilization method used in Malaysia is gamma radiation because it is the fastest, easiest and the most importanty it is a cold process. However, there is one major drawback to this method because gamma sterilization requires relatively high irradiation dose; minimum of 25 kGy as required by ISO 11137 standard. This high sterilization dose is known of it capability to break chemical bonds in a medical devices and caused its mechanical properties (tensile strenght) to decline. In this study, hybrid gamma-peroxide vulcanized natural rubber latex (HGPVL) films were post-gamma irradiated at 25 and 35 kGy to investigate the extent of damage or degradation when the samples were exposed to extreme irradiation dose. At 25 and 35 kGy post-gamma irradiation, tensile strenght of HGPVL films were found to decrease at 8.0 % and 15 % respectively compared to control (26.5 MPa) due to degradation of their polymeric network.

Keywords: gamma, sterilization, latex.

INTRODUCTION

Preparation of prevulcanized natural rubber latex via hybrid gamma –peroxide vulcanization has successfully produced prevulcanized latex with good mechanical and physical properties which has the potential to be a key ingredient in the production of premium latex dipped products such as surgical glove, condom and etc. (Sofian et al. 2018a, Sofian et al. 2018b, Sofian et al. 2018c). However, the durability of hybrid gamma-peroxide vulcanized latex films against post-gamma irradiation should be studied if it is to be used to produce surgical glove. This is because every medical products or devices has to be sterilized at minimum of 25 kiloGray (kGy) before being marketed to consumers as required by ISO 13485: 2016 and ISO 11137: 2017 standards.

Nowadays there are several methods of sterilization of medical devices. The most commonly used sterilization method in Malaysia are ethylene oxide gas (EtO) and gamma radiation. Between the two methods, gamma irradiation sterilization is a cold process, fastest and easiest. However, there is one major drawback to this method; gamma sterilization requires relatively high radiation dose as required by ISO 11137 (2017) standard and this high dose sterilization are capable to break chemical bonds (crosslinking) in a medical devices and caused its mechanical properties (tensile strength, modulus at 500 % and modulus at 700 %) to decline (Anne 2012).

In this study, two post-gamma radiation dose parameters were selected at 25 kGy and 35 kGy. As previously described, the ISO 11137: 2017 standard has set the minimum radiation dose to sterilized medical devices at minimum 25 kGy. However, in this study another parameter of radiation dose which is 35 kGy were also been choosed. The reason why we choosed this parameter because during the sterilization process, each set of medical devices (i.e. gloves) will receive different doses even if they are placed in the same radiation container. For example, each medical device in the same container can received a various radiation doses between 25 to 29 kGy even though the radiation machine has been set for minimum 25 kGy radiation parameter. This phenomenon is known as dose distribution and occur due to the design of a radiation plant itself (Ruzalina et al. 2014). So the objective of this study is to examine the effects on the mechanical and physical properties of the hybrid gamma-peroxide vulcanized natural rubber latex (HGPVL) films when exposed to extreme radiation doses.

MATERIALS AND METHODS

Materials

The latex utilized in this work were a high ammonia type of latex (HA latex) supplied by Revertex (M) Pt. Ltd., Malaysia. The sensitizer and co-sensitizer used were hexanediol diacrylate (HDDA) supplied by Allnex, China and tert-butyl hydroperoxide (*t*-BHPO) supplied by Fluka, Switzerland respectively. The stabilizer used was potassium laurate supplied by Tiarco Chemical (M) Pt. Ltd., Malaysia and the antioxidant used was Aquanox Lp supplied by Aquaspersion (M) Pt. Ltd., Malaysia. These materials were used as received.

Methods

Preparation of hybrid gamma-peroxide vulcanized natural rubber latex films

One kilogram of hybrid gamma-peroxide vulcanized natural rubber latex (HGPVL) were prepared based on compounding formulation in Table 1 and then send to MINTec-Sinagama Plant, Malaysian Nuclear Agency to be irradiated at 6 kGy (Current activities of Co-60 are 447000 Curie with dose rate of 2.08 kGy/hr). Then the HGPVL was formed into nine latex films by coagulant dipping method (Sofian et al. 2018b).

Table 1: Compounding formulation of HGPVL

Materials	Part perhundred rubber (phr)
NR Latex (62% Total Solid Content)	100
Stabilizer	0.06
HDDA	2.50
t-BHPO	0.10
Antioxidant	2.50
Water	Add to 52% Total Solid Content

Irradiation of hybrid gamma-peroxide vulcanized natural rubber latex films at high irradiation doses

Nine latex films that was prepared earlier were divided to three set of latex films (each set consists of 3 latex films). Each set of the latex films then undergoing post-gamma irradiation at 0, 25 and 35 kGy respectively. Upon completion of post-gamma irradiation, mechanical properties (tensile strenght, modulus at 500 % elongation (M500) and modulus at 700 % elongation (M700)) and gel content (extraction of samples in toluene for 8 hours using Soxhlet apparatus) of the latex films were measured (ASTM D3616-95.2014; ASTM D412. 2016; Jayasuria et al. 2001).

RESULTS AND DISCUSSION

Post-gamma radiation on mechanical properties of hybrid gamma-peroxide vulcanized natural rubber latex films

In 1974, Wilson has conducted a study on the chemical properties of polymers exposed to radiation. The study was originally intended to find a plastic material that can withstand ionizing radiation from nuclear reactor and by coincidence he discovered something more interesting. From the studied, he found out that ionizing radiation not only caused degradation to some plastics but also capable to make some plastics become stronger after exposure to the radiation. This indicates that some polymeric materials may undergo crosslinking when exposed to ionizing radiation (Wilson 1974). Generally, crosslinking and degradation reactions occur simultaneously but at different rates (Fig. 1). A polymer material becomes stronger under ionizing radiation when the crosslinking rate exceeds its degradation and weakens on the contrary. The crosslinking and degradation ratios are different for different types of polymers and depend on the radiation conditions (Makuuchi 2003).

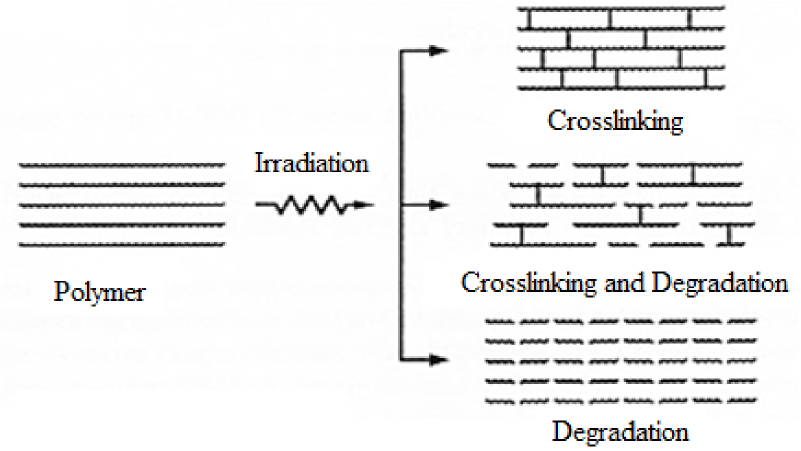

Figure 1: Effect of crosslinking and degradation on polymers by ionizing radiation

Base on this study, HGPVL films that underwent post-gamma radiation at high radiation doses did not show a significant change in their physical (colour) appearance compared to control as shown in Figure 2. Besides, it was found that the smell of all the films not signifantly different compared to control. This proves that the use of water-based Aquanox Lp antioxidant are better than oil-based antioxidants such as Irganox 1520 which often produced burning odors to the films (Sofian et al. 2015).

Figure 2: Effect of post- radiation doses on physical of HGPVL films

Studies on mechanical properties of HGPVL films irradiated at high doses (above 25 kGy) were performed according to the ISO 11137: 2006 and ISO 13485: 2003 standards requirement. This is very important since modulus and tensile strength values are considered as the commercial importance parameter, typically in gloves production. Modulus values are always referred as the degree of crosslinking in the films, whilst tensile strength value is referred to the extent of the film undergo stress [Roslim et. al., 2015].

Tensile strength, M500 and M700 of the films that were post-gamma irradiated at 0 kGy (control), 25 kGy and 35 kGy were measured. The tensile strength obtained is compared to the tensile strength of the control film to determine the high dose effect on its mechanical properties.

The results of tensile strength, modulus at 500 % and modulus at 700 % for post-gamma radiation of HGPVL are shown in Table 2. Based on the experimental results, it was found that post-irradiated HGPVL films at high doses showed a deterioration in the tensile strenght, M500 and M700 . Film at 25 kGy post-irradiated showed decreasing of tensile strength, M500 and M700 at 7.7 %, 34.5 % and 35.8 % respectively compared to control. Although there is a slight decrease of tensile strength (24.4 mPa), it is still complies to the requirements of ASTM D3577-19 (2019).

Table 2: Mechanical properties of post-irradiated HGPVL films

Radiation dose	Mechanical properties		
	Tensile strenght, mPa	Modulus at 500 %, mPa	Modulus at 700 %, mPa
0 (Control)	26.5	4.1	13.3
25	24.4	2.7	8.5
35	22.6	2.5	7.7

Post-gamma irradiated HGPVL films at extreme dose of 35 kGy was conducted in order to study it's mechanical properties effect on the films. Based on the tensile strength, modulus at 500 % and modulus at 700 % results in Table 2, it shown that more significant degradation of the film compared to 25 kGy post-gamma irradiation. The tensile strength, modulus at 500 % and modulus at 700 % of the film decreased by 14.5 %, 38.2 % and 41.9 % respectively compared to control. The deterioration on tensile strength down to 22.6 mPa resulted the HGPVL failed to comply with the ASTM D3577-01a standard.

The degradation of the mechanical properties of the films that undergone post-gamma radiation are due to reduction of crosslink density caused by the chain scission mechanism that occurs when films are expose to high irradiation dose (Makuuci 2003; Wicha et al. 2020). This finding can be support by observing the deterioration on the modulus properties of the post-gamma radiation films. Further evidence to support the finding has been collected from the gel content analysis.

Effect of irradiation doses on gel content of post-irradiated HGPVL films

The crosslink or gelled polymers only swell up and not dissolve in any solvent, in other words it's defined as that percent of a sample that does not dissolve in toluene (ASTM D3616-95, 2014; Jayasuria 2001). Figure 3 shows the relationship between gel content and post-gamma irradiated doses. The gel content of HGPVL films declined with the increasing of post-gamma irradiated dose indicating degradation in crosslink density of the latex films. As discussed earlier, the tensile strength is closely related to the crosslink density of the latex film. The decreasing of crosslink density when the films were exposed to extreme irradiation dose also caused a decrease in the tensile strength of the film (Table 2).

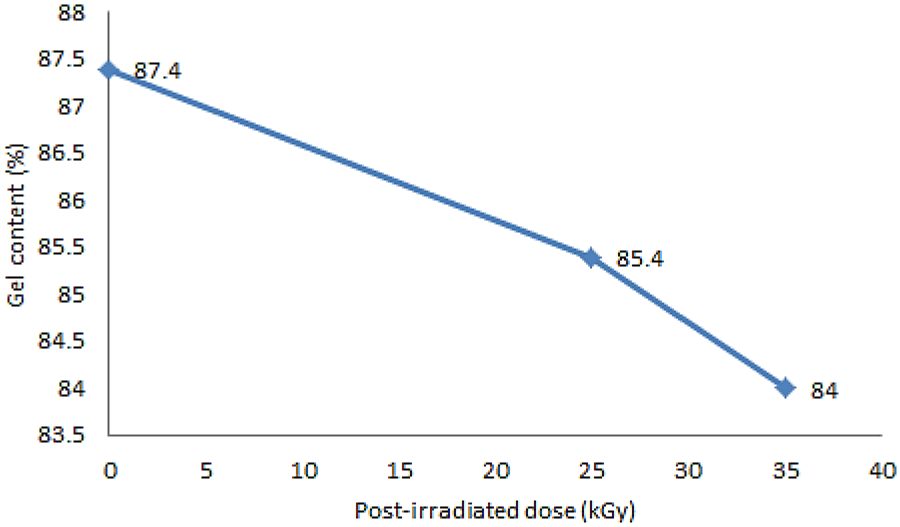

Figure 3: Effect of post-irradiation doses on gel content of post-irradiated HGPVL films

CONCLUSIONS

Post-gamma radiation at 25 kGy for sterilization purposed on HGPVL film was found to cause 8.0 % decrement of tensile strenght and 2.3 % of gel content compared to the control and still comply with the ASTM D3577-01a requirements. As for the HGPVL film that subjected to extreme post-gamma radiation (35 kGy), the tensile strenght decreased significantly (almost 15 %) due to degradation of their crosslinking and therefore failed to comply to ASTM D3577. However, from this studies it showed that HGPVL has the potential to be used in the production of premium latex dipped products such as surgical gloves.

ACKNOWLEDGEMENTS

The authors would like to express their deepest appreciation to Revertex (M) Sdn. Bhd., Malaysia for supplying HA latex to be used in this research and RAYMINTEX Plant, Malaysian Nuclear Agency for financial and technical supports of this work.

REFERENCES

11137-3:2017, ISO. (2017). *Sterilization of health care products-radiation-part 3: Guidance on dosimetric aspects of development, validation and routine control.* Geneva, Switzerland: International Organization for Standardization ISO Central Secretariat.

13485:2016, ISO. (2016). *Medical devices Quality management systems.* Geneva, Switzerland: International Organization for Standardization ISO Central Secretariat.

Anne, S. (2012). *Sterilisation of Biomaterials and Medical Devices.* Woodhead Publishing Limited.

D3577-19, ASTM. (2019). *ASTM International, West Conshohocken, PA.*

D3577-19, ASTM. (2019). *Standard Specification for Rubber Surgical Gloves.* West Conshohocken, PA: ASTM International.

D3616-95, ASTM. (2014). *Standard Test Method for Rubber—Determination of Gel, Swelling Index, and Dilute Solution Viscosity.* West Conshohocken, PA: ASTM International.

D412, ASTM. (2016). *Standard Test Methods for Vulcanized Rubber and Thermoplastic Elastomers—Tension.* West Conshohocken, PA: ASTM International.

Jayasuria, M. M., K. & Yoshi, F. Makuuchi. (2001). Radiation vulcanization of natural rubber latex using TMPTA and PEA. *European Polymer Journal* 37: 93-98.

Makuuchi, K. (2003). *An Introduction to Radiation Vulcanization of Natural Rubber Latex.* Bangkok T.R.I. Global Co., Ltd.

Roslim, R., Jefri, J., Manroshan Singh, J. S., Siti Noor Suzila, M. U. H. & Amir Hashim, M. Y. (2015). *Characterizing Physical Properties of Peroxide Vulcanized Natural Rubber Latex Films.* Trans Tech Publications Ltd. Switzerland.

Ruzalina, B., S. Hasan, A. W. Ahsanulkhaliqin, K. Jamilah, G. Cosmos, M. N. Zarina, dan A. & Sofian, I. Mohd Khairul. (2014). Establishment of dose correlation during dose mapping on medical devices product. *Paper Presented at Seminar R&D Agensi Nuklear,Bangi, Selangor, Malaysia.*

Sofian, I., Khairiah, B., Chantara, T. R. & Hasni, M. A. (2018c). Tensile properties of pre-vulcanised natural rubber latex films via hybrid radiation and peroxide vulcanisations. *ASM Sci. J.* 11(2): 67-75.

Sofian, I., Khairiah, Badri., Chantara, T. R., & Noor Hasni, M. A. (2018b). Enhancing Mechanical Properties of Prevulcanized Natural Rubber Latex via Hybrid Radiation and Peroxidation Vulcanizations at Various Irradiation Doses. *Radiation Effects and Defects in Solids* Vol.173 Issue 5-6 pg. 427-434.

Sofian, I., Khairiah, Badri., Chantara, T. R., Noor Hasni, M. A., Chai, C. K. & Mohd Noorwadi, M. L. (2018a). Preliminary Study on Peroxide Vulcanization with Gamma Irradiation. *Jurnal Sains Nuklear Malaysia* 30(2): 39–46.

Sofian, I., Mohd Noorwadi, M. L., Syuhada, R., Chai, C. K., Muhammad, K. M. Y., Saiful, O., Najib, M. Z., Hafizuddin, M. & Hasni, M. A. (2015). *Effect of dose rate on radiation prevulcanized natural rubber latex tensile strength and physical properties.* Volume 47, 2015. Report no, INIS-MY-2016-042, International Nuclear Information System (INIS).

Wicha, G., Intharaprasit, K., Wimolmala, E., Markpin, T. & Saenboonruang, K. (2020). Effects of post-gamma irradiation on swelling and mechanical properties of gamma vulcanized natural rubber latex (GVNRL) films. *IOP Conf. Series: Materials Science and Engineering.* 773(2020)012010.

Wilson, J. E. (1974). *Radiation chemistry of monomers, polymers and plastics.* New York: Marcel Dekker, Inc.