The role of Vitamin E and Hindered Amine Light Stabilizers in suppressing crosslinks in irradiated Polyethylene

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Introduction: Ultra-high molecular weight polyethylene (PE) components of total joint replacement prostheses are usually crosslinked by ionizing radiation since an increase in crosslink density can lead to a decrease in the rate of particulate wear of these components [1, 2], which is important to decrease the incidence of particle-induced osteolysis. The irradiated, crosslinked PEs (or XPE) are usually thermally stabilized after radiation to decrease the number of trapped free radicals to prevent long term oxidative degradation. However, thermal treatment causes scission and decrease the crosslink density. This process can be avoided by incorporating antioxidants, such as Vitamin E (alpha-tocopherol) into the PE component either before radiation [5, 6] or diffused into the component after the radiation [7]. It is known that Vitamin E stabilizes PE from oxidation even at a low weight percentage of 0.1% [5]. However, there is a concentration dependent decrease in crosslink density of PE if Vitamin E is incorporated prior to radiation since it is a radical scavenger and suppresses crosslinking. A recent study showed that three types of hindered amine light stabilizer (HALS) were able to prevent oxidation in PE without significant suppression of crosslinking since they only become radical scavengers after irradiation [8]. In this study, we quantified the crosslink density for irradiated PE containing Vitamin E and a variety of HALS stabilizers in order to determine the extent of crosslink suppression by each stabilizer.

Materials and Methods: Compression molded sheets of GUR® 1020 containing 0%, 0.05%, 0.1% and 0.75% Vitamin E respectively were provided by Ticona (Oberhausen, Germany) along with GUR 1020 containing 0.75% of various Hindered Amine Light Stabilizers (HALS) as shown in Table 1. The relatively high weight percentage of 0.75 was chosen in order to be able to clearly distinguish between the crosslink densities of irradiated GUR 1020 containing the various stabilizers. GUR 1020 without stabilizers served as control.

Results: Equilibrium swelling experiments showed that control GUR 1020 PE had the lowest swell ratio and molecular weight between crosslinks (Mw) were calculated using equations (1), (2) and (3) as shown below:

\[ q_{eq} = \frac{V_1}{V_0} - 1 \]  
\[ V_0 = \frac{1}{V_1} \left[ \ln(1-q_{eq}^{-1}) + q_{eq}^{-1} + Xq_{eq}^{-2} \right] \]  
\[ M_w = \frac{1}{V_1} \left[ \left( \frac{X}{V_1} \right)^{1/3} \right] \]  

where \( V_1 \) = 136 cm³/mol, \( X = 0.33 + 0.55/q_{eq}^{-1} \) [2] and \( V_0 = 920 \text{ g/dm}^3 [9] \).

Crosslink densities for 100 kGy electron beam irradiated GUR 1020 containing 0% (Control), 0.05, 0.1 and 0.75% Vitamin E (average ± standard deviation).

Table 1. Sample ID for various HALS stabilizers in GUR 1020 PE.

<table>
<thead>
<tr>
<th>Sample ID</th>
<th>Description</th>
</tr>
</thead>
<tbody>
<tr>
<td>HALS 1</td>
<td>GUR 1020 containing 0.75% Chimasorb® 2020</td>
</tr>
<tr>
<td>HALS 2</td>
<td>GUR 1020 containing 0.75% Tinuvin® 622</td>
</tr>
<tr>
<td>HALS 3</td>
<td>GUR 1020 containing 0.75% Tinuvin® 791</td>
</tr>
<tr>
<td>HALS 4</td>
<td>GUR 1020 containing 0.75% SONGNOX® 1010</td>
</tr>
<tr>
<td>HALS 5</td>
<td>GUR 1020 containing 0.75% Tinuvin® 770</td>
</tr>
</tbody>
</table>

Discussion: This study showed that there was a concentration dependent decrease in crosslink density in PE containing Vitamin E with nearly a 60% decrease in crosslink density at 0.75%. In general, all HALS stabilizers suppressed crosslinking by a much smaller amount than Vitamin E added to the same weight percentage with HALS 1 showing no suppression and HALS 2 showing only 6.4% lower crosslink density. This indicates that HALS may be preferable stabilizers for GUR 1020 PE compared to Vitamin E purely from the standpoint of suppression of crosslinking when the stabilizers are premixed into PE prior to irradiation. However, the biocompatibility as well as effectiveness of HALS as antioxidants must be comprehensively evaluated before being implemented for this clinical application requiring a high level of crosslinking in irradiated PE.

Significance: This study has strong clinical relevance since it quantifies the decrease in formation of crosslinks in irradiated PE containing two types of antioxidants. Crosslink density has been shown to be inversely correlated to wear rates of PE components, which must be decreased to prevent particle induced osteolysis in total joint replacement prostheses.


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