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Mapping the scientific research on the gamma irradiated polymers degradation (1975–2018)

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ABSTRACT

Keywords:

Gamma irradiation
Polymer
Degradation
Bibliometrics
Keyword analysis

The global research trend on the degradation of gamma irradiated polymers from 1975 to 2018 was studied with a bibliometric approach. With 1339 papers published during this period, the investigation showed a growing interest overtime since 1975. The main countries working on that topic are USA, China, Japan and Europe. From 1975 to 1998, Japan was the first country to publish in this field. From 1998 to 2008, USA came on the top whereas from 2009 to 2018 China detained the most papers published. Networks analysis based on countries, authors and keywords showed interesting facts. First, a worldwide collaboration is growing overtime. Secondly, a multi-technique approach is necessary to study the degradation of the gamma irradiated polymers. Different work areas were highlighted. First, the journal sources study reveals three main domains: the nuclear, the biomedical and the polymer science domain. In a second time, the biology domain is the most discussed domain regarding at the top five most cited publications. Finally, the biopharmaceutical domain is the most represented domain regarding at the top five most recent publications.

1. Introduction

Bibliometric methods supply a portrait of the production of the sciences and the technology, measured by the production of the scientific publications, and flows of knowledge in the system of the sciences and of the technology, measured by joint signatures in the scientific publications. The purpose of this paper is to obtain a global view on the scientific studies concerning the degradation induced by gamma irradiation on polymers. Interest on other ionizing irradiations will be treated in another paper. For scientific articles the most worked domains are the resistance in the degradation of materials (i.e. nuclear domain) and sterilization (i.e. medical, pharmaceutical and food packaging domains).

In the nuclear domain, several studies are carrying on in this field about the stabilization of radioactive waste or the degradation of polymers used in nuclear plants (Pourmand et al., 2017; Nishad et al., 2012; Funabashi et al., 1991). In the medical (Valente et al., 2016) and pharmaceutical domains, polymers are present in numerous medical devices as syringes, gloves, devices of drip and taking, or still the single-use bags. They are also used as biomaterials – biological or synthetical materials which can be introduce into body tissue as part of an implanted medical device. The food packaging (Madera-Santana et al.,

2016) is also studied in the degradation of gamma irradiated polymers field as the irradiation acts as bactericide and fungicide solutions.

For these three domains, the gamma irradiation is used as a sterilization process which allows the eradication of microorganisms on the material (ISO 11137-1, 2006) it has a bactericidal and fungicide effect. The radio-sterilization corresponds to a sterilization made by means of ionizing radiations. It causes a break of the double helix of DNA (Aquino, 2012) so preventing any possible replication.

The gamma sterilization presents numerous advantages. It allows to sterilize samples at room temperature without important heating and to handle them directly in their definitive packaging. This guarantees the sterility in the time by avoiding every risk of possible re-contamination. It is about a continuous, reliable and reproducible process.

Unfortunately, the gamma irradiation presents also inconveniences. The polymers undergo modifications during a radiation exposure. Some factors could influence the degradation of polymers under irradiation, as their chemical composition, additives, or the presence of oxygen in the environment. The damages waited on the irradiated polymers are potentially the formation of radicals (Audran et al., 2015), small molecules, inter and intra chain bonding, insaturations, colorings (Gaston et al., 2018) and formation of oxidized molecules (Gaston et al., 2016a, 2016b), resulting in crosslinking and/or chain scission.

The aim of the present study is to have a global overview of academic research between 1975–2018 on polymer degradation after gamma irradiation. It will allow us to highlight emerging domains in the gamma irradiated polymers degradation field to identify whatever the applications how such investigations have been conducted. The work is led by studying the evolution of the annual publications, the different countries of publications, the sources used and by establishing authors-countries and keywords network. Identifying thus main polymers of interest for gamma irradiation purpose, main analytical tools used, and main properties impacted will be carried out.

2. Methodology

Data were obtained on June 27, 2018 from Web of Science Core Collection (WOS) databases from 1975 to 2018 as follows:

- The search was limited to titles, abstracts and keywords. Duplications were automatically removed.
- All journals have been considered
- Different words combinations have been selected: “gamma irradiation” and “polymer*”, “gamma irradiation” and “film*”, “gamma irradiation” and “resin*”, “gamma irradiation” and “packaging”. All these sets have been assembled. The result has been combined with “gamma irradiation and degradation” set using the connector “and”. The Asterix describe the possibility to consider the word either in plural or in singular.
- Outputs were the authors, the journal publication, the country of universities, the institutions and keywords.

Data were then exported as text file and analyzed with Mateo Analyzer software.

It is important to note that only publications from Web of Science Core Collection are collected. Indeed, some organization such as International Atomic Energy Agency has their own collection (International Nuclear System Information). Also, Web Of Science has mainly publications written in English. Countries could have their own journals and write publications in Japanese or Russian for example. All these publications are then not considered in this study.

3. Findings and discussion

3.1. Yearly distribution

A total of 1339 different papers have been found between 1975 and 2018 on the degradation of gamma irradiated polymers with the criteria defined in previous chapter. The number of papers published every year is shown in Fig. 1. There is only one paper published in 1976 and 1991. The reason why there is only 2 papers before 1991 can be explained by the fact that papers may have not been scanned. From 1991, there is a constant increase overtime. The maximum is observed in 2016 with 97 papers.

3.2. Country of publication and evolution

Since 1975, there are 72 different countries working on that topic. They are colored in blue in function of the number of publications in the Fig. 2. The top five country (representing more than 5% of total publications) is composed of United States of America, China, Japan, France and Egypt respectively and represents 35% of total publications (Table 1). First, it is not surprising that United States of America, China, Japan and France appear in the top five countries publishing on that topic. Indeed, these countries are the main global nuclear power (Portugal-Pereira et al., 2018). However, Russia should also appear in this list. It does not, probably because Russia publishes on their own journal in Russian. Secondly, we note that Egypt takes place in the top five. Egypt does not take part of the global nuclear power, but it detains

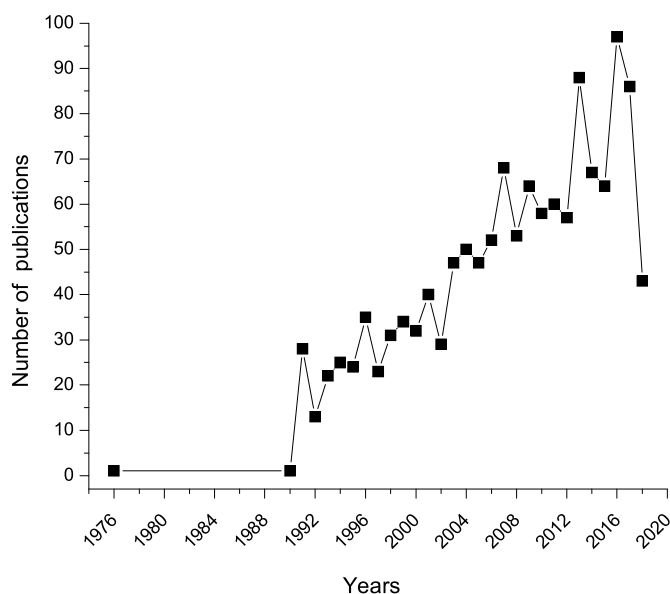


Fig. 1. Number of publications overtime (1975–2018).

two nuclear power plants intended for research (Supersberger and Führer, 2011). They have then worked on that topic in the previous years.

In the Fig. 3, we can observe the evolution of publication numbers overtime for the top 5 countries. Although United States of America is the country which detains the most of publication in this field, it does not record the maximum of papers every year.

From 1990 to 1998, Japan was the country to publish the maximum of papers (orange triangle). From 1998 to 2008, USA detained the maximum of papers with an average of 9 papers per year (green square). Since 2008, China (blue circle) publishes with an average of 12 papers per year contrary to 7 papers for the USA. France (red triangle down) and Egypt (purple diamond) increase the number of publications overtime.

Over the past few years, China shows an increasing interest regarding the degradation of gamma irradiated polymers field. Fossil fuels used in the world has a clear detrimental effect on life conditions and environment. Authorities and researchers from China are then taking a closer look on the nuclear energy to reduce the pollution issues (Hsiao and Chen, 2018). Indeed, in 2006 Chinese State Council issued 2005–2020 National plan for Medium- and long-term Nuclear Power Development. The will is to increase the number of power plant by four to eight every year until 2030. At the moment, China detains 40 nuclear power plants in operation and 20 under construction (China Nuclear Power, 2018).

3.3. Sources used

Publications referring about degradation of gamma irradiated polymers are recorded in 426 different academic sources of publications. *Radiation Physics and Chemistry* represents the most productive journal with 160 papers (12% of total papers) and it is followed by *Polymer Degradation and Stability* with 108 papers (Table 2).

Focusing on the domains pointed by the quartiles, it indicates us a global view regarding the use of the gamma irradiated polymers area. The first five sources to publish in this field have an impact factor (IF) between 1.4 and 3.5 contrary to *Biomaterials* with an IF of 8.806. However, when we focus on the quartiles, it is interesting to note that *Radiation Physics and Chemistry* is in the first IF quartile in the Nuclear Science & Technology domain. Moreover, *Nuclear Instruments & Methods in Physics Research Section B: Beam Interactions with Materials and Atoms* is in the second IF quartile in the Nuclear Science & Technology

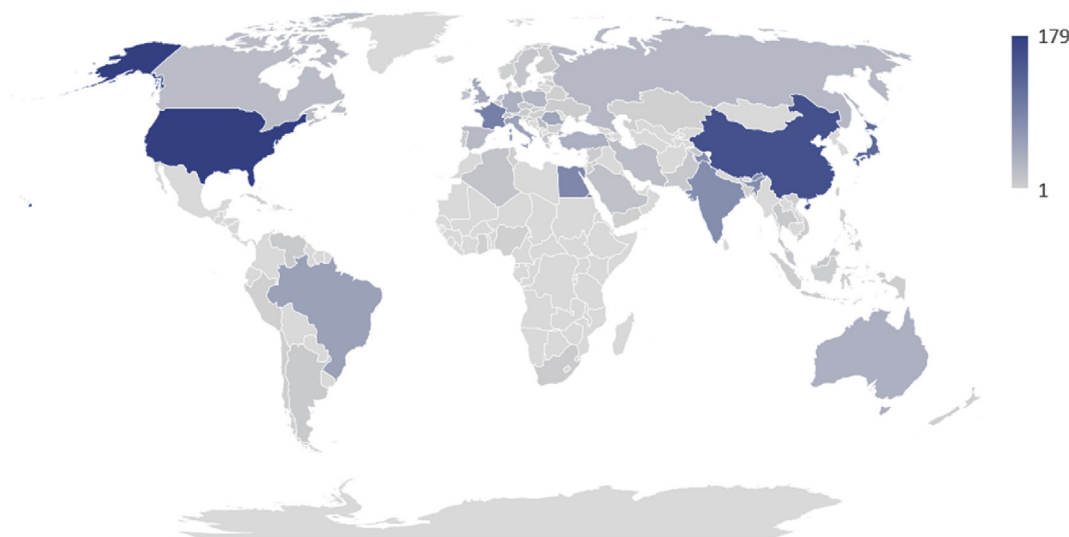


Fig. 2. World map of countries publishing in the degradation of gamma irradiated polymers field (1975–2018).

Table 1

Number of papers for the first five countries.

Rank	Country	Number of papers	% of 1471 ^a
1	United States of America	179	12.2
2	China	155	10.5
3	Japan	121	8.2
4	France	94	6.4
5	Egypt	84	5.7

^a The sum of total publications is 1471 (vs 1339 as recorded in part 2.1.) because when one paper is written by researchers coming from two different countries, the paper is counted twice (one for each country.).

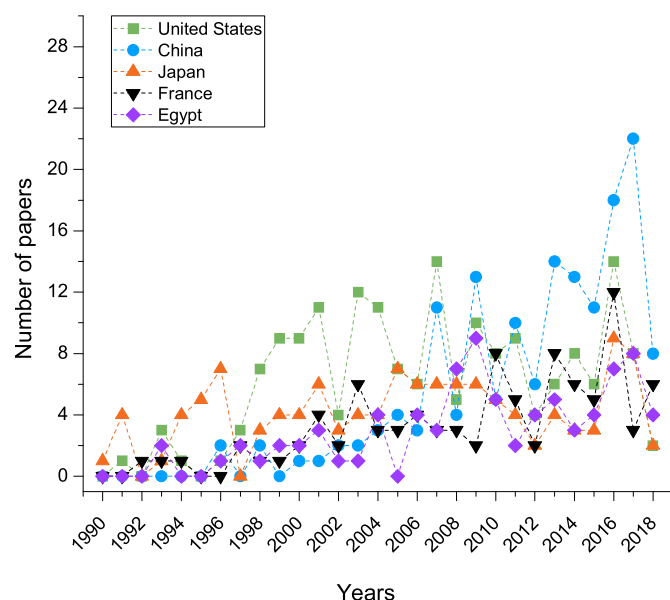


Fig. 3. Publications number evolution for United States, China, Japan, France and Egypt (1975–2018).

domain. The IF of these journals is relatively low as it is very specialized and not read by a large audience. On the other hand, *Biomaterials* journal is in the first IF quartile in Engineering, Biomedical domain and in Materials Science, Biomaterials domain. Finally, *Polymer Degradation and Stability*, *Journal of Applied Polymer Science* and *Polymer* journals are in the first two IF quartiles in the Polymer science domain. These three

journals publish articles on fundamental and applied polymers science.

In conclusion, by analyzing the academic sources of publications, three main domains appear: the nuclear, the biomedical, and the polymer science research. With the will to build more power plant in the world, researchers are working deeply on the degradation of the constitutive materials in the nuclear plants. They work on the degradation mechanisms of insulation, materials composed of EVA (Ethylene Vinyl Acetate) and EPDM (Ethylene Propylene Diene Monomer) (Gong et al., 2017; Šarac et al., 2016a, 2016b). To go further, we looked at the first three most cited publications referring about the keyword *nuclear* (in the whole research i.e 1339 papers). The first paper, with 150 citations, is a review and it highlights the degradation of fluoropolymers irradiated (for example in nuclear facilities) (Forsythe and Hill, 2000). The second most cited publication (with 58 citations) is a review which studies the use of microspheres in radio-nuclide therapy (Nijssen et al., 2002). Finally, the third paper (with 22 citations) reports how functionalized graphene can act as a radical scavenger in gamma irradiation environment (Xia et al., 2016).

3.4. Most cited publications

3.4.1. Most cited publications

In Table 3, it is recorded the top 10 of the most cited publications in the “gamma irradiated polymers degradation” field. They have been published around 20 years ago. It means that important work has mainly been done years ago in this domain. We do not find the highest IF of this research in this table (which is 24.6 corresponding to Progress in Polymer Science journal) likely because the journal is more general and publishes articles about polymer science and engineering. Considering the first five most cited publications, the cells and molecular biology domain appears as the most investigated domain in the degradation of gamma irradiated polymers field. It is important to note that with “polymer” as an entrance keyword in the initial search, all existing organic and inorganic polymers such as the ones used in the life sciences (i.e. proteins) appear. A distinction has then to be done between the life sciences and the material sciences. Regarding the other publications (from rank 6 to 10), the biomedical/medical domain is highlighted with the study of drug delivery (Song et al., 1997) and drug release (Alexis, 2005).

3.5. Most recent publications

The Table 4 records the last 5 articles published when the research with Web of Science has been done. One publication (rank 2) refers to

Table 2
Top 10 of sources used with Impact Factor and Quartile in Category.

Id	Source publication	# Publications	Percentage	Impact Factor (2017)	Q1	Q2
1	Radiation Physics and Chemistry	160	12%	1.435	Nuclear Science & Technology (8/33)	NA
2	Polymer Degradation and Stability	108	8%	3.193	Polymer Science (16/87)	NA
3	Journal of Applied Polymer Science	78	6%	1.901	NA	Polymer science (38/87)
4	Nuclear Instruments & Methods in Physics Research Section B: Beam Interactions with Materials And Atoms	60	4%	1.323	NA	Nuclear Science & Technology (11/33)
5	Polymer	20	1%	3.483	Polymer Science (14/87)	NA
6	Biomaterials	19	1%	8.806	Engineering, Biomedical (1/78)	NA
7	Carbohydrate Polymers	17	1%	5.158	Materials Science, Biomaterials (1/33)	NA
8	Radiation Effects and Defects in Solids	17	1%	0.526	Chemistry, Applied (2/71)	NA
9	Journal of Polymer Science Part A-Polymer Chemistry	16	1%	2.588	Chemistry, Organic (6/57)	Polymer science (23/87)
10	Polymer Bulletin	15	1%	1.589	Polymer Science (7/87)	NA

the nuclear domain. It is interesting to note that the biopharmaceutical domain appears with the publication in rank 3 and 5. Indeed, some polymers manufactured as multilayer film with a barrier properties layer in the middle can be used in the pharmaceutical domain to prevent cross contamination (Gaston et al., 2016a).

3.6. Network analyses

3.6.1. Countries/authors

To study the evolution of researchers and country working on the field of gamma irradiated polymers degradation, three networks have been built. The first one shows the authors-countries network for the period 1975–1998 (Fig. 4), the second one for the period 1999–2008 (Fig. 5) and the third one represents the last period 2009–2018 (Fig. 6).

To build a network with Mateo analyzer software, two factors must be considered. The first one is called “Pairs Frequency (PF)”, it represents the number of papers published between one author and one country. The number is located on the line between these both. The second factor is “Forms Frequency (FF)”. It is the number of papers published for one author or one country. The number is written in top right of the author name or country. To ease the reading, it is necessary to fix these 2 criterions. For the 3 networks (Fig. 4–6), $PF \geq 1$ and $FF \geq 4$. It is important to keep in mind that sometimes a group of authors are linked to one country but only one of them is truly connected to the country as he works for both countries. For example, in Fig. 6, France and Russia are linked only by Marques, SRA.

Regarding the first period (Fig. 4) one main sub-network is observed and it is constituted of Japan (25 papers), USA (16 papers), Australia (10 papers), Germany (6 papers) and Switzerland (5 papers). Japan is predominant with 11 authors connected and 25 publications. This first observation confirms the data obtained in Fig. 3. The second sub-network is represented by France (6 papers) and Canada (5 papers). They published one paper together.

For the second period (1999–2008, Fig. 5), the main sub-network observed in Fig. 4 has grown and US is now predominant with 88 papers contrary to 50 papers for Japan (in the Fig. 4). This sub-network represents the worldwide sub-network. Indeed, USA are linked with Australia, Italy and Japan. China appears in the sub-network working with Japan. China has 31 papers published during this period. UK, Ireland, Brazil and Egypt are also present. Once again, the observation obtained in Fig. 3 is confirmed: USA was the first country to publish during this period (1999–2). The second main sub-network is composed of Romania (24 papers), France (31 papers), Germany (19 papers), Algeria, Venezuela (7 papers), Belgium (5 papers) and Portugal (4 papers). This second sub-network constitutes the European network.

Overtime, it is interesting to note that researchers all over the world start to work together. Indeed, during the first period only Japanese, north Americans and Australians published together. In the second period, there are more publications and consequently a larger interest regarding this field.

For the third period (2009–2018, Fig. 6), four sub-networks are observed. The larger one (represented, in the first period by Japan and in the second period by USA) is represented now by China (120 papers), USA (75 papers), India (54 papers), Egypt (51 papers) and Japan (46 papers). There are 20 countries in total representing this sub-network. The second one is represented by France (57 papers) and Russia (15 papers). The third one is composed of Turkey and Ireland. Surprisingly, Australia (16 papers) left the main sub-network and works with Serbia (14 papers) with two authors.

All these observations highlight the fact that the international collaboration grows overtime. However, all these networks show that knowledge of gamma irradiated polymer degradation is mainly distributed in Japan, US, China and France overtime and comfort the observations and conclusions obtained in section 2.3.

3.6.2. Key words

Analyzing keywords of publications within the search “degradation of gamma irradiated polymers” it enables an overview of which polymers are studied, how they are studied and for what purpose. Fig. 7 represents a network built on a content analysis which quantifies the number of different co-occurrences in all papers. Two criteria are considered. The first one represents the keyword frequency in the whole search (within the 1339 papers) and this number is indicated at the top right of the keyword (Fig. 7). The other one is the number on the line between two keywords. It represents the co-occurrence keyword frequency. To ease the network reading, the minimum keyword frequency is fixed at 9 and the minimum co-occurrence keyword frequency at 12. The network is divided in 5 parts (circled in yellow) in Fig. 7. *Gamma irradiation* and *degradation* keywords are not taken into account in the discussion as they constitute the main keywords of the initial search.

On the right side, we can observe the different materials studied. The polymers mainly studied are *polyethylene* (111 times), *biodegradable polymers* (63 times) and *polypropylene* (44 times). Polyethylene remains the polymer the most studied in the field of gamma irradiation polymers degradation. Indeed, it appears with a frequency of 111. It is not surprising as it is often studied in the biomedical domain. For example, the UHMPWE (Ultrahigh Molecular Weight Polyethylene) used as articular implant is known to be oxidized (Jahan et al., 1991; Premnath et al., 1996; McKellop et al., 1999). Moreover, it can be used as cable

Table 3
Top 10 most cited publications.

Rank	Publication name	Authors	Average citations per year	Times cited (2018)	Date	Source publication and IF ^a
1	Specific proteolytic cleavage of poly (ADP-ribose) polymerase - an early marker of chemotherapy-induced apoptosis	Kaufmann, Sh; Desnoyers, S; Ottaviano, Y Et Al	57.5	1552	September 1993	Cancer Research (IF = 8.38)
2	Differentiation between cellular apoptosis and necrosis by the combined use of in-situ tailing and nick translation technique	Gold, R; Schmied, M; Giegrich, G; Et Al	26.1	678	August 1994	Laboratory Investigation (IF = 3.68)
3	Enhanced photocatalytic activity of Ce ³⁺ -TiO ₂ for 2-mercaptobenzothiazole degradation in aqueous suspension for odor control	Li, Pb; Li, Xz; Hou, Mf; Et Al	24.1	361	May 2005	Applied Catalysis A-General (IF = 4.63)
4	Gamma sterilization of UHMWPE articular implants: an analysis of the oxidation problem	Premnath, V; Harris, Wh; Jasty, M; Et Al	11.9	286	September 1996	Biomaterials (IF = 10.3)
5	Comparison of apoptosis in wild-type and fas-resistant cells: Chemotherapy-induced apoptosis is not dependent on Fas/Fas ligand interactions	Eischen, Cm; Kottke, Tj; Martins, Ln; Et Al	10.7	245	August 1997	Blood (IF = 16.6)
6	Formulation and characterization of biodegradable nanoparticles for intravascular local drug delivery	Song, CX; Labhasetwar, V; Murphy, H; et al.	10.6	243	January 1997	Journal of controlled release (IF = 7.90)
7	Factors affecting the degradation and drug-release mechanism of poly (lactic acid) and poly [(lactic acid)-co-(glycolic acid)]	Alexis, F	15.3	229	January 2005	Polymer international (IF = 2.43)
8	Gamma-initiated, photo-initiated, and thermally initiated oxidation of linear low-density polyethylene - a quantitative comparison of oxidation-products	LACOSTE, J; CARLSSON, DJ	7.79	218	March 1992	Journal of polymer science part A-polymer chemistry (IF = 2.59)
9	Regulating bone formation via controlled scaffold degradation	Alsberg, E; Kong, HJ; Hirano, Y; et al.	12.7	215	November 2003	Journal of dental research (IF = 5.13)
10	Effect of sterilization method and other modifications on the wear resistance of acetabular cups made of ultra-high molecular weight polyethylene - A hip-simulator study	McKellop, H; Shen, FW; Lu, B; et al.	9.10	182	December 2000	Journal of bone and joint surgery-American volume (IF = 4.716)

^a IF from 2018.

Table 4
Top 5 most recent publications.

Rank	Publication title	Authors	Source publication	Date
1	Electron-Beam radiation effects on the structure and properties of polypropylene at low doses rates	Wang, Heng-ti; Jiang, Hai-Qing; Shen, Rong-Fang; et al.	Nuclear Science and Techniques (IF = 0.779)	June 2018
2	Hydration effect on ion exchange resin irradiated by swift heavy ions and gamma rays	Boughattas, I; Labed, V; Gerenton, A; et al.	Journal of Nuclear Materials (IF = 2.447)	June 2018
3	Reconciliation of pH, conductivity, total organic carbon with carboxylic acids detected by ion chromatography in solution after contact with multilayer films after gamma-irradiation	Dorey, Samuel; Gaston, Fanny; Dupuy, Nathalie; et al.	European Journal of Pharmaceutical Sciences (IF = 3.466)	May 2018
4	Single step synthesis of Schottky-like hybrid graphene - Titania interfaces for efficient photocatalysis	Yi, Zhifeng; Merenda, Andrea; Kong, Lingxue; et al.	Scientific Reports (IF = 4.122)	May 2018
5	Monitoring of the discoloration on irradiated PE and EVA films to evaluate antioxidant stability	Gaston, Fanny; Dupuy, Nathalie; Marque, Sylvain R.A; et al.	Journal of Applied Polymer Science (IF = 1.90)	May 2018

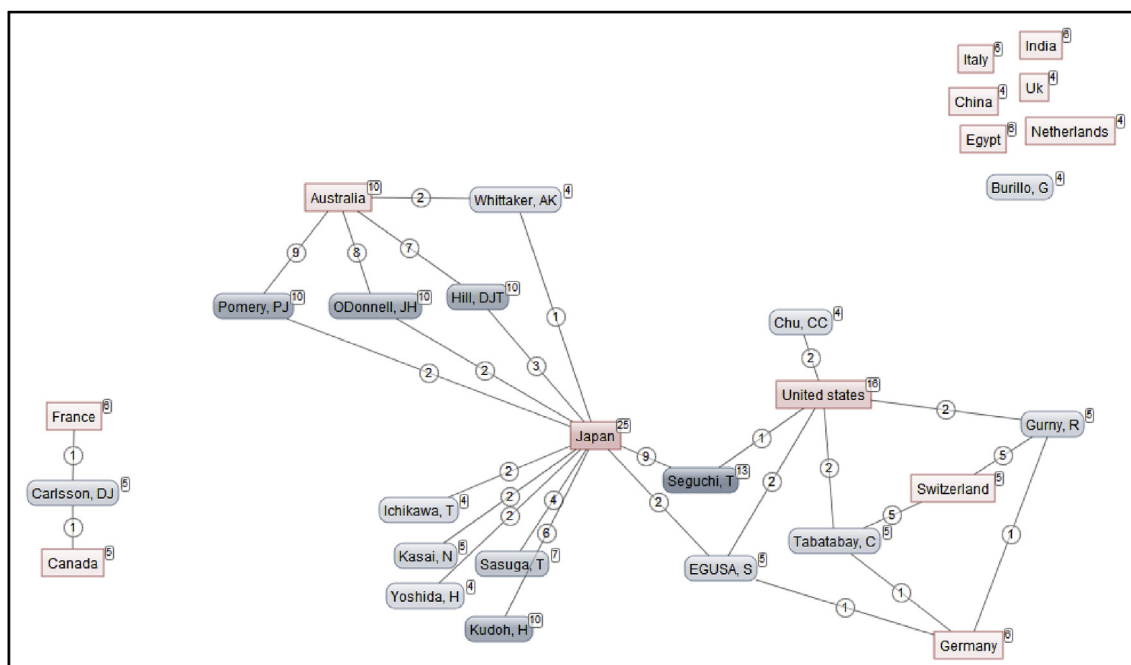


Fig. 4. Authors-countries network (1975–1998).

component with other polymers in nuclear power plant (Kurihara et al., 2011; Cassidy et al., 2016). Also, polyethylene is one of the main polymers used in food packaging. Researchers work on the degradation of gamma irradiated polymers used in food packaging industry (Buchalla et al., 1993; Goulas et al., 2004; Buchalla and Boess, 2000). Polyethylene is followed by biodegradable polymers with 63 occurrences which refers directly to the biomedical/biology domains such as the study of biodegradable particles on drug released (Siepmann et al., 2004; Montanari et al., 1998; Lee et al., 2002). Polypropylene comes in third position as it is often used as a matrix material. For example, Oliani et al. study the effects of aging to poly (propylene) and poly-

(propylene) modified by irradiation (Oliani et al., 2016). Other polymers such as hydrogel, PLGA Poly(Lactic-co-Glycolic Acid), biomaterials or chitosan belong also to the biomedical/biology domains as well. In this section, it appears nanocomposites and nanoparticles with a frequency of 38 and 34 papers respectively. It means that the degradation of smart gamma irradiated polymers is also studied. Indeed, incorporating nanomaterials in polymer matrices, polymers properties can be significantly increased. Xia et al. show that anchor ceria nanoparticles onto graphene sheets and incorporate them into epoxy resins could act as radical scavenger (Xia et al., 2017). This phenomena could then delay the ageing and the degradation. Moreover, Visakh et al. published

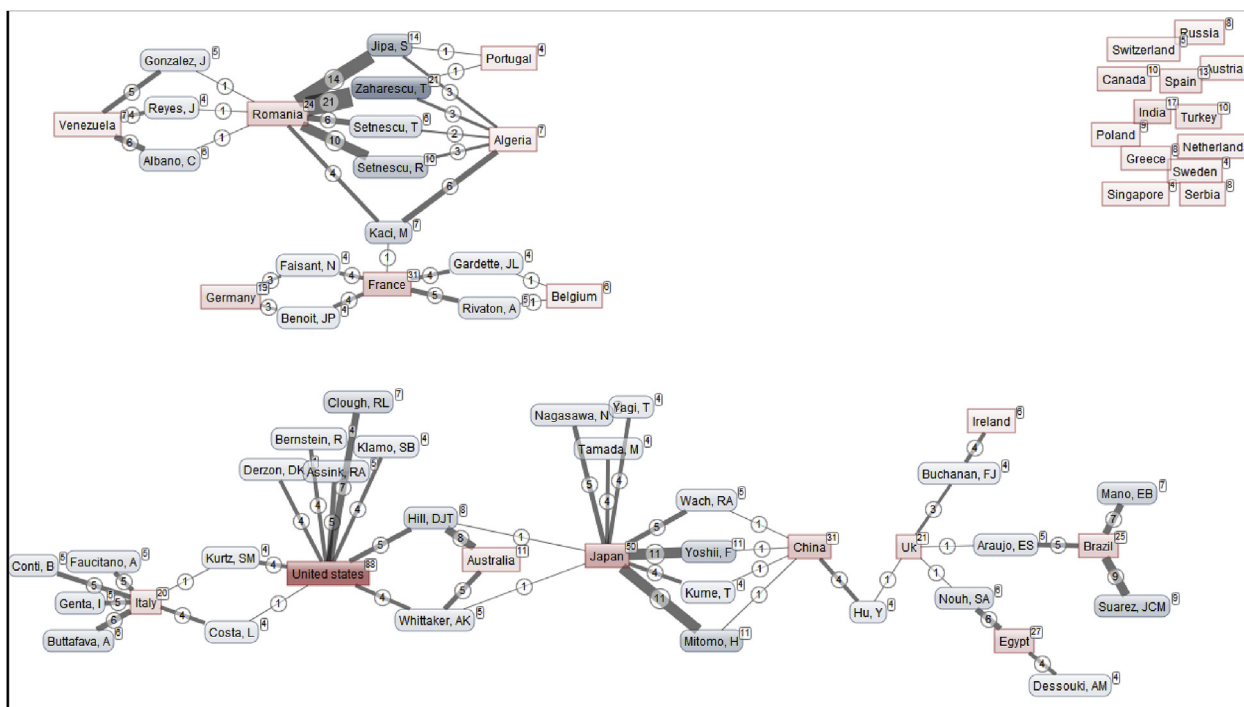


Fig. 5. Authors-countries network (1999–2008).

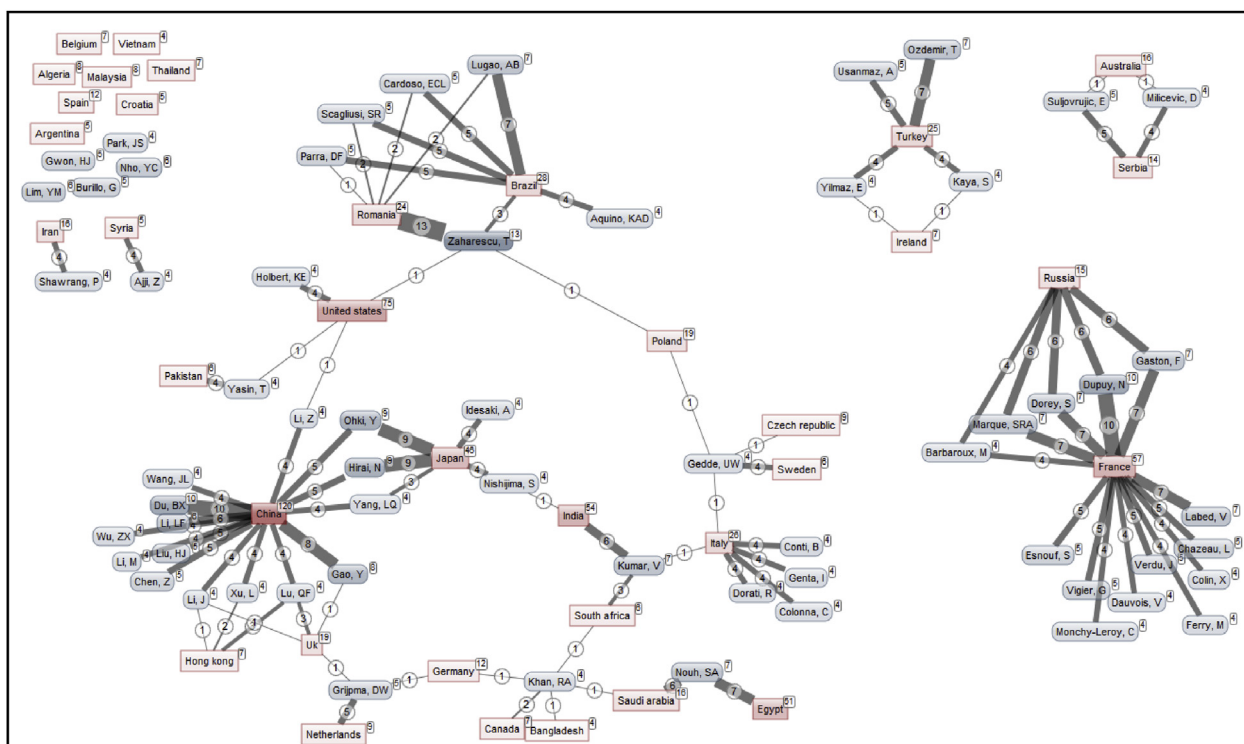


Fig. 6. Authors-countries network (2009–2018).

the fact that introducing aluminium particles into epoxy composites, thermal and mechanical properties for materials irradiated up to 100 kGy (Visakh et al., 2017) are improved.

At the bottom, the analytical techniques used to study the degradation of these polymers are grouped: *ESR* (Electron Spin Resonance) with 168 occurrences, *FTIR* (Fourier Transform InfraRed

spectroscopy) with 72 occurrences, *XRD* (X-Ray Diffraction) and *NMR* (Nuclear Magnetic Resonance) with 26 and 24 occurrences respectively are used. *ESR* is the analytical technique the most cited. This technique allows to monitor radical species in a polymer. It is known that gamma irradiation induces radical's formation by breaking bonds (Klepac et al., 2014). Therefore, radicals can react with other molecules and create

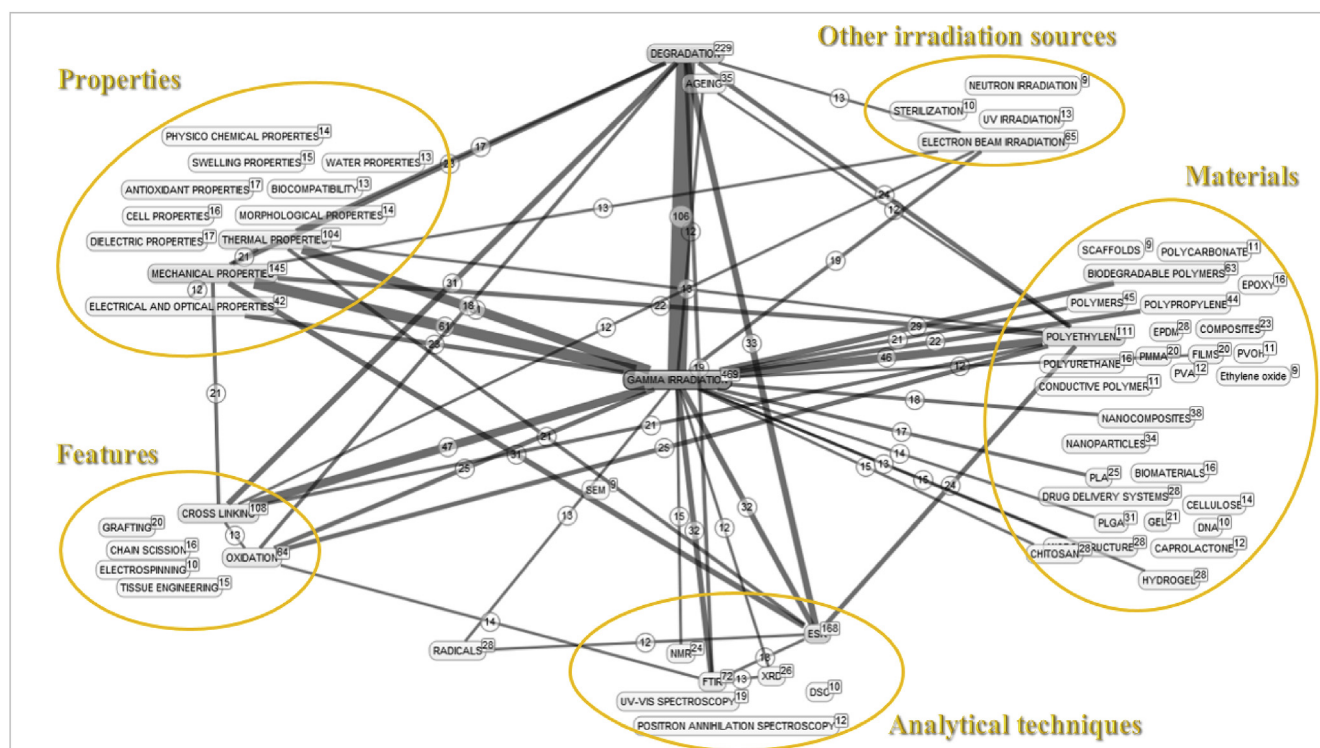


Fig. 7. Key words network (1975–2018) with a keyword's frequency of minimum 9 and a co-occurrence keywords frequency of minimum 12.

new chemical bonds and then new chemical species. The properties will then be modified, and the degradation of the gamma irradiated polymers will be partly explained.

At the top left, the polymers properties with *mechanical properties* and *thermal properties* keywords are gathered. They are the most recorded words with 145 and 104 times respectively.

At the bottom left, the features studied on polymers are grouped. It represents the result of polymer degradation. The main features studied are *crosslinking* and *oxidation* with a frequency of 108 and 64 respectively. These two keywords are linked and appear both in 13 publications. Crosslinking has two different meanings here. On the one hand, crosslinking is used as a technique to improve properties. For example, crosslinking can be used to improve mechanical integrity of biopolymers (Delgado et al., 2018), or crosslinking can be used simultaneously with irradiation to synthesize the formation of silver nanoparticles (Alcántara et al., 2019). On the other hand, crosslinking can also be an unwanted consequence of ionizing irradiations and can cause materials degradation such as a lower elongation at break for silicone foam (Liu et al., 2018).

Importantly, *ESR*, *FTIR*, *oxidation*, *cross linking* and *mechanical properties* are linked together. It explains that to study the degradation of gamma irradiated polymers, a multi-techniques approach must be investigated to understand the degradation phenomena observed on gamma irradiated materials. For example, Damir Klepac et al. studied the behavior of polyethylene and polypropylene films under gamma irradiation. They used FTIR technique to observe new chemical bands and therefore highlight an oxidation of the film. Then they used ESR technique to explain the formation of these new bands (Klepac et al., 2014). Another study reports the attenuation of the loss of toughness provoked by irradiation when radicals are scavenging by the presence of UHMWPE/MWCNT nanocomposites (Martínez-Morlanes et al., 2011).

At the top-right, alternative ionizing irradiations are grouped such as UV-irradiation and electron beam irradiation. *Electron beam* is linked with *gamma irradiation* and these two keywords appear together in 65 publications. Comparative studies can be made to select or compare one type of ionizing irradiation to another. For example, Suyong Kim et al. compared electron beam and gamma irradiation on dissolving micro-needle encapsulated with ascorbic acid 2-glucoside and find that electron beam irradiation does not degrade the drug encapsulated in contrary to gamma irradiation (Kim et al., 2018).

With 65 occurrences in the whole research (1339 papers), electron beam ionizing irradiation seems to appear as an emergence domain in the field of gamma irradiated polymers degradation. Since 2006, we observe an increase of papers published each year.

4. Conclusion

This bibliometric investigation from 1975 to 2018 on the gamma irradiated polymers degradation has shown an important growth of publications overtime since 1975. By analyzing the journal sources of the 1339 publications recorded, they can be distributed in three main domains: nuclear, biomedical and polymer science. However, focusing on the top ten most cited publications, the biology domain stands out followed by the biomedical domain. The biopharmaceutical domain, not highlighted with journal sources analysis, constitutes the most represented domain in the top five most recent publications. The search is mainly distributed over the USA, China, Japan and Europe. The analysis of publications country shows that before 1998 Japan detained the most publications. Between 1998 and 2008 USA were the first to publish before China has taken the lead since 2008. The author-country networks highlight a worldwide collaboration growing overtime. The keywords network points out mostly the biomedical science which is one of the main areas of work highlighted in the sources' study. The main polymers studied are polyethylene, polypropylene (both often used as a model in several applications) and biodegradable polymers

(mainly studied in biomedical applications). For most of these polymers, the evolution of mechanical, thermal, electrical and optical properties is explored. A multi technique approach is used to identify the main degradation (cross linking and oxidation) pathways with Electron Spin Resonance, Nuclear Magnetic Resonance, X-Ray Diffraction and Fourier Transformation InfraRed techniques.

In the same way, the investigation shows that researchers seem to work on another irradiation sources such as electron beam irradiation or X-ray.

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Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

References

- Alcántara, M.T.S., Lincopan, N., Santos, P.M., Ramirez, P.A., Brant, A.J.C., Riella, H.G., Lugão, A.B., 2019. Simultaneous hydrogel crosslinking and silver nanoparticle formation by using ionizing radiation to obtain antimicrobial hydrogels. *Radiat. Phys. Chem.* 165, 108369. <https://doi.org/10.1016/j.radphyschem.2019.108369>.
- Alexis, F., 2005. Factors affecting the degradation and drug-release mechanism of poly (lactic acid) and poly[(lactic acid)-Co-(Glycolic acid)]. *Polym. Int.* 54 (1), 36–46. <https://doi.org/10.1002/pi.1697>.
- Aquino, K.A. da S., 2012. Sterilization by gamma irradiation. *Gamma. Radiat.* <https://doi.org/10.5772/34901>.
- Audran, G., Dorey, S., Dupuy, N., Gaston, F., Marque, S.R.A., 2015. Degradation of γ -irradiated polyethylene-ethylene Vinyl alcohol-polyethylene multilayer films: an ESR study. *Polym. Degrad. Stab.* 122, 169–179. <https://doi.org/10.1016/j.polydegstab.2015.10.021>.
- Buchalla, R., Boess, C., 2000. Analysis of volatile radiolysis products in gamma-irradiated LDPE and polypropylene films by thermal Desorption $\pm$ gas Chromatography $\pm$ mass spectrometry. *Appl. Radiat. Isot.* 19.
- Buchalla, R., Schuttler, C., Bogl, K.W., 1993. Effects of ionizing radiation on plastic food packaging materials: a review. *J. Food Prot.* 56, 8.
- Cassidy, J., Nesaie, S., McTaggart, R., Delfanian, F., 2016. Mechanical response of high density polyethylene to gamma radiation from a cobalt-60 irradiator. *Polym. Test.* 52, 111–116. <https://doi.org/10.1016/j.polymertesting.2016.04.005>.
- China nuclear power | Chinese nuclear energy - world nuclear association. <http://www.world-nuclear.org/information-library/country-profiles/countries-a-f/china-nuclear-power.aspx>, Accessed date: 12 September 2018.
- Delgado, L.M., Fuller, K., Zeugolis, D.I., 2018. Influence of cross-linking method and disinfection/sterilization treatment on the structural, biophysical, biochemical, and biological properties of collagen-based devices. *ACS Biomater. Sci. Eng.* 4 (8), 2739–2747. <https://doi.org/10.1021/acsbiomaterials.8b00052>.
- Forsythe, J.S., Hill, D.J.T., 2000. The radiation Chemistry of fluoropolymers. *Prog. Polym. Sci.* 25 (1), 101–136. [https://doi.org/10.1016/S0079-6700\(00\)00008-3](https://doi.org/10.1016/S0079-6700(00)00008-3).
- Funabashi, K., Chino, K., Kikuchi, M., Horiuchi, S., Tsuchiya, H., 1991. Properties of a radioactive waste pellet package using cement-glass. *Nucl. Technol.* 96 (2), 185–191. <https://doi.org/10.1318/NT91-A34604>.
- Gaston, F., Dupuy, N., Marque, S.R.A., Barbaroux, M., Dorey, S., 2016a. One year monitoring by FTIR of γ -irradiated multilayer film PE/EVOH/PE. *Radiat. Phys. Chem.* 125, 115–121. <https://doi.org/10.1016/j.radphyschem.2016.03.010>.
- Gaston, F., Dupuy, N., Marque, S.R.A., Barbaroux, M., Dorey, S., 2016b. FTIR study of ageing of γ -irradiated biopharmaceutical EVA based film. *Polym. Degrad. Stab.* 129, 19–25. <https://doi.org/10.1016/j.polydegstab.2016.03.040>.
- Gaston, F., Dupuy, N., Marque, S.R.A., Gignes, D., Dorey, S., 2018. Monitoring of the discoloration on γ -irradiated PE and EVA films to evaluate antioxidant stability. *J. Appl. Polym. Sci.* 135 (18), 46114. <https://doi.org/10.1002/app.46114>.
- Gong, Y., Hu, S.-M., Yang, X.-L., Fei, J.-L., Yang, Z.-G., Shi, X.-Q., Xie, Y.-C., Guo, A.-H., Xu, J.-F., 2017. Comparative study on degradation of ethylene-propylene rubber for nuclear cables from gamma and beta irradiation. *Polym. Test.* 60, 102–109. <https://doi.org/10.1016/j.polymertesting.2017.03.017>.
- Goulas, A.E., Riganakos, K.A., Kontomina, M.G., 2004. Effect of ionizing radiation on physicochemical and mechanical properties of commercial monolayer and multilayer semirigid plastics packaging materials. *Radiat. Phys. Chem.* 69 (5), 411–417. <https://doi.org/10.1016/j.radphyschem.2003.08.013>.
- Hsiao, C.Y.-L., Chen, H.H., 2018. The contagious effects on economic development after resuming construction policy for nuclear power plants in coastal China. *Energy* 152, 291–302. <https://doi.org/10.1016/j.energy.2018.03.131>.
- ISO 11137-1, 2006. International Organization for Standardization, Sterilization of Health Care Products - Radiation - Part 1: Requirements for Development, Validation and Routine Control of a Sterilization Process for Medical Devices.

- Jahan, M.S., Wang, C., Schwartz, G., Davidson, J.A., 1991. Combined chemical and mechanical effects on free radicals in UHMWPE joints during implantation. *J. Biomed. Mater. Res.* 25 (8), 1005–1017. <https://doi.org/10.1002/jbm.820250807>.
- Kim, S., Lee, J., Shayan, F.L., Kim, S., Huh, I., Ma, Y., Yang, H., Kang, G., Jung, H., 2018. Physicochemical study of ascorbic acid 2-glucoside loaded hyaluronic acid dissolving microneedles irradiated by electron beam and gamma ray. *Carbohydr. Polym.* 180, 297–303. <https://doi.org/10.1016/j.carbpol.2017.10.044>.
- Klepac, D., Ščetar, M., Baranović, G., Galić, K., Valić, S., 2014. Influence of high doses γ -irradiation on oxygen permeability of linear low-density polyethylene and cast polypropylene films. *Radiat. Phys. Chem.* 97, 304–312. <https://doi.org/10.1016/j.radphyschem.2013.12.005>.
- Kurihara, T., Takahashi, T., Homma, H., Okamoto, T., 2011. Oxidation of cross-linked polyethylene due to radiation-thermal deterioration. *IEEE Trans. Dielectr. Electr. Insul.* 18 (3), 878–887. <https://doi.org/10.1109/TDEI.2011.5931077>.
- Lee, T.H., Wang, J., Wang, C.-H., 2002. Double-walled microspheres for the sustained release of a highly water soluble drug: characterization and irradiation studies. *J. Control. Release* 83 (3), 437–452. [https://doi.org/10.1016/S0168-3659\(02\)00235-3](https://doi.org/10.1016/S0168-3659(02)00235-3).
- Liu, B., Huang, W., Ao, Y.-Y., Wang, P.-C., An, Y., Chen, H.-B., 2018. Dose rate effects of gamma irradiation on silicone foam. *Polym. Degrad. Stab.* 147, 97–102. <https://doi.org/10.1016/j.polymdegradstab.2017.11.016>.
- Madera-Santana, T.J., Meléndrez, R., González-García, G., Quintana-Owen, P., Pillai, S.D., 2016. Effect of gamma irradiation on physicochemical properties of commercial poly(lactic acid) clamshell for food packaging. *Radiat. Phys. Chem.* 123, 6–13. <https://doi.org/10.1016/j.radphyschem.2016.02.001>.
- Martínez-Morlanes, M.J., Castell, P., Martínez-Nogués, V., Martínez, M.T., Alonso, P.J., Puértolas, J.A., 2011. Effects of gamma-irradiation on UHMWPE/MWNT nano-composites. *Compos. Sci. Technol.* 71 (3), 282–288. <https://doi.org/10.1016/j.compscitech.2010.11.013>.
- McKellop, H., Shen, F., Lu, B., Campbell, P., Salovey, R., 1999. Development of an extremely wear-resistant ultra high molecular Weight polyethylene for total hip replacements. *J. Orthop. Res.* 17 (2), 157–167. <https://doi.org/10.1002/jor.1100170203>.
- Montanari, L., Costantini, M., Signoretti, E.C., Valvo, L., Santucci, M., Bartolomei, M., Fattibene, P., Onori, S., Faucitano, A., Conti, B., et al., 1998. Gamma irradiation effects on poly(dl-lactide-co-glycolide) microspheres. *J. Control. Release* 56 (1–3), 219–229. [https://doi.org/10.1016/S0168-3659\(98\)00082-0](https://doi.org/10.1016/S0168-3659(98)00082-0).
- Nijssen, J.F., Schip, A.D., Hennink, W., Rook, D., van Rijk, P., Klerk, J.M.H., 2002. Advances in nuclear oncology: microspheres for internal radionuclide therapy of liver tumours. *Comput. Mater. Continua (CMC)* 9 (1), 73–82. <https://doi.org/10.2174/0929867023371454>.
- Nishad, P.A., Bhaskarapillai, A., Velmurugan, S., Narasimhan, S.V., 2012. Cobalt (II) imprinted chitosan for selective removal of cobalt during nuclear reactor decontamination. *Carbohydr. Polym.* 87 (4), 2690–2696. <https://doi.org/10.1016/j.carbpol.2011.11.061>.
- Oliani, W.L., Komatsu, L.G.H., Lugao, A.B., Parra, D.F., 2016. Thermal ageing and accelerated weathering of HMSPP: structural and morphological studies. *Macromol. Symp.* 367 (1), 18–23. <https://doi.org/10.1002/masy.201500135>.
- Portugal-Pereira, J., Ferreira, P., Cunha, J., Szklo, A., Schaeffer, R., Araújo, M., 2018. Better late than never, but never late is better: risk assessment of nuclear power construction projects. *Energy Policy* 120, 158–166. <https://doi.org/10.1016/j.enpol.2018.05.041>.
- Pourmand, P., Hedenqvist, M.S., Furó, I., Gedde, U.W., 2017. Radiochemical ageing of highly filled EPDM seals as revealed by accelerated ageing and ageing in-service for 21 years. *Polym. Degrad. Stab.* 144, 473–484. <https://doi.org/10.1016/j.polymdegradstab.2017.09.008>.
- Premnath, V., Harris, W.H., Jasty, M., Merrill, E.W., 1996. Gamma sterilization of UHMWPE articular implants: an analysis of the oxidation problem. *Biomaterials* 17 (18), 1741–1753. [https://doi.org/10.1016/0142-9612\(95\)00349-5](https://doi.org/10.1016/0142-9612(95)00349-5).
- Šarac, T., Quiévy, N., Gusarov, A., Konstantinović, M.J., 2016a. Influence of γ -irradiation and temperature on the mechanical properties of EPDM cable insulation. *Radiat. Phys. Chem.* 125, 151–155. <https://doi.org/10.1016/j.radphyschem.2016.03.024>.
- Šarac, T., Quiévy, N., Gusarov, A., Konstantinović, M.J., 2016b. The study of temperature and radiation induced degradation of cable polymers: a comparison between the mechanical properties of industrial and neat EPDM. *Procedia. Struct. Integr.* 2, 2405–2414. <https://doi.org/10.1016/j.prostr.2016.06.301>.
- Siepmann, J., Faisant, N., Akiki, J., Richard, J., Benoit, J.P., 2004. Effect of the size of biodegradable microparticles on drug release: experiment and theory. *J. Control. Release* 96 (1), 123–134. <https://doi.org/10.1016/j.jconrel.2004.01.011>.
- Song, C.X., Labhasetwar, V., Murphy, H., Qu, X., Humphrey, W.R., Shebuski, R.J., Levy, R.J., 1997. Formulation and characterization of biodegradable nanoparticles for intravascular local drug delivery. *J. Control. Release* 43 (2–3), 197–212. [https://doi.org/10.1016/S0168-3659\(96\)01484-8](https://doi.org/10.1016/S0168-3659(96)01484-8).
- Supersberger, N., Führer, L., 2011. Integration of renewable energies and nuclear power into north african energy systems: an analysis of energy import and export effects. *Energy Policy* 39 (8), 4458–4465. <https://doi.org/10.1016/j.enpol.2010.12.046>.
- Valente, T.A.M., Silva, D.M., Gomes, P.S., Fernandes, M.H., Santos, J.D., Sencadas, V., 2016. Effect of sterilization methods on electrospun poly(lactic acid) (PLA) fiber alignment for biomedical applications. *ACS Appl. Mater. Interfaces* 8 (5), 3241–3249. <https://doi.org/10.1021/acsami.5b10869>.
- Visakh, P.M., Nazarenko, O.B., Sarath Chandran, C., Melnikova, T.V., Nazarenko, S. Yu, Kim, J.-C., 2017. Effect of electron beam irradiation on thermal and mechanical properties of aluminum based epoxy composites. *Radiat. Phys. Chem.* 136, 17–22. <https://doi.org/10.1016/j.radphyschem.2017.03.032>.
- Xia, W., Xue, H., Wang, J., Wang, T., Song, L., Guo, H., Fan, X., Gong, H., He, J., 2016. Functionalized graphene serving as free radical scavenger and corrosion protection in gamma-irradiated epoxy composites. *Carbon* 101, 315–323. <https://doi.org/10.1016/j.carbon.2016.02.004>.
- Xia, W., Zhao, J., Wang, T., Song, L., Gong, H., Guo, H., Gao, B., Fan, X., He, J., 2017. Anchoring ceria nanoparticles on graphene oxide and their radical scavenge properties under gamma irradiation environment. *Phys. Chem. Chem. Phys.* 19 (25), 16785–16794. <https://doi.org/10.1039/C7CP02559A>.