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Review Article

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Mapping the scientific research on the ionizing radiation impacts on polymers (1975–2019)

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Abstract: A bibliometric approach allowed us to study the global research trend on the impact of ionizing irradiations on polymers from 1975 to 2019. The investigation revealed 1,015 publications with growing interest since 1990. The research is split into three main categories: polymer science, nuclear science technology, and chemistry physical. The three main ionizing irradiations studied in this research are gamma, electron beam, and X-ray irradiations. The impact of ionizing irradiations on polymers under gamma irradiation is the most commonly studied field with 578 publications among the 1,015 publications. Electron beam irradiation is the second most studied field followed by X-ray irradiation. Whatever the irradiation modalities, publications focus on material degradation and material improvement studies.

Keywords: ionizing irradiations, polymer, bibliometrics, gamma, e-beam, X-rays

1 Introduction

Since a long time, the field of polymer degradation took a central place in polymer science and technology. Studies and research in this field have shown a high increase either in the industry or universities. Indeed, there has been increasing demand of its applications and

the researchers have shown deep interest in the study of polymer degradation. There are different types of degradation such as thermal, mechanical, or chemical degradations (1). These degradations can be induced by ionizing irradiations. These ionizing irradiations can modify polymers by absorbing the radiations, breaking the polymer chain (1), and then inducing a loss of polymer properties. Ionizing irradiation is a mandatory process used to sterilize products by breaking the deoxyribonucleic acid (DNA) of microorganisms and killing it or inhibiting its reproduction ability (2). This process is also used in the polymer science field to obtain polymers with desired properties. For example, crosslinking induced by ionizing irradiation is used to improve the mechanical properties of polymers (3). Grafting is another method to improve polymer properties by adding functional side chains (4).

Regarding the sterilization process, ionizing irradiation is used in different areas such as in food or medical applications. For example, ionizing radiation has been widely accepted as an effective technique for reducing or eliminating pathogens in foods. Currently, gamma irradiation is the most common process to sterilize products. However, due to several disadvantages such as the price rising of ^{60}Co sources, the antipathy toward the nuclear (5) and the safety issue due to nuclear power plants, two irradiation processes are emerging: electron beam irradiation and X-ray irradiation. The electron beam irradiation is a continuous process and the source of the electron beam is an accelerator that converts electricity using a medium, i.e., a tungsten filament source (2). X-rays are photons that are generated by using an electron beam accelerator. The electron beam is converted to photons by accelerating the electrons into a high density material (i.e., tungsten, steel, or tantalum) (2). Any type of ionizing radiation can affect the performance of polymers. In order to have a global view on the polymers irradiated by ionizing irradiations research, a bibliometric approach has been used. This bibliometric approach allow to analyze the knowledge and the structure of a specific scientific research. Previously, a similar study has been conducted studying the degradation of gamma-irradiated polymers (6). The aim of this study is then to establish an overview

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of the research between 1975 and 2019 on the impact of the different ionizing irradiation processes on polymers. Expanding the research from gamma irradiation to ionizing irradiations, in general, is necessary as industries are tempted to adopt other ionizing irradiations. The work is led by studying the evolution of publications, the different countries involved in publications and by establishing a keyword network. Keyword networks will help us to identify the main polymers of interest and the main analytical tools.

2 Methodology

Data were obtained on 16 December 2019 from Web of Science (WOS) Core Collection databases from 1975 to 2019 as follows:

- The search was limited to title only. Title refers to the title of a journal article, proceeding paper, book, or book chapter. Duplications were automatically removed.
- All journals have been considered.
- The search was separated in three parts (i.e., #1, #2, and #3):
 - #1: ethylene, polyethylene terephthalate (PET), propylene, silicon*, poly*, ethylene propylene diene monomer (EPDM), polystyrene-b-poly(ethylene-butylene)-b-polystyrene, poly styrene-butadiene-styrene, poly(terephthalate de butylene), thermoplastic elastomer, elastomer*, rubber*, polyvinyl chloride (PVC), polyamide, cellulose, poly tetrafluoroethylene (PTFE), polyetheretherketone, ethylene vinyl alcohol, acrylonitrile butadiene styrene, polyoxymethylene, perfluoroalkoxy, polychlorotrifluoroethylene, polyvinyl fluoride, polyvinylidene fluoride (PVDF), ethylene tetrafluoroethylene, fluorinated ethylene propylene, “styrenic block copolymer”
 - #2: “gamma irradi*,” “electron beam irradi*,” “e beam irradi*,” “x ray irradi*,” “ionizing rad*,” “ionizing irradi*”
 - #3: change*, degradation*, modification*, effect*
- The three sets were then crossed using the connector “and.”
- The asterisk allows us to consider the word with different writing.
- Outputs were the authors, the journal publication, the country of universities, the institutions, and keywords.

Finally, data were exported as text file and analyzed with Mateo Analyzer software, version: 4.2 150720.

Only publications from WOS Core Collection are collected and mentioned in this article. Some organizations such as International Atomic Energy Agency have their own collection (International Nuclear System Information).

Moreover, WOS has mainly publications written in English. All papers published in various journals but not recorded in WOS are not considered in this study. Existing bibliography may be delivered through conferences (7,8) and cannot be taken into account in the present survey.

3 Findings and discussion

3.1 Evolution of the impact of ionizing irradiations on polymer research overtime

During the past 44 years, the scientific literature on the impact of ionizing irradiations on polymers has grown (Figure 1). A total of 1,015 publications have been published during this period. Two periods can be observed: the first part (1975–1990) is represented as the first period of the research by showing a constant number of papers published with an average of 11 papers per year. The second part (1991–2019) shows a growing interest with an increase of the publications every year.

3.2 Domains of research interest about the ionizing radiation impact on polymers

All 1,015 publications are gathered in different categories. These categories are defined by WOS Core Collection and

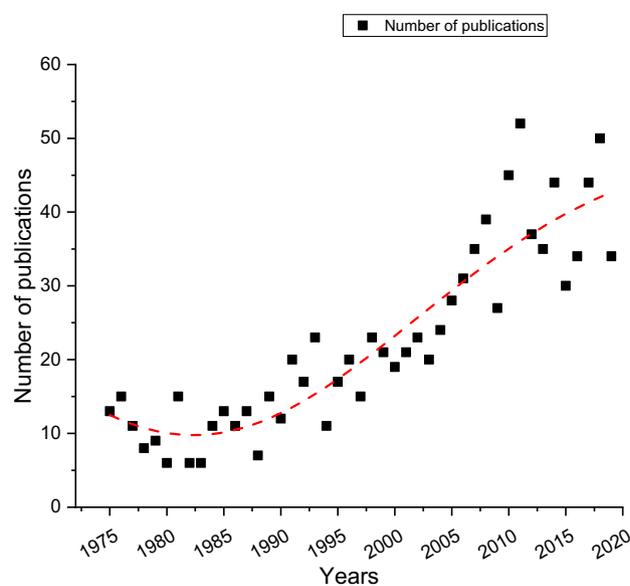


Figure 1: Number of publications overtime. The dashed line is given to ease the reading.

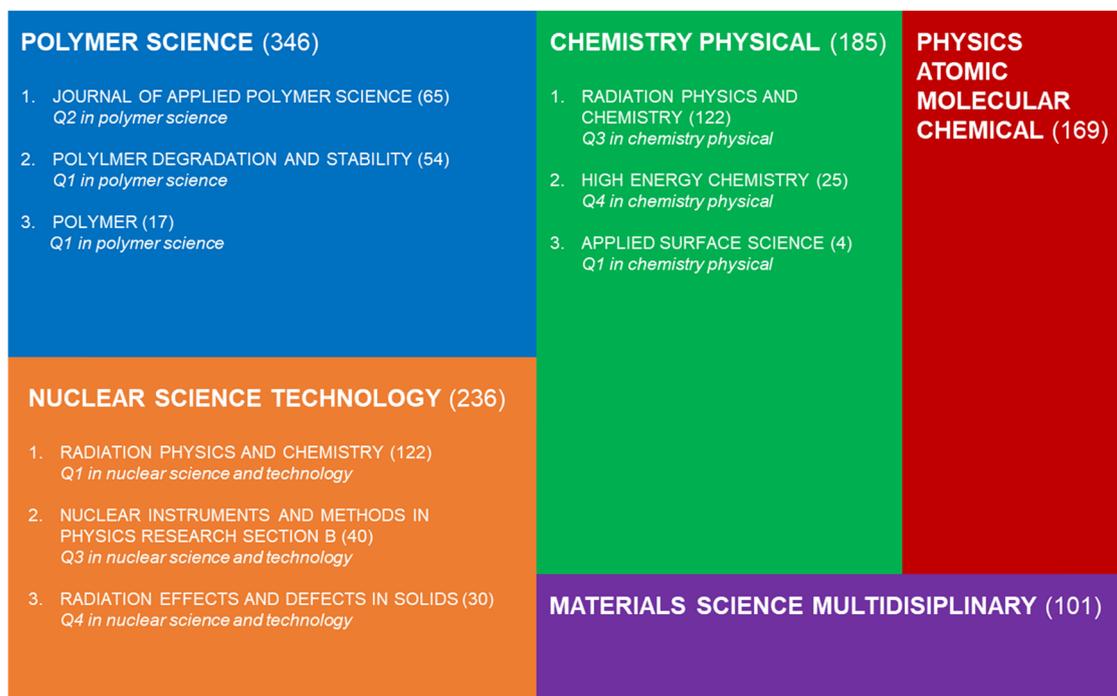


Figure 2: First five WOS categories of the whole research. The first three journals for each category are given.

they are classified following the journal source of the publication. Figure 2 shows the first five categories. The first category is defined as “polymer science” gathering 346 articles. This category gathers articles which are published in journals such as *Journal of Applied Polymer Science* (with 65 papers), *Polymer Degradation and Stability* (with 54 papers), and *Polymer* (with 17 papers). These journals belong, respectively, to quartiles Q1 or Q2 in the polymer science JCR® Category (Figure 2). This category gathers articles presenting all applications of polymers and all types of polymers.

The second category highlighted is “nuclear science technology” with 236 papers. The three main journals publishing articles in this category are *Radiation Physics and Chemistry* (122 papers), *Nuclear Instruments and Methods in Physics Research Section B: Beam Interactions with Materials and Atoms* (40 papers), and *Radiation Effects and Defects in Solids* (30 papers). These journals belong, respectively, to quartiles Q1, Q3, or Q4 in the nuclear science and technology JCR® Category. This category covers all aspects of the interaction of energetic beams with all types of materials.

The third category is defined by “chemistry physical” and includes 185 papers. It consists of articles published in journals such as *Radiation Physics and Chemistry* (122 papers), *High Energy Chemistry* (25 papers), and *Applied Surface Science* (four papers). These journals belong, respectively, to quartiles Q1, Q3, or Q4 in the chemistry physical

JCR® Category. These publications refer mainly to high energy chemistry, such as the interaction of high-energy particles with matter or studies on surfaces, interfaces, nanostructures, and their applications.

It is inferred that analytical chemistry is used as a tool to investigate polymer properties only.

3.3 Worldwide research distribution of the ionizing radiation impact on polymers

Figure 3 represents the different countries involved in the research on the impact of ionizing irradiations on polymers. The country which detained the highest record of publications is Egypt with 129 papers. It is followed by United States of America (USA) with 126 papers, India with 95 papers, Japan with 81 papers, and China with 74 papers. The top ten of the countries publishing such articles are listed in Table 1.

3.4 Focus on gamma, electron beam (e-beam), and X-ray irradiations as ionizing irradiations

To study deeply the different ionizing irradiations in the global research on the impact of ionizing irradiations on

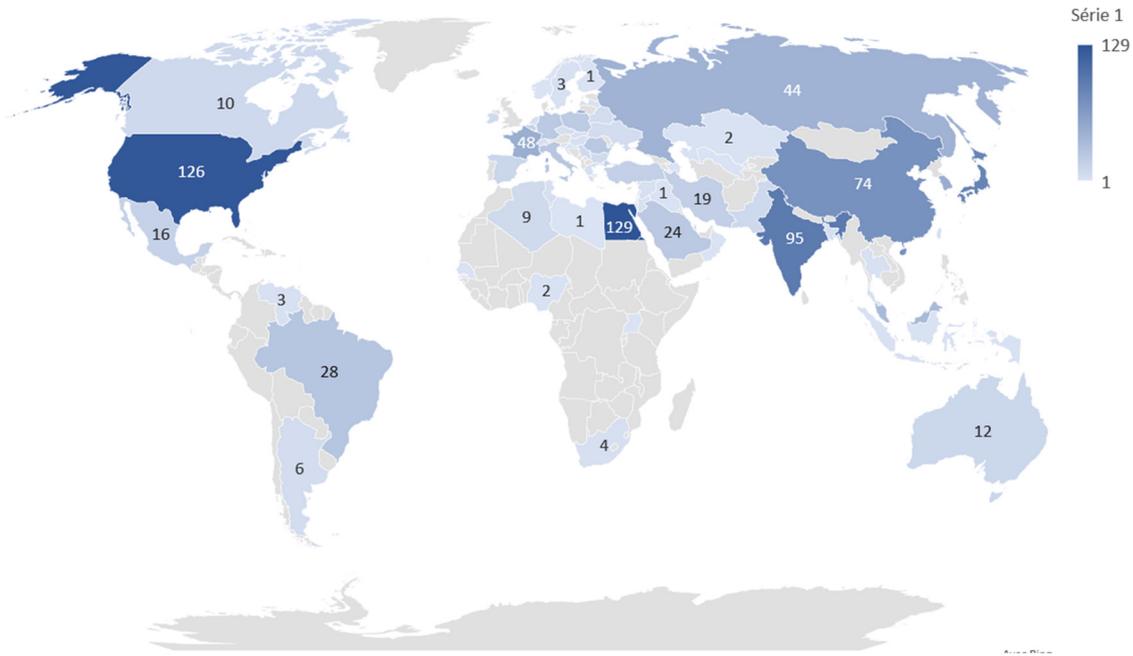


Figure 3: Worldwide research distribution between 1975 and 2019.

polymers, we focused on the three ionizing irradiation keywords in the initial research: gamma, electron beam, and X-ray irradiations. Figure 4 shows the number of papers published from 1975 to 2019 on X-ray, e-beam, and gamma irradiation. The publication is linked to one irradiation modality when the keyword gamma, electron beam, or X-ray appears in the title. Publications linked to the general ionizing irradiations keyword are also shown in Figure 4. These publications do not mention X-rays, e-beam, or gamma in the title but only ionizing irradiation. They can focus only on gamma, e-beam, or X-rays, or make a comparison for example.

With a total of 578 publications, gamma irradiation (in blue) is the most common irradiation modality with the

highest number of publications since 1975 and a constant increase. Chernobyl accident in 1986 and Fukushima Daiichi accident in 2011 seem to participate to the increase after 1990 and after 2011, respectively. We can observe two plateaus, the first one between 1975 and 1985 and the second one between 1990 and 2005.

Table 1: Top ten of the countries publishing in the ionizing irradiated polymer research field (1975–2019)

Rank	Countries/territories	# Publications	%
	World	1,015	100
1	Egypt	129	13
2	USA	126	12
3	India	95	9
4	Japan	86	8
5	China	74	7
6	South Korea	49	5
8	France	48	5
9	Russia	44	4
10	England	41	4

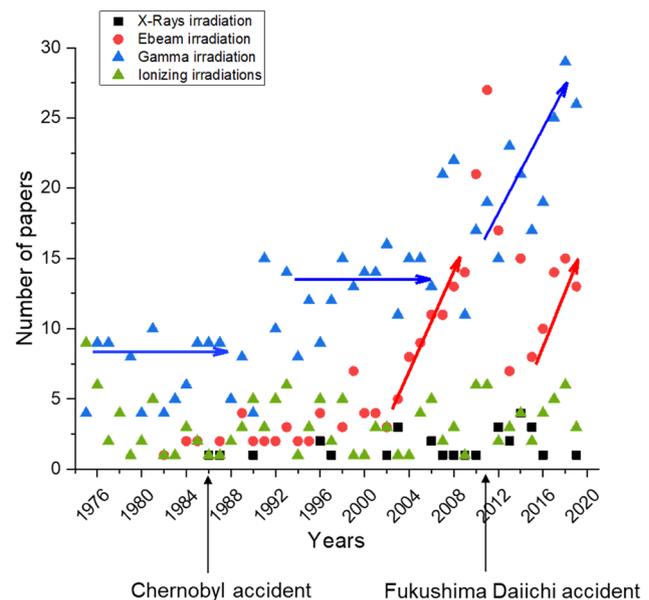


Figure 4: Number of papers overtime (1975–2019) referring to X-ray irradiation (black), e-beam irradiation (red), and gamma irradiation (blue). The fitting curves (dashed line) are given to ease the reading.

Electron beam irradiation (in red) comes in second place with a total of 267 publications. On an average, two papers are published per year from 1975 to 2000. From 2000 to 2010, the curve shows an increase from 4 papers to 27 papers. The interest decreased after 2010–2013 but since 2014 the number of publications has increased. This increase could also be explained by the Fukushima Daiichi nuclear accident, which happened in 2011. This increase is smaller than the one observed at the same period with gamma irradiation. Finally, X-ray irradiation (in grey) counts the lowest number of publications with 30 publications since 1975. It is important to keep in mind that even if electron beam and X-rays seem to be less studied than gamma irradiation, in this present study only the “impact” of these ionizing radiations is considered. However, in the literature, many studies report on the X-ray and e-beam “technologies” (9–11).

We now focus on the keywords of publications within the search on the ionizing irradiation impact on polymers to provide an overview of the research orientation, the type of polymers investigated, the analytical techniques used, and associated purpose. Figure 5 represents a network built on a keyword analysis. All keywords shown in Figure 5 can be clustering in three columns following the three main ionizing irradiation modalities: gamma,

electron beam, and X-ray irradiations. For each of these irradiations, keywords are assembled following four levels: materials, properties, features, and analytical techniques and will be reviewed presently.

The main materials investigated in gamma during this period are polyester, microspheres, polystyrene, rubber, poly lactic-*co*-glycolic acid (PLGA), polyvinyl alcohol (PVA), composites, biodegradable polymers, EPDM, PET, polyethylene, polypropylene, and PVC. These polymers represent mainly the packaging domain or biodegradable polymers. They are linked to the sterilization, work area largely investigated today as it is mainly used in the medical domain.

The main materials investigated in electron beam during this period are PTFE, polydimethylsiloxane (PDMS), PVDF, PET, polyethylene, and polypropylene. These polymers are historical polymers and they also have been studied under gamma irradiation in the sixties (12). These publications do not appear in this article as data are collected from 1975.

The main materials investigated in X-rays are PVC and cellulose. PVC constitutes one of the main polymers used in the blood-contacting applications and therefore the soft X-ray irradiation investigated. Other materials (not represented in Figure 5) such as polypeptides or

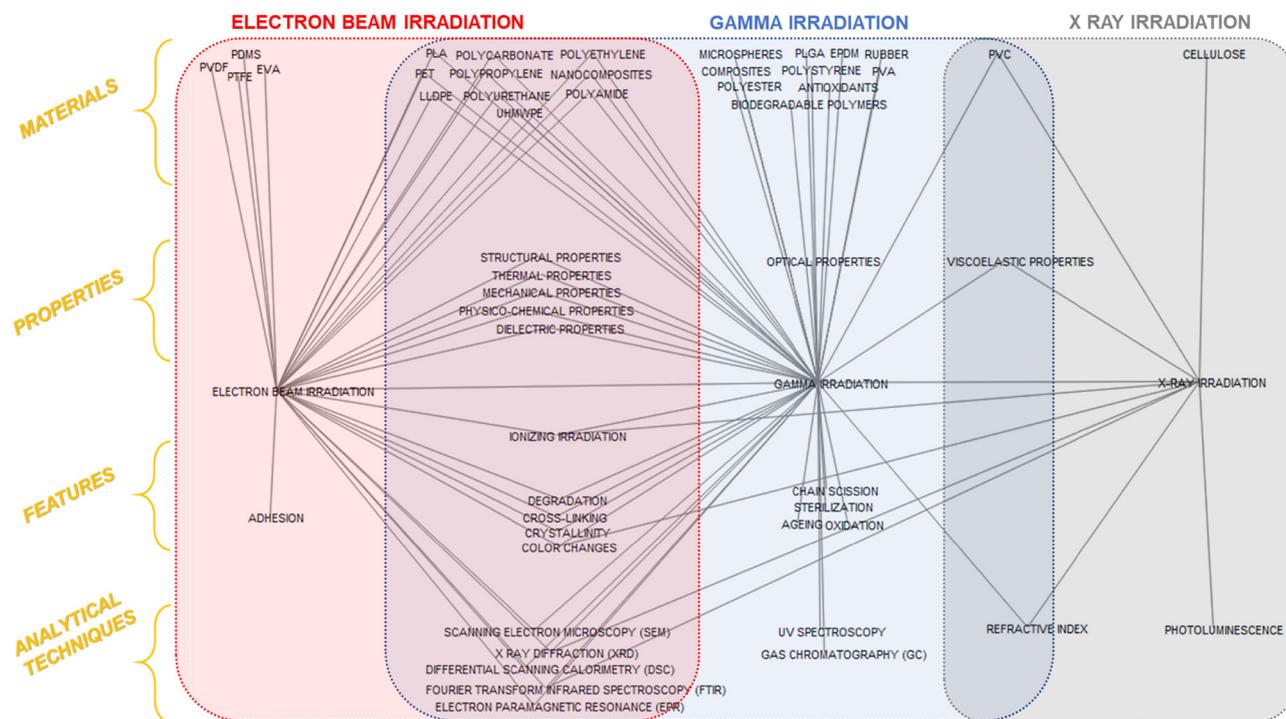


Figure 5: Keyword network (1975–2019) with a keyword’s frequency of minimum four for keywords linked with e-beam and gamma irradiation and two for those linked with X-ray irradiation.

cancer cells can be treated with X-rays in the medicine or biology domains. As it is explained before, X-ray irradiation seems to be less studied than gamma irradiation and electron beam as only the “impact” of these ionizing radiations is considered in this article. Moreover, there is only one industrial X-ray irradiator able to sterilize pallet which has been built in 2010 in Switzerland, and which could explain the lower proportion of X-ray irradiation.

Whatever the modality of irradiation, the polymer properties examined are thermal properties, physico-chemical properties, structural properties, dielectric properties, optical properties, viscoelastic properties, and mechanical properties.

To scrutinize these properties, the main analytical techniques used are Fourier transform infrared spectroscopy, X-ray diffraction, differential scanning calorimetry, electron paramagnetic resonance (EPR), and scanning electron microscopy.

We can observe that the 1,015 articles are split into two categories. The first one regroups articles which study the unwanted and uncontrolled modifications of the polymers during irradiation, whereas the second category regroups articles based on the polymer’s modification using irradiation as a tool to improve the original material.

For the first category, which regroups articles based on the degradation of the polymers under irradiation, we can find polymers such as polyethylene, polypropylene, polyester, polystyrene, PLGA, rubber, EPDM, PVC, PTFE, PVDF, and cellulose.

Miyagawa et al. studied the mechanical properties and the changes in the molecular weight distribution of low density polyethylene caused by gamma and electron beam irradiation and showed crosslinking reactions caused by irradiation (13). Other studies report on the effect of electron beam irradiation on polyethylene analyzing mechanical properties or thermal properties. Low density polyethylene shows an increase in trans vinylene double bond under electron beam irradiation (14). When high density polyethylene (HDPE) is irradiated with an electron beam under 250 kGy, a crosslinking is observed in amorphous area, whereas above 250 kGy, crosslinking is observed in the crystal domain area (15). Waste polyethylene electron beam irradiated showed enhanced physicochemical properties (16). HDPE studied with positron annihilation lifetime spectroscopy under gamma and electron beam irradiations showed no change in the lifetimes (17).

Polypropylene is widely used as a material of choice in single-use medical devices (i.e., syringes). Researchers studied the impact of gamma irradiation and electron

beam on the physicochemical and mechanical properties of the polypropylene syringes. Globally, they find minor polymer property degradation when syringes are sterilized with electron beam irradiation than gamma irradiation (18–21). Other study also reports less degradation when high crystalline polypropylene is irradiated with electron beam than gamma (22).

Polyester is studied under gamma irradiation. Buttafava et al. showed that chain scission is induced by irradiation for up to 100 kGy and crosslinking dominates between 100 and 300 kGy (23). Polyester such as estane was studied under electron beam and gamma irradiation. The study highlights that estane primarily undergoes rapid cross-linking and partially undergoes scission reactions (24).

Polystyrene is another material of interest in this category. Nichol et al. worked on the determination of the yields of crosslinking and scissions of polystyrene by gamma irradiation (25).

PLGA is a type of polymer mainly studied in the biomedical domain. The gamma irradiation on poly(lactico-glycolic) bioabsorbable microspheres shows a substantial effect on initial molecular weight distribution but no effect on mass loss (26).

Rubber is used in nuclear applications. Rivaton et al. showed chemical changes in gamma-irradiated EPDM and EPR films (27). In another study, the irradiation dose rate on commercial isobutylene–isoprene rubbers has been defined as an important parameter to control the degradation of butyl rubbers (26).

PVC is another polymer which can be used for nuclear industry. Relative humidity appeared to be a factor limiting the effects of temperature on the formation of HCl from pure gamma-irradiated PVC at high dose rates (28). PVC is also widely used in the biomedical domain as blood storage bags, tubing, or catheters. Manfredini et al. highlighted the process of oxidation taking place at the surface during soft X-ray exposure (29).

PTFE is known to have excellent chemical and physical properties. Jinglong et al. showed that the increase of irradiation dose can promote C–F and C–C scissions (30).

PVDF is widely used in industrial fields such as chemical and petrochemical process or wire and cable as it shows many outstanding properties such as high mechanical strength or excellent chemical resistance. The melting temperature increases for PVDF films irradiated at low dose (<30 kGy), whereas at higher irradiation doses (>30 kGy), it decreases with increasing irradiation dose (31). Also, electron beam irradiation increases the residual yield at high temperature of PVDF film due to the formation of crosslinked network structures (32).

Cellulose nitrate shows no significant changes when it is analyzed using energy-dispersive X-ray fluorescence spectrometry. This means that it should not show significant modifications under X-ray radiation (33).

For the second category which regroups articles based on the study of the properties improvement of original polymers using ionizing irradiation, polymers such as polyethylene, PET, PTFE, PDMS, and PVA are studied. Ultra-high molecular weight polyethylene is known to be used in biomedical domain as an implant. To improve properties, studies are undertaken by modifying the polymer using gamma irradiation (34,35) or electron beam irradiation (36). PET fibers can be grafted using electron beam irradiation with quaternary ammonium salt to improve the antibacterial activity of PET (37). PTFE and PDMS are jointly studied in the biomedical domain as they can be assembled. Indeed, it is well known that PTFE is poor radiation resistance (38). Homogeneous low voltage electron beam irradiation can be used as a useful method for quick adhesion of PDMS/PTFE without the use of heat and glue (39). Electron beam can also be used as a useful tool for quick lamination of PDMS and PTFE (40). PVA has been studied under gamma irradiation with and without crosslinker. The study shows that the addition of cross-linker led to high degrees of interaction between the cross-linker and the macromolecules of polymeric chains (41).

Depending on the type of polymers, it can be concluded that researchers show different aspects of degradation of polymers. For polymers such as polyethylene, polypropylene, polyester, polystyrene, PLGA, rubber, EPDM, PVC, PTFE, PVDF, and cellulose, whatever the irradiation modalities (i.e., gamma, electron beam, or X-rays), the topic of interest is the degradation. For polymers like polyethylene, PET, PTFE, PDMS, and PVA, the aim of the studies is to improve properties using crosslinking, chain scission, adhesion etc. under irradiation.

4 Conclusion

In this present study, the attention was focused on a bibliometric investigation from 1975 to 2019 on the impact of ionizing irradiations on polymers. Over the 1,015 publications, growing interest has been highlighted since 1990. We have shown that the research is distributed over three main categories: polymer science, nuclear science technology, and chemistry physical. Through these categories we find three main sterilization modalities: gamma, electron beam, and X-ray irradiations. The

impact of ionizing irradiations on polymers under gamma irradiation is the most studied field followed by electron beam irradiation which constitutes the second interest regarding ionizing irradiation. Finally, X-ray irradiation seems to be at the beginning of the research as a hint of the recent readiness of the X-ray technology for sterilization. By analyzing keywords in publication title and making a keyword network, we showed that, whatever the irradiation modality, the degradation is one of the aspects studied with polyethylene, polypropylene, polyester, polystyrene, PLGA, rubber, EPDM, PVC, PTFE, PVDF, and cellulose. The other aspect studied is the property improvement using crosslinking, grafting, and adhesion under irradiation. The latter regroups polymers like polyethylene, PET, PTFE, PDMS, and PVA.

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