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Research Progress in Enhancing the Radiation Resistance of Polymer Material

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Abstract
Compared with the traditional method of sterilization, radiation sterilization has multiple advantages. The polymer will occur oxidative degradation in radiation environment, resulting in yellow coloration, mechanical properties decrease and other metamorphic. Enhancing the radiation resistance of polymer is an issue worthy of attention. Researchers mainly enhance the radiation resistance of polymer by adding antioxidant, heat stabilizer, light stabilizer and so on. In this review, the mechanism and the synergistic or antagonistic effect between all kinds of stabilizers enhancing the radiation resistance will be introduced.

1. Introduction

In recent years, people's attention to health care is growing as the continuous improvement of the living level of consumption, leading to the increasing of the investment on health care and the promoting of the modification of the medical equipment research. People begin to focus on appropriate physical or chemical means to kill or remove microorganisms. The traditional ways of sterilization such as moist heat sterilization, dry heat sterilization, sterilizing gas does not apply to the poor thermal stability of polymers. What is worse, ethylene oxide sterilization will produce toxic residues [1].

However, the sterilisation of vital single-use medical devices is carried out by electron beam or γ irradiations, which performs to be safer, cleaner and faster. On the other hand, the degradation and oxidation reaction of the polymer materials caused by the action of high energy radiation can make the material yellow and the performance destroyed.. [2] To solve the problem of discoloration of the irradiation of polymers, the usual way is adding different antioxidants, light stabilizers and other substances to increase stability and antioxidant properties of the materials, enhancing resistance to the electrochromic properties of the radiation.

There are a lot of researches having been done to solve the problem of polymer in application in poor weather resistance, poor stability. However, study devoting to enhancing polymer material resistant of the electrochromic properties of the radiation research is still relatively small, which is a new direction for the concern and research.

2. Research Status of Polymer Antioxidant for Irradiation Sterilization

2.1. Main Types of Antioxidant and Its Radiation Resistant Mechanism

In the process of synthesizing polymers, the molecular chains and oxygen molecules of the polymer have automatic reaction, which causes the material to be aged. The addition of antioxidants is the most effective method to prevent aging of polymers. In many researches, due to the antioxidant mechanism and material resistant to irradiation performance of the modified mechanism of the same, scholars pay attention to the addition of an antioxidant to improve material irradiation resistance. Common antioxidants are divided into: hindered phenolic antioxidant and phosphite ester antioxidant, thiol antioxidant and composite antioxidant. As antioxygen oxidation process of different antioxidants on the basis of structure and performance for different materials, and some antioxidants and antioxidant between exist synergism. [2]

The process of oxidation is as follows:

(1) an initial step, in which R^* are formed:

$$RH \xrightarrow{\Delta} R^* + H^*$$

$$\mathrm{RH} + O_2 \xrightarrow{\Delta} R^* + HO_2^*$$

(2) a propagation step, during which ROO^* and $ROOH$ is produced:

$$R^* + O_2 \xrightarrow{\Delta} ROO^*$$

$$ROO^* + RH \xrightarrow{\Delta} ROOH + R^*$$

(3) a disproportionation step, during which $ROOH$ are decomposed:

$$ROOH \xrightarrow{\Delta} RO^* + HO^*$$

$$ROOH + RH \xrightarrow{\Delta} RO^* + R^* + H_2O$$

$$2ROOH \xrightarrow{\Delta} RO^* + ROO^* + H_2O$$

$$RO^* + RH \xrightarrow{\Delta} ROH + R^*$$

(4) a termination step, in which R^* are deactivated:

$$2ROO^* \rightarrow ROOR + O_2$$

$$2ROO^* \rightarrow 2RO^* + O_2$$

$$ROO^* + R^* \rightarrow ROOR$$

$$2R^* \rightarrow R - R$$

Antioxidants inhibit oxidation mainly through several mechanism. Trapping and scavenging free radical mechanism on which antioxidants could have reaction with the free radical produced in the process of oxidation reaction, interrupting free radical chain reaction. Antioxidants with the role of this include carbon black, some nitroso compounds, stable free radical compounds etc. The electron donating mechanism interrupt the chain reaction of the free radical by electron donating, which contains GW-608, GW-650, etc. The proton giving mechanism means proton is given by the antioxidants to the destroy chain reaction, and the representative of these are hindered phenoland aromatic amines. The mechanism of hydrogen peroxide decomposition refers to decomposition of hydrogen peroxide by ionic mechanism, preventing the accelerated reaction of free radical caused by hydrogen peroxide. Common antioxidants of this type include phosphite ester and thiol antioxidant. There are antioxidants reducing the activity of metal ions, because the metal ions can be achieved by one electron oxidation - reduction reaction, to accelerate the decomposition of hydrogen peroxide free radical, to accelerate the polymers from oxidation. Antioxidant complex the harmful metal ion, reducing catalytic activity of the ions, so as to enhance the oxidation resistance of the material. This type of antioxidant is mainly oxime organic matter.

The mechanism of inhibiting oxidation is as follows:

(1) Trapping and scavenging free radical mechanism

$$\mathrm{Ar}^* + RO^* \rightarrow ArOR$$

$$\mathrm{Ar}^* + R^* \rightarrow ArR$$

(2) The electron donating mechanism:

$$ROO^* + Ar - \ddot{N} - R_2 \rightarrow ROO{:}^- Ar - N - R_2^+$$

(3) The proton giving mechanism:

$$Ar_2 - NH + ROO^* \rightarrow ROOH + Ar_2{-}N^*$$

$$Ar_2 - N^* + ROO^* \rightarrow Ar_2 - NOOR$$

$$Ar - OH + ROO^* \rightarrow ROOH + Ar_2 - O^*$$

$$Ar_2 - O^* + ROO^* \rightarrow ROO - O - Ar$$

(4) The hydrogen peroxide decomposition mechanism:

$$ROOH \rightarrow RO^* + {}^*OH$$

$$ROOH \rightarrow ROO^- + {}^+H$$

(5) The metal ions deactive mechanism:

$$\mathrm{ROOH} + M^{\mathrm{m}+} \rightarrow RO^* + M^{(m+1)^+} + OH^-$$

$$\mathrm{ROOH} + M^{(\mathrm{n}+1)+} \rightarrow ROO^* + M^{\mathrm{n}^+} + H^+$$

Among these types of antioxidants above, usually hindered phenolic antioxidant act to be the main antioxidant. The main mechanism is trapping and scavenging free radicals, interrupting the oxidation process in the chain growth. Hindered phenolic antioxidant include single phenol, bisphenol, polyphenol, nitrogen heterocyclic polyphenols. Single phenol and bisphenol antioxidants with lower

molecular weight have the volatility and the migration of the big shortage. Polyphenol antioxidants are current leading products with high molecular weight, good compatibility with polymer and excellent antioxygen performance.

Phosphite antioxidant and thio auxiliary antioxidant are auxiliary antioxidant, through whose changes in its molecular phosphorus or sulfur atom valence to decompose the hydrogen peroxide into low molecular activity. According to the classification of molecular structure, it can be mainly classified as thioester antioxidant, thiobisphenol antioxidant and sulfide type phenol etc.

Composite antioxidant is the combination of different main antioxidants and auxiliary antioxidant., or different types of antioxidants. The synergistic effect of different antioxidants achieve a good antioxidant effect. [14]

2.2. Research Status and Development of Antioxidant

Bi Dazhi [3] studied ternary and binary composite antioxidant on high density polyethylene anti-aging effect. The anti aging effect of ternary system of hindered phenolic Irganox 1010 and phosphite ester Irganox 168 mixed binary system at 230 DEG C is higher than binary system of Irganox 1010, Irganox 168 and antioxidant HP136.The result on 260 DEG C is on the contrary.

Tao Yuanyuan [4]further studied the effect of antioxidant and light stabilizer blends on the properties of high density polyethylene. It is found that the binary mixed system of Irganox 1010 and Irganox 168with ultraviolet absorbent Uv326 and light stabilizer hindered amine 770 compound composite has greatly improved on polyethylene photooxidation aging performance.

Xia Hongbiao[5]studied on the antioxidant of PVC anti aging discoloration. Hindered phenolic antioxidant and metal passivator compound can effectively prevent the oxidative degradation reaction of PVC..

Chen Jiabo [6]studied the influence of Irganox 1010 on polyethylene photo degradation membrane structure and performance. obtained the conclusion that Irganox 1010 can inhibit the photo oxidation degradation of polyethylene.

Wu Peng [7] studied the hindered phenolic Irganox 1010, phosphate Irganox 168 of LLDPE alone can enhance the anti ageing performance, hindered phenol, phosphite antioxidant and hindered amine light stabilizer has a synergistic effect.

Wang Hui Liang [8] studied different antioxidants on the irradiated PP induced color performance impact, found under the same irradiation dose, yellow degree minimum when adding Irganox 1076,than comes thio antioxidant PS-802, yellow degree maximum when adding Irganox 168, which confirmed that the irradiation modification ability of phenolic antioxidants is the strongest.

3. Research Status of Polymer Heat Stabilizer for Irradiation Sterilization

3.1. Main Types of Heat Stabilizer and Its Radiation Resistant Mechanism

Common heat stabilizers of polymer are lead salts, metal soaps, organic tin, rare earth and composite stabilizers. Calcium zinc stabilizer of metal soap can be used to improve the radiation resistance of PVC. [9] The main reason of PVC irradiation induced color is that it occurs intramolecular removal reaction of hydrogen chloride, after high energy radiation, beside the degradation and oxidation reaction, which results in the formation of peroxides, discoloration of polymers. Zinc soap can inhibit the initial coloration of vinyl chloride by esterification of polyvinyl chloride on polyvinyl chloride molecules. Calcium soap can not exchange vinyl chloride, but it can capture the hydrogen chloride generated from the reaction of polyvinyl chloride which is favorable for long-term stability. Therefore, the overall performance of the calcium zinc composite stabilizer is initially inhibited and the long-term stability is good. [10]

The mechanism of inhibiting oxidation is as follows:

(1) Zinc soap and Calcium soap react generating complex as figure 1.

(2) The ester group in the complex is continuously substituted by chlorine atoms on the PVC molecule chain, thus inhibiting PVC generating HCl as figure 2.

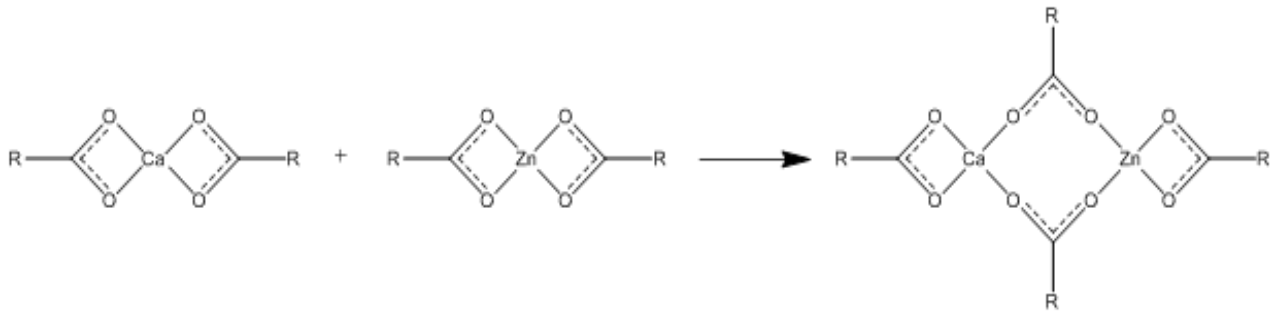

Figure 1. *Zinc soap and Calcium soap react.*

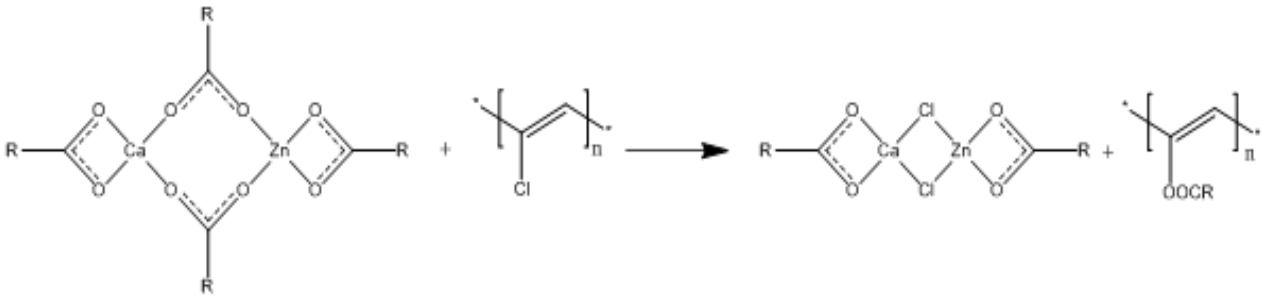

Figure 2. *PVC and calcium zinc composite stabilizer react.*

3.2. Research Status and Development of Heat Stabilizer

There are few studies on the heat stabilizer used to improve the radiation resistance of the material. Some researchers have studied the effect of calcium and zinc stabilizer on the weathering resistance, and the photo stability of the material is indirectly corroborated.

Xu Peng [11] explored the effect of the amount of calcium zinc stabilizer on the improvement of weather resistance. When calcium zinc stabilizer added to a certain amount, the effect of the aging properties was small.

Peng Zhaorong [12]Studied the effect of different stabilizers on the PVC radiant discoloration. The structure of the spectrometer was characterized by ultraviolet spectrometer and Fu Liye transform infrared spectrometer. The results showed that the calcium stearate / stearic acid zinc complex stabilizer, epoxy soybean oil, phosphite and light stabilizer can improve PVC radiation resistant photochromic properties. Among them, the modification effect with calcium stearate / zinc stearate appeared to be the best, than comesepoxy soybean oil, phosphite and light stabilizer worse. In the same dose, adding calcium / zinc stearate composite stabilizer PVC had excellent resistance to radiation discoloration performance.

4. Research Status of Polymer Light Stabilizer

4.1. Main Types of Polymer Light Stabilizer and Its Radiation Resistant Mechanism

Light stabilizer is a stabilizer used to prevent the material degeneration due to the photochemical effect of material, which is the main stabilizer for improving the radiation resistance of materials. Light stabilizer include UV absorber, quencher, light shielding agent and hindered amine light stabilizer. [13-16]

Ultraviolet light absorption type light stabilizer can convert light energy into heat energy by its molecular structure to avoid the light oxidation of materials because of direct absorption of light, which include two benzophenone and three benzotriazole etc.

The quenching agent is similar to UV absorber to avoid the oxidation of materials by transferring light energy. What is different, the quenching agent react with the excited functional groups of high energy and high chemical reactivity of the energy produced by the light energy, transferring the energy of functional groups. In industry, the quenching agent is a complex of two valence nickel which have certain harm to human body. In recent years, the use of quantity is gradually reduced.

The light shielding agent includes carbon black, titanium dioxide, zinc oxide, etc. The effect of light shielding agent on the reflection and shielding of the light energy is achieved to prevent the photo oxidation..

The application of the optical shielding agent, ultraviolet absorbent and quenching agent in the 60's in twentieth Century was industrialized, when HALS became industrial production until the mid 70's.However, the number of varieties and the development speed of hindered amine light stabilizer is higher than that of other light stabilizer.

Hindered amine light stabilizer is a kind of steric effect of organic amine compounds whose 2,2,6,6- four methyl -4-piperidyl as parent structure. Under the action of light energy, the chain reaction of the polymer occurs. Hindered amine light stabilizer will be oxidized by the peroxide and oxide produced by the chain reaction, producing nitrogen oxygen free radicals, reacting with the hydrogen peroxide and free radicals, and capturing the reactive products, thus eliminating the hydrogen peroxide and free radicals in the materials. The nitrogen oxygen free radicals also have the ability to capture the active free radical of polymer. After the capture, nitrogen oxygen free radicals will return to the original nitrogen free radical state, and can capture hydrogen peroxide and free radicals again. At the same time, hindered amine light stabilizer has the ability to quench singlet oxygen. Although the quenching efficiency of hindered amine in amine state is very low, it can be greatly improved when Hindered amine is oxidized to the nitrogen oxygen free radicals. This reaction makes the singlet oxygen from the excited state to the ground state, interfering with the light oxidation reaction before the chain initiation reaction.

The mechanism of hindered amine light stabilizer to inhibit the oxidation is as follows:

(1) Decomposition reaction of hydro peroxides by hindered amine as figure 3.

(2) Radical-trapping mechanism of hindered amine as figure 4.

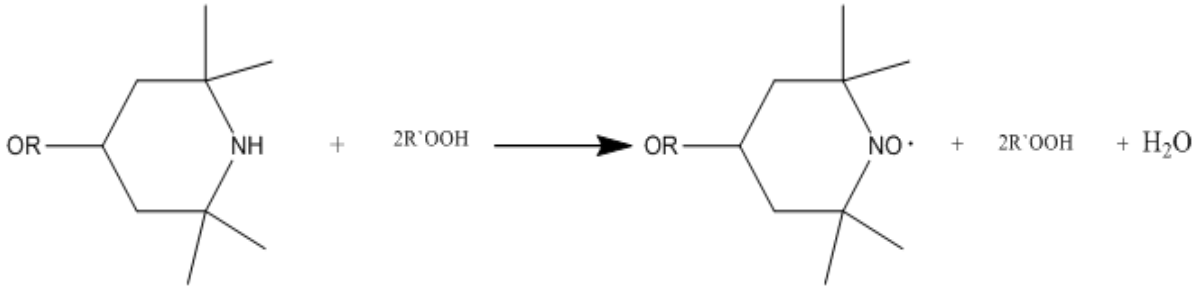

Figure 3. *Hydro peroxides and hindered amine react.*

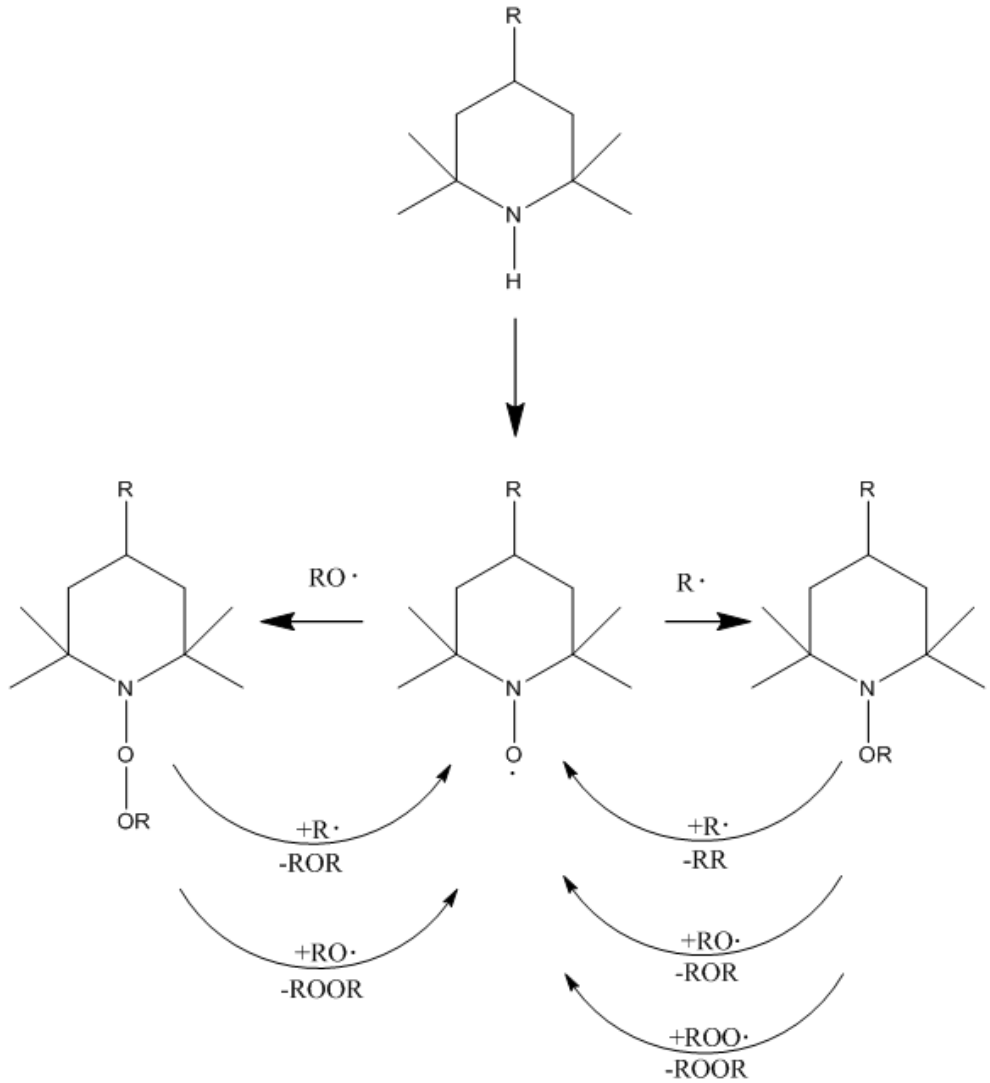

Figure 4. *Radical-trapping mechanism of hindered amine.*

4.2. Research Status and Development of Polymer Light Stabilizer

Entering the 90's in twentieth Century, the scholars discovered that HALS not only had good light stability, but also had some thermal stability. Qi Zhihao[17] studied on anti aging properties of PP fabric thermal with the HALS stabilizer (Ciba Chimassorb994).

Results show that anti aging performance with phenolic antioxidants, HALS light stabilizer with composite melt spinning made PP mesh fabric is better than using phenol or phosphite antioxidant. The ratio of phenol antioxidant and HALS to 3:1, the fabric achieves the best anti aging effect. Therefore, the high relative molecular weight HALS not only has excellent light stability effect, but also has the effect of thermal stability.

Zhu Fuhai [18] applied Chimassorb994 to the polyethylene system and got similar conclusions.

Due to the hindered amine cannot absorb more than 260 nm ultraviolet light, in order to increase the polymer anti UV ability, scholars of the hindered amine light stabilizer and ultraviolet absorber do achieve.

Pan Jiangqing[19-20]studied light stability of polypropylene with hindered amine light stabilizers with ultraviolet absorbers. The results show that there is a good synergistic effect between Tinuvin-144 and UV-531 or UV-327.The hindered amine and ultraviolet absorber have mutual protective effect.GW-540 and UV-531 are used to show the co – isotropic, but with UV-327 and showing weak antagonism.

In addition, there have also been a number of studies on the combination of the hindered amine and antioxidants which is related to the structure of hindered amine and antioxidant.

Zhang Liji [21] studied the effect of antioxidant and light stabilizer in polyethylene, discovering that phenolic antioxidants and nitrogen oxygen free radical reacted easily. Irganox 1010 and light stabilizer GW-544 had antagonistic effect. Although antioxidants will capture free radicals, hydrogen peroxide decomposition, hindered phenolic antioxidant itself is susceptible to degradation.

Wang Huiliang [22-27] studied hindered amine light stabilizer modification of PE, PP materials resistant to irradiation induced color performance, discussing the mechanism of hindered amine light stabilizer enhancing the radiation resistance of polymer. The results show that, phenolic Irganox 1076 and HALS and show synergistic effect. Hindered amine PDS have synergistic effect with Irganox 1076 or Irganox 1010, but no antioxidant 1098.

You Jizeng[28] studied the application of the hindered amine light stabilizer in the LDPE agricultural film, resulting in synergistic effect of hindered amine Chimassorb944LD and phenolic antioxidant.

When hindered amine and phosphorous acid auxiliary antioxidants combine, the acid produced by the hydrolysis of the sub phosphate can be removed and the anti oxygen efficiency can be improved. Wang Huiliang [22-27] studied the effect of sub phosphate ester compound of PDS or BW-10LD, which showed synergistic effect. Zhang Liji [21]observed the counter effect between GW544 and Irganox 168.

When hindered amine and phosphorothioate antioxidants are used, the antioxidant is easily oxidized and decomposed to produce acidic substances, reacting with hindered amine with alkalineand reducing the effect of each other.Hu Xingjun[29] studied hindered amine Tinuvin770 with antioxidant DLTR or Santonox R. Compared with phosphorus containing antioxidants, it showed a clear counter effect. Thiobisphenol and other sulfur-containing antioxidants are not ideal with hindered amine.

5. Conclusion

With people pay more and more attention to the health and safety of medical activities, irradiation methods in medical sterilized product industry will occupy an increasingly important position, which requires the resistance to the irradiation of polymer materials research still need further development. At present, the main idea of solving this problem is by adding stabilizer. And it is urgent to solve the formula and proportion of the stabilizer.

Table 1. *The synergistic or antagonistic effect between stabilizers.*

Types of stabilizers		Hindered phenolic	phosphite ester	thiol	UV absorber	Hindered amine
antioxidant	Hindered phenolic	-	Synergistic	Synergistic	Unreported	Synergistic
	phosphite ester	Synergistic	-	Unreported	Unreported	antagonistic
	thiol antioxidant	Synergistic	Unreported	-	Unreported	antagonistic
light stabilizer	UV absorber	Unreported	Unreported	Unreported	-	Synergistic
	Hindered amine	Synergistic	antagonistic	antagonistic	Synergistic	-

In summary, hindered phenolic antioxidant and phosphite ester antioxidant respectively act to capture free radicals and decompose peroxide, which has a synergistic effect in the polymer oxidation process. Due to ultraviolet UV absorber can absorb the ultraviolet wavelength of more than 260nm which hindered amine can not, their mixture shows synergistic effect. Because of its easy oxidation decomposition of sulfur-containing antioxidants producing acidic substances, there shows antagonistic effect bewteen sulfur-containing antioxidants and alkaline hindered amine. There is a synergistic effect between hindered phenolic antioxidant, phosphite ester antioxidant and hindered amine light stabilizer. More work should be tried to solve the current problem by the synergistic effect between light stabilizer and antioxidant to improve the material's resistance to irradiation properties. Once the problem of polymer radiation is solved, the application of radiation sterilization in the medical industry will be more promising.

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