Analysis of nuclear radiation damage effect on typical materials

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Abstract. In order to evaluate the effects of nuclear radiation on several typical materials, the mathematical models of radiation damage characteristics of three materials are established based on the analysis of radiation environment and damage forms. In order to simplify the calculation, the principles and indexes of damage estimation are determined according to the radiation damage process of three typical materials: polymer materials, high-energy explosives, and metal materials. The results show that the pure nuclear radiation environment has no obvious effect on the properties of three typical materials. It lays a theoretical foundation for radiation hardening of devices.

1. Introduction
For devices working in nuclear radiation environment, radiation defects such as vacancies, interstitial atoms and some complexes may become fatal problems and seriously affect their reliability. Because of the strong radioactivity of nuclear materials such as uranium and plutonium, the internal structure and functional materials of nuclear equipment are inevitably exposed to the radiation environment and sustained effects of radiation, which puts forward higher requirements for radiation stability of devices. In this process, whether the structure and performance of materials can meet the requirements is an important issue related to the safety and reliability of equipment. The materials inside nuclear equipment can be divided into three categories: metal materials, high-energy explosives, and polymer materials [1-3].

The study of materials is the basis to improve the performance parameters and reliability of devices, so it is very important to study the types and characteristics of defects introduced by particle irradiation in materials. With the development of atomic energy technology, the related research on radiation damage has been gradually carried out. The radiation damage of materials has been studied by means of radiation damage test and computer simulation by domestic and foreign scientific research institutions. The materials involved include metals, macromolecule materials, etc. (proton, neutron, and gamma-rays) There are significant differences in radiation damage modes of different materials. Dislocation is the main form of radiation damage of crystal materials, while the main form of radiation damage of polymer compounds is the breakage of molecular bonds. In the long-term experiments and simulations, for a given material, the researchers analyzed and summarized the relationship between environmental conditions (such as radiation dose rate, type of radiation) and radiation damage. Based on the experimental results, the mechanism and process of radiation damage were speculated. Neutron irradiation degrades the crystalline quality of materials.

With the development of science and technology, devices need to work effectively in more extreme...
environments, and the extreme conditions of the environment put forward higher requirements for the anti-irradiation characteristics of devices. In order to predict the device degradation in the radiation environment, it is necessary to understand the degradation mechanism of the device in the radiation environment. The radiation damage effect of materials is a complex process. This paper chooses the empirical method to evaluate the radiation damage of materials based on the typical radiation field data, which provides a reference for practice and validates it when the test conditions are available.

2. Analysis of nuclear radiation environment

Nuclear radiation environment includes nuclear explosion and nuclear power environment. Numerous high-energy particles and intense electromagnetic pulses generated by nuclear weapon explosion are called nuclear explosion environment. Nuclear power plants, submarines and carriers equipped with nuclear energy will also generate certain nuclear radiation around them, which is called nuclear power environment. High-energy particles in nuclear radiation environment mainly include fast neutron flow, high-energy electron flow, gamma-ray, X-ray, alpha-ray, beta-ray and so on. The latter two are easily absorbed by the atmosphere with short range and little influence on electronic equipment and components. Our main concern is fast neutrons and gamma rays, which have higher fluxes and doses than the space environment.

In "Monte Carlo calculation of radiation field inside typical plutonium core abroad", taking the relevant equipment of the United States as an example, Monte Carlo method is used to simulate the radiation field inside plutonium core. The variation law of photon and neutron influence rate with radius is described, and the representative points are analyzed by energy spectrum. In this paper, the photon and neutron energy deposits in typical materials are calculated by MCNP (Monte Carlo N Particle Transport Code) program. As shown in table 1, under this assumption, the radiation damage of typical materials is estimated. Silicone rubber, HMX and steel are selected as macromolecule and high molecular weight respectively.

| Material    | Energy deposition/(Jg⁻¹s⁻¹) |
|-------------|-----------------------------|
|             | Photon/10¹⁹ Neutron/10²¹      |
| Silicon rubber | 1.35 6.30                  |
| HMX         | 1.04 4.85                   |
| Steel       | 2.35 10.9                   |

3. Analysis of nuclear radiation damage forms

According to the different interaction between energetic particles and semiconductor materials and devices, the radiation effect can be divided into ionization effect and displacement effect. The former generates electron holes by breaking the energy band balance, while the latter causes some atoms to move away from the normal lattice position, leaving vacancies at the same time. Ionization damage generally requires much less energy than displacement damage. Due to the different internal structure of the three materials, the damage process and mechanism are different in the radiation environment.

Heavily charged particles (such as protons) pass through the material and interact with electrons in the material, losing energy bit by bit. Once enough energy is lost to excite an electron, the particle loses energy through a nuclear collision. When the particle is slow, it can capture electrons to form neutral atoms, such as protons into hydrogen atoms. The slowdown of heavy charged particles is almost entirely due to the Coulomb scattering of atom electrons, so the slowdown is continuous. Because the heavy charged particles are relatively large, it is difficult for the particles to be diverted by the atomic and nuclear electrons of the material. Usually, the trajectory of the particles is a straight line. Neutral particles (such as neutrons) can release directly ionized particles or undergo nuclear transformation. The basic interaction between neutrons is scattering and absorption, which includes capture and fission. The neutron collides with the material nucleus, and the irradiation defect is introduced. Sometimes the
material is activated, resulting in transmutation doping. For the electron irradiation with large energy, because the electron mass is relatively small.

3.1. The basic process of radiation damage of high energy explosives
The effect of radiation on high-energy explosives can be attributed to the breaking of molecular bonds: particles pass through materials to produce secondary electrons, secondary electrons cause the breaking of molecular bonds due to ionization effect, and the final effect is the decomposition of explosives. The weakest bond of an explosive molecule usually breaks first. A series of subsequent reactions will occur after the breaking of molecular bonds, and these reactions are accompanied by exothermic phenomena as shown in figure 1.

![Figure 1. Compton effects.](image1)

![Figure 2. Structure of the polymer materials.](image2)

3.2. The basic process of radiation damage of polymer materials
The long chain structure of polymer materials makes a small amount of radiation events lead to a large proportion of molecular damage as shown in figure 2. Therefore, radiation damage to polymer materials is more significant in theory. Crosslinking, degradation and gas generation are the main changes of polymer materials in radiation field. Crosslinking and main chain breaking have the greatest impact on polymer materials. Crosslinking and degradation of polymer materials usually occur simultaneously in the radiation field.

3.3. The basic process of radiation damage of metal material
The atoms in the metal material are arranged regularly and repeatedly, thus forming different lattices. In the process of penetrating metal materials, the incident particles transfer part of the energy to the lattice atoms in the form of elastic collision. The collision of radiation particles into metal materials and lattice atoms will trigger a series of collision processes, which can be divided into primary and secondary processes. The primary process produces primary dislocation atoms, and the secondary process is cascade collision process.

Based on the analysis of radiation damage process of three different materials, the target of radiation damage estimation is determined.

- High-energy explosives mainly study the effect of radiation on molecular decomposition.
- The influence of radiation on logarithmic mean molecular weight (LMW) of polymer materials was investigated.
- The influence of radiation environment on dislocation density is mainly investigated for metal materials.

4. Theoretical model of material damage
4.1. Damage model of high energy explosives
The basic process of radiation damage of high-energy explosive molecule is the generation of secondary electrons by radiation passing through the material. The breakage of molecular bond caused by ionization effect leads to the breakage of molecule. The calculation of radiation damage focuses on the decomposition rate of explosive molecules. According to the characteristics of radiation damage effect
of high-energy explosive molecules, Yang et al put forward four hypotheses to simplify the calculation, and gave the formula for calculating the decomposition rate, as shown in formula (1)[4].

$$\nu = \left( I_n E_n + I_p E_p \right) / E_0$$

(1)

Here, $\nu$ is the decomposition rate of explosive, mol•s$^{-1}$; $I_n$ and $I_p$ are the intensity of neutron and photon, $E_n, E_p$ are the average energy deposition of neutron and photon in explosive, J; $E_0$ is the activation energy of substance, J•mol$^{-1}$.

In order to calculate the decomposition rate of explosives directly, formula (1) is adjusted as shown in formula (2).

$$\eta = \frac{e_n + e_p}{M \cdot E_0}$$

(2)

where $\eta$ is the decomposition rate of explosive, i.e. the proportion of decomposed molecules to the total number of molecules; $e_n, e_p$ are the deposition of neutron and photon energy per unit mass explosive, J•kg$^{-1}$; $M$ is the molar mass of explosive, kg•mol$^{-1}$.

4.2. Damage model of polymer materials

Degradation and crosslinking are the most fundamental changes of polymer materials in the radiation field, and these two processes usually occur simultaneously. In the process of radiation degradation, no monomer molecule is usually formed, and the main chain breaking during radiation degradation usually results in shorter molecular chains, which reduces the average molecular weight. Based on the above experimental results, the computational model is simplified as follows:

- Radiation degradation occurs only when radiation acts on polymer materials.
- The action of rays on molecular chains only breaks the weakest chemical bonds in the main chain.

Under the above assumptions, the number of main chain breaks per mole of material can be calculated by formula (3).

$$N = \left( U_{m,n} + U_{m,p} \right) \cdot N_A / U_{m,0} ,$$

(3)

where $N$ is the number of breaks in the main chain; $U_{m,n}$ and $U_{m,p}$ are the energy per mole of neutrons and photons, J•mol$^{-1}$; $U_{m,0}$ is the bond energy of the weakest chemical bond in the main chain;, J•mol$^{-1}$, $N_A$ is the Avogadro constant, 6.02×10$^{23}$ mol$^{-1}$. Assuming that the number-average molecular weight of irradiated polymer material is $M_0$, the number-average molecular weight $M$ after irradiation can be calculated by formula (4).

$$M = M_0 \left[ 1 + \frac{M_0}{N_A} N \right]$$

(4)

4.3. Damage model of metal material

Metal materials belong to crystals, which will produce crystal defects after being irradiated. The dislocation density caused by incident particles can be used to measure the radiation damage caused by incident particles.

The number and energy spectrum of primary dislocation atoms in the differential energy region can be calculated according to the total flux of the incident particle, the energy spectrum and the energy transfer cross section when the particle collides with the lattice atom. The relationship between the primary dislocation atoms with energy $E$ and the number of dislocation atoms $n(E)$ is established, and the total number of dislocation atoms is calculated.

Dislocation cascade is usually calculated by the Kinchin-Pease model. Cascade collisions are initiated by primary dislocation atoms with energy $E$, and finally $n(E)$ dislocation atoms are generated.
In any intermediate stage, the moving atom generates the same number of dislocation atoms as the original primary dislocation atom, so \( n(E) \) is conservative, as shown in equation (5).

\[
n(E) = n(E-T) + n(T)
\]

The primary dislocation atom with energy \( E \) is transferred to the stricken atom. The probability of energy transfer between \( T \) and \( T+dT \) is \( K(E,T)dT \), multiplied by differential cross section \( \frac{K(E,T)}{\sigma(E)}dT \) on both sides of equation (5), and obtained from 0 to \( E \) integral.

\[
\bar{n}(E) = \int_0^E \left[ n(E-T) + n(T) \right] \frac{K(E,T)}{\sigma(E)}dT
\]

In the formula, \( \sigma(E) \) is the total cross section. The atomic potential of Kinchin-Pease model is calculated by rigid sphere model. For rigid sphere collision,

\[
\frac{K(E,T)}{\sigma(E)} = \frac{1}{E}
\]

From the transformation of formulas (6) and (7), the average number of dislocation atoms \( \bar{n}(E) \) produced by primary dislocation atoms with energy \( E \) can be calculated by formula (8).

\[
\bar{n}(E) = \frac{2}{E} \int_0^E \bar{n}(T)dT
\]

Research shows that there exists a lower energy value \( E_{\text{low}} \). When the energy \( E_{\text{low}} \) is reached, the moving neutral atom cannot collide with the stationary atom in the solid, resulting in ionization. In order to maintain some positive charges, the minimum energy of the moving particle must be about its mass number \( M \) times keV. If the energy of the primary dislocation atom is higher than \( E_{\text{low}} \), the dislocation atom can not be produced until the energy loss of the primary dislocation atom is reduced to \( E_{\text{low}} \). If the energy of the primary dislocation atom is lower than \( E_{\text{low}} \), the atomic collision will be considered instead of the electron blocking.

The average number of dislocation atoms produced by incident particles with energy \( E \) can be obtained by solving equation (9) considering the energy loss caused by electron blocking.

\[
\begin{cases}
\bar{n}(E) = 1 & E_d < E \leq 2E_d \\
\bar{n}(E) = \frac{E}{2E_d} & 2E_d < E \leq E_{\text{low}}
\end{cases}
\]

In the formula, \( E_d \) is the off-position threshold energy. Kinchin-Pease model is a commonly used model for cascade calculation of dislocations. The theoretical values of the number of dislocation atoms radiated by electrons, protons and neutrons are basically in accordance with experimental values, which are got by the model. The number of dislocation atoms caused by a single incident particle can be calculated by the Kinchin-Pease model. The total number of dislocation atoms in the region can be solved by summing the number of dislocation atoms caused by all incident particles. To further simplify the calculation process, the following two hypotheses are proposed:

- All incident particles collide with atoms in the region, and the energy deposits completely in the region.
- When the incident particle collides with the atom, the energy is completely transformed to the kinetic energy of the primary dislocation atom.

The above two assumptions maximize the energy deposition of incident particles and the kinetic energy of primary dislocation atoms. The total number of dislocation atoms calculated from these assumptions will be larger than the actual situation, and the maximum dangerous situation is considered in the estimation process. Based on the above assumption, the total number of dislocation atoms can be
expressed as:

\[
N_{\text{loc}} = \int_0^\infty \pi(E) \Phi(E) \, dE
\]  

(10)

Among them, \( \Phi(E) \) is the energy spectrum of the incident particle. The total number of dislocations per unit cross-sectional area is the dislocation density.

5. Estimation of radiation damage

5.1. Radiation damage of polymer materials

Molecular weight directly affects the properties of polymer materials. With the decrease of molecular weight, the tensile strength, compression set rate and other performance indicators will deteriorate to varying degrees. Chen et al found that when the number-average molecular weight difference of silicone rubber and other polymer materials is greater than 10%, the density, elongation, compression modulus and other performance indicators begin to change significantly. Therefore, a 10% reduction in number-average molecular weight is taken as the failure critical value [5].

Taking silicone rubber as an example, the radiation damage of polymer materials was estimated. The excellent properties of silicone rubber are related to the chemical structure of polysiloxane, whose simplified unit structure is shown in figure 3. The number-average relative molecular weight is between \(10^4\) and \(10^5\). The bond energy of the silicone-oxygen bond on the main chain is 451 kJ· mol\(^{-1}\).

![Figure 3. Unit structure of silicone rubber.](image)

According to the model assumption, the energy deposited by the incident particles is completely used to destroy the silicon oxygen bonds on the main chain. The average energy deposition in the silicon foam in unit time of the photon is \(1.35 \times 10^{-19}\) J· g\(^{-1}\) and the neutron is \(6.30 \times 10^{-21}\) J· g\(^{-1}\). The average molecular weight of silicone rubber is between \(10^4\) and \(10^5\), assuming that it is \(5 \times 10^4\). Taking \(E_n = 6.75 \times 10^{15}\) J· mol\(^{-1}\), \(E_p = 3.15 \times 10^{16}\) J· mol\(^{-1}\), \(E_0 = 451\) kJ· mol\(^{-1}\), according to the formula (1), the breaking frequency of the main chain of per mole silicone rubber foam in unit time is:

\[
N = \frac{6.75 \times 10^{-15} + 3.15 \times 10^{-16}}{451 \times 1000} \cdot N_\Lambda = 9391.2
\]

Relative molecular weight of silicone rubber stored for 1 year is:

\[
M = \frac{50000}{1 + \frac{50000}{N_\Lambda} \times 1.56 \times 10^{-20} \times 3600 \times 24 \times 365 N_\Lambda} = 5.00 \times 10^4 \text{ g} \cdot \text{mol}^{-1}
\]

The relative variable of relative molecular weight is:

\[
\Delta M = \frac{|M - M_0|}{M_0} = 2.46 \times 10^{-8}
\]

The calculation results show that when the influence of radiation on the number average molecular weight of silicon foam in radiation environment is considered separately, the relative change amount and the absolute change amount are all very small. In most cases, the main chain of macromolecule
compounds is a carbon chain, and the bond energy does not change significantly. From this mechanism, the number-average molecular weight will not have a significant impact.

5.2. Radiation damage of high energy explosives

For high-energy explosive molecules, spontaneous decomposition is a common phenomenon. The decomposition depth is related to the density and detonation velocity of explosive. When the decomposition depth of explosive reaches 0.1%, its performance will change significantly. Therefore, the decomposition depth of 0.1% is usually regarded as the failure critical value [6]. In this paper, the radiation damage of HMX is estimated by using the decomposition depth as an index. HMX is the explosive with the best comprehensive military performance. Its chemical structure is shown in figure 4.

![Molecular structure of HMX](image)

**Figure 4.** Molecular structure of HMX.

In radiation environment, the average energy deposition a year is $3.28 \times 10^{-12}$ J·g$^{-1}$ for photons and $1.52 \times 10^{-13}$ J·g$^{-1}$ for neutrons. The activation energy $E_0$ of HMX is 220.5 kJ·mol$^{-1}$. The radiation decomposition rate of HMX is calculated by substituting data into formula (2).

$$
\eta = \frac{3.28 \times 10^{-12} + 1.52 \times 10^{-13}}{220.5 \times 1000} = 4.60 \times 10^{-13}
$$

The decomposition rate of HMX radiation stored for one year is calculated from the decomposition rate, which is $4.60 \times 10^{-13}$. The results show that the decomposition rate of HMX is very low when the effect of radiation on the decomposition of HMX is considered separately in the radiation field. Taking the decomposition depth as the index of radiation damage, the radiation damage of high-energy explosive was investigated. The results show that the rate of decomposition of explosive molecule caused by radiation is very low. In practice, the interaction between radiation and matter has energy loss, and the ionization effect of neutrons is very small, that is, the decomposition rate of molecules in the real radiation environment is lower than that estimated. Therefore, the influence of radiation on high-energy explosives can be neglected.

5.3. Radiation damage of metal materials

Taking steel as representative, the damage of metal materials in radiation field is estimated. The total number of dislocation atoms produced by a single incident particle can be calculated by formula (10). In the most dangerous case, the minimum dislocation threshold energy is 50 eV [7]. The energy loss caused by electronic blocking is treated as equation (9), $E_{\text{low}} = 56$ keV. By substituting the above data into equation (10), the dislocation atoms generated by a single incident particle are as follows:

$$
\begin{align*}
\pi(E) &= \begin{cases} 
\frac{E}{0.05} & \text{if } E < 56 \text{ keV} \\
1120 & \text{if } E \geq 56 \text{ keV}
\end{cases}
\end{align*}
$$

The collision spectra of photons and neutrons in this region are calculated by MCNP program, which
are as shown in figure 5.

![Figure 5. Photon and neutron energy spectra of collisions in steel. (a) Photon Energy Spectra and (b) Neutron Energy Spectra.](image)

It is further assumed that all radiation particles in the region collide with beryllium atoms in the lattice, resulting in atomic dislocations. The total number of dislocation atoms can be calculated by formula (10). By substituting the energy spectrum data, the total number of dislocation atoms caused by photons and neutrons in the spherical region with a diameter of 1 cm is obtained can be got, where the number of photons is 7.54×10^5 cm^-2 s^-1 and the number of neutrons is 2.59×10^3 cm^-2 s^-1.

The atomic dislocation density caused by radiation in steel material is 2.37×10^12 cm^-2 under storage for one year. The direct result of photon and neutron radiation is that a large number of dislocation atoms and vacancies are generated in the bombarded material. These micro lattice defects will affect the macroscopic properties of the material. Therefore, dislocation density is used as an index to investigate the radiation damage of steel materials.

At present, no research results accurately describe the quantitative relationship between dislocation density and mechanical properties of materials, but researches show that the dislocation density of steel and other metal materials caused by hot working is 10^{14} cm^-2, and there is no significant change in mechanical properties of materials under these conditions. It is believed that when the dislocation caused by radiation is considered separately in radiation field environment, the properties of steel materials do not change significantly.

6. Conclusions
Under the assumption of maximum danger, a mathematical model was established and the effects of radiation environment on the main properties of three typical materials were estimated. The estimated results show that the HMX radiation decomposi
tion rate is 4.60×10^{-13}, the relative change of molecular weight of polymer material is 2.46×10^{-8}, which is much smaller than the index of performance change. The dislocation density of metal crystal caused by radiation is about 2.37×10^{12} cm^-2, which is only equivalent to that caused by thermal processing of metal material. In conclusion, it is believed that the pure radiation environment has no obvious effect on the properties of three typical materials. The results are in good agreement with experimental analysis results in the study of radiation effects.

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