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The expression revealing variation trend about radiation resistance of aromatic polymers serving in nuclear environment over absorbed dose

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HIGHLIGHTS

- Radiation resistance of aromatic polymers was studied.
- Percolation theory was employed to elucidate the critical phenomenon.
- A novel two-parameter radiation resistance model was proposed and verified.

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ABSTRACT

For polymeric materials applied in nuclear environment, the macroscopic properties usually remain unchanged after irradiation for several years or decades up to a threshold dose at which the deterioration of materials begins to take place. In this paper, the general radiation response of aromatic polymers is firstly reviewed and discussed. Then percolation theory is employed innovatively to elucidate the critical phenomenon over the service life for polymeric materials with high radiation resistance. For a better quantitative evaluation, a novel two-parameter radiation resistance model is proposed by the method of analogy between two nuclear-related phenomena. Six epoxy systems are employed from the published literatures to verify the novel model and the result shows that it is reliable and helpful in not only estimating the radiation damage over the service period but also multi-objective optimum design of polymeric materials.

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1. Introduction

Polymeric materials have extensive applications in nuclear power station, accelerator, aerospace, nuclear medicine devices and nuclear waste devices with their advantages of lightweight and excellent mechanical properties. Polymers suffer from rigorous environments such as radiation, high temperature, stress and corrosion, among which radiation causes the largest damage undoubtedly. Under high dose of radiation, energy deposits in materials and accumulates gradually, leading to changes in material microstructures. Macroscopic performance of materials deteriorates dramatically when total absorbed dose reaches a certain value (Birkina et al., 1967; Egusa et al., 1988; Gilfrich and Wilski, 1992; Ozdemir and Usanmaz, 2008; Sasuga, 1991), which will determine service lifetime of materials. Therefore, radiation resistance of polymeric materials becomes another significant research field in addition to radiation shielding performance.

Evaluation of radiation resistance of polymers can be accomplished from both microscopic and macroscopic views. From the micro perspective, the concentration of free radicals is the most commonly used indicator characterizing resistance to irradiation under the conditions of low temperature (usually 77 K), vacuum and low dose (Devasahayam et al., 2001; Heiland et al., 1996). Another evaluation way is accomplished in term of gas evolution (Hegazy et al., 1992a,b). The microscopic evaluation methods have some limitations in reflecting the eventual macro-properties of materials. While from the macroperspective, each material has a synthesis of physical and chemical properties such as mechanical performance, thermal properties, optical properties, electrical properties. A concept "determining characteristic indicator of radiation resistance" was introduced by Klinshpont et al. (1994), which means among all the properties only one determining and predominant indicator should be used for materials with different functional purposes. For instance, for materials with functions of structure, electrical engineering and thermal insulation, the

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determining characteristic indicators are tensile strength or bending strength, electrical conductivity and heat conductivity, respectively.

To obtain the radiation resistance of various polymers, numerous irradiation experiments have been carried out under different conditions in the last several decades, most of which show the variation tendency of macro-properties of polymers with the increased absorbed dose. As early as 1953, Oak Ridge National Laboratory (ORNL) had published data about the radiation stability of polymers (Bopp and Sissman, 1953). Swallow (1973) presented a listing including a wide range of available polymers at that time on radiation effects. Hanks and Hamman (1969) also showed the relative radiation resistance of various organic materials based upon changes in physical properties. Wundrich (1984) presented a review of the radiation resistance in the form of tables for several plastic and elastomeric materials and discussed the main factors that influence the radiation effects. Seguchi and Morita (1999) dealt with the data on radiation resistance and presented tables of radiation resistance of four groups of polymers: thermoplastics, elastomers, all aromatic polymers and composite materials. Hu et al. (2008) made a qualitative comparison on the radiation resistance among several radiation shielding materials. More extensively and intensively, European Organization for Nuclear Research (CERN) released a sequence of reports about the mechanical resistance of materials under different radiations, which nearly encompassed all the thermoset and thermoplastic polymers (Schönbacher and A.S.-I. 1979a, 1979b; Schönbacher and M.T., 1989; Tavlet and Schönbacher, 1998; Beynel and Schönbacher, 1982).

Among all the polymers, the aromatic polymers have been known to show greater radiation resistance in general than the aliphatic ones, attributed to the energy dissipation by resonance within the benzene ring structure (Brown and Odonnell, 1979; Campbell, 1981; Chen et al., 2009; Elsa et al., 1993; Hill et al., 1984). Thus the interest of this paper mainly refers to those aromatic materials with high radiation resistance applied in nuclear radiation environment, such as epoxy resins (Bonin et al., 1998; Laricheva, 2008; Rinaldi and Maura, 1993), polyimides systems (Apel et al., 2001; Coltman and Klabunde, 1982; Sasuga, 1991), phenolic resins (Gilfrich et al., 1991a, b), polystyrene (Spiegelberg et al., 1995; Tagawa, 1986), polyesters (Aliev et al., 2006) as well as their composites.

Although a volume of work has been devoted to the radiation effects of polymers and many experiments have been done regarding the radiation stability of polymers, little research has been carried on the theoretical description of radiation response over the service life and giving models of the radiation resistance of polymers, which will be the main two concerns of this paper. In our previous work (Hu et al., 2008), radiation shielding performance of polymer-matrix composites has been investigated adequately and quantitative evaluation of this performance has been finished by genetic algorithms combined with the Monte Carlo N-particle code. Once the radiation resistance model of polymers is established, the realization of multi-objective optimum design of polymeric materials with both excellent radiation shielding and excellent radiation resistance performances will be in the near future.

It should be stated that this paper just focuses on the performance of radiation resistance of polymers, thus the effect factors of temperature, oxidation, corrosion, etc. are beyond the research scope.

2. Response of polymers to radiation

Primarily, the response of polymers to radiation will be discussed systematically. For most polymeric materials with certain radiation resistance, their response to radiation presents a threshold feature (Birkina et al., 1967; Gilfrich and Wilski, 1992). Before the threshold dose, the macro-properties of materials are considered to be approximately unchanged, as shown in Fig. 1.

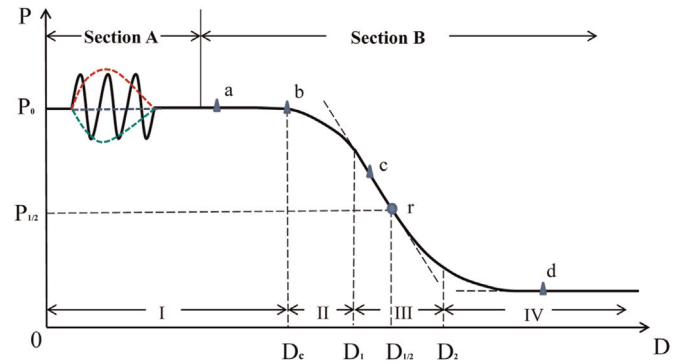

Fig. 1. Radiation response process of polymeric materials over their service life. Horizontal axis represents the accumulated dose by symbol D. Vertical axis represents the term "determining characteristic indicator of radiation resistance" mentioned earlier by symbol *P*. (For interpretation of the references to color in this figure, the reader is referred to the web version of this article.)

Fig. 1 describes the response of polymers to radiation over the service life in nuclear environment, which is divided into two parts (sections A and B) as a classification based on the absorbed dose. Section A refers to the relatively low dose exposure (usually in Gy or kGy) while section B refers to the rather high accumulated dose (usually in MGy).

In section A, ionizing radiation energy begins to deposit in polymers, leading to multiple reactions among which crosslinking and scission are the main two effects. The black wave line expresses the competition process of those two major reactions and such a competition results in two opposite phenomena. If crosslinking effect dominates, properties of polymers will gain an enhancement, as shown by the red broken line. On the contrary, the dominance of scission effect will give rise to the degradation of materials, as shown by the green broken line. Otherwise for some materials with high radiation insusceptibility, no obvious variation can be observed, as shown by the blue broken line. However, there are some polymers the radiation mechanisms of which are still not understood completely. Take epoxy resins for example, Burnay (1980) and Ngono-Ravache et al. (2001) found that crosslinking was the predominant degradation mechanism. The opposite opinion was come up with by other researchers (Davenas et al., 2002; Devanne et al., 2005a, 2005b; Djouani et al., 2013; Longieras et al., 2007; Vignoud et al., 2001a, 2001b) that the degradation of epoxy resin was apparently dominated by the scission effect. This contradiction has been explained by Longieras et al. (2006) who pointed out that the above epoxy materials investigated had different chemical structures and curing processes, and the irradiation conditions (like radiation source, temperature, dose rate, oxygen concentration) conducted were also quite different.

Section A shows the radiation effect of polymers, but one fact should be noted that no matter one polymer belongs to crosslinking or scission type, the mechanical properties must degrade with increase in the accumulated dose and finally become unusable in its service life. That means section B is actually the major concern of the study on radiation resistance of polymers. In section B, the classification of crosslinking and scission type is meaningless and will not be in consideration. Of great significance in practical engineering should be the critical dose at which the property mutation generates and the subsequent deterioration process.

According to the degree of radiation damage, four regions are marked out for further discussion. Regions I, II, III and IV stand for

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