A TESTING PROGRAM TO EVALUATE THE EFFECTS OF SIMULANT MIXED WASTES ON PLASTIC TRANSPORTATION PACKAGING COMPONENTS

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ABSTRACT

Based on regulatory requirements for Type A and B radioactive material packaging, a Testing Program was developed to evaluate the effects of mixed wastes on plastic materials which could be used as liners and seals in transportation containers. The plastics evaluated in this program were butadiene-acrylonitrile copolymer (Nitrile rubber), cross-linked polyethylene, epichlorohydrin, ethylene-propylene rubber (EPDM), fluorocarbons [Viton™ and Kel-F™ (PCTFE)], high-density polyethylene (HDPE), butyl rubber, polypropylene, polytetrafluoroethylene, and styrene-butadiene rubber (SBR). These plastics were first screened in four simulant mixed wastes. The liner materials were screened using specific gravity measurements and seal materials by vapor transport rate (VTR) measurements. For the screening of liner materials, Kel-F, HDPE, and XLPE were found to offer the greatest resistance to the combination of radiation and chemicals. The tests also indicated that while all seal materials passed exposure to the aqueous simulant mixed waste, EPDM and SBR had the lowest VTRs. In the chlorinated hydrocarbon simulant mixed waste, only Viton passed the screening tests. In both the simulant scintillation fluid mixed waste and the ketone mixture waste, none of the seal materials met the screening criteria. Those materials which passed the screening tests were subjected to further comprehensive testing in each of the simulant wastes. The materials were exposed to four different radiation doses (1.4, 2.9, 5.7, and 37 kGy) followed by exposure to a simulant mixed waste at three temperatures (18, 50, and 60°C) and four different exposure times (7, 14, 28, 180 days). Materials were tested by measuring specific gravity, dimensional, hardness, stress cracking, VTR, compression set, and tensile properties. The second phase of this Testing Program involving the comprehensive testing of plastic liner has been completed and for seal materials is currently in progress. This phase involved testing the above properties in the first type of mixed waste simulant, an aqueous Hanford Tank simulant. The results of the latter phase in this experimental program have shown that PCTFE had the greatest durability after exposure to gamma radiation followed by exposure to the Hanford Tank simulant mixed waste. EPDM and butyl rubber were found to be remarkably resistant under the above conditions. These results are described in some detail in this paper.
INTRODUCTION

Hazardous and radioactive materials packagings permit transportation of such materials without posing a threat to the health or property of the general public. To achieve this aim, regulations have been written establishing general design requirements for such packagings. While no regulations have been written specifically for mixed waste packaging, regulations for the constituents of mixed wastes, i.e., hazardous and radioactive substances, have been codified. The design requirements for both hazardous [USDOT a] and radioactive [USDOT b] materials packaging specify packaging compatibility, i.e., that materials of the packaging and any contents be chemically compatible with each other. Furthermore, Type A [USDOT c] and Type B (USNRC) packaging design requirements stipulate that there be no significant chemical, galvanic, or other reaction between the materials and contents of the package. Based on these requirements, a Chemical Compatibility Testing Program [Nigrey et al (1994)] was developed in the Transportation Systems Department at Sandia National Laboratories. This Testing Program aims to assure any regulatory body that the issue of packaging compatibility towards hazardous and radioactive materials has been addressed.

EXPERIMENTAL

Materials

The selected materials were five plastics liner materials having known chemical resistance to a large number of classes of chemicals. The selected plastics were high-density polyethylene (HDPE), cross-linked polyethylene (XLPE), polypropylene (PP), and fluorocarbon plastics [polychlorotrifluorethylene (PCTFE, see Kel-F™ in Reference Section), and polytetrafluoroethylene (Teflon)]. Based on previous screening results (Nigrey et al, 1994), the elastomeric seal materials Ethylene Propylene Diene Monomer (EPDM) and butyl rubber were also evaluated in a separate study.

Simulant Preparation

The simulant mixed waste form used in this testing phase was an aqueous alkaline simulant Hanford Tank waste. It was prepared by dissolving 179 g (2.10 moles) of sodium nitrate and 50 g
(0.73 moles) sodium nitrite in deionized water (600 mL) using a 4-L beaker. After these salts had completely dissolved, 82 g (2.05 moles) sodium hydroxide was added under stirring and slight heating using a magnetic hotplate. To this hot (~70°C) stirred solution, 17 g (0.107 moles) cesium chloride and 16 g (0.0952) strontium chloride were added. Finally, 32 g (0.301 moles) of sodium carbonate was added to the solution. This latter addition resulted in the formation of a copious amount of white precipitate. Based on its insolubility, it is believed that this precipitate is strontium carbonate. To the resulting mixture another 400 mL of deionized water was added to bring the total volume of water used to 1 L. After cooling to near ambient temperature, the stirred mixture was stored in Amber Glass Bottles. It should be mentioned that the procedure described above was scaled up threefold to give 3-L batches of the simulant. All chemicals used in the preparation of the waste simulant were ACS reagent grade chemicals.

Sample Preparation
Standardized test methods were used to cut, condition, and test the materials. The geometry of the material samples was specified by the test method. The samples were cut using an expulsion press (Part # 22-16-00) and dies manufactured by Testing Machines Inc., Amityville, NY. For example, the rectangular (~2.5 cm x 5 cm x 0.3 cm) samples required for specific gravity and hardness measurements were cut in the expulsion press fitted with an Expulsion Straight Edge Die (Part #23-10-06). Rectangular (~2.5 cm x 7.7 cm x 0.3 cm) samples required for dimensional measurements were cut in the expulsion press fitted with an Expulsion Straight Edge Die (Part #23-10-07). Rectangular (~1.3 cm x 3.8 cm x 0.3 cm) samples required for stress cracking measurements were cut in the expulsion press fitted with an Expulsion Straight Edge Die (Part #23-14-36). Sample discs (~1.3 cm diameter x 0.318 cm thick) required for elastomer compression set measurements were cut in the expulsion press fitted with a custom circular cutter from CCS Instruments, Akron, OH. Larger discs (~6.8 cm diameter x 0.3 cm thick) required for Vapor Transport Rate (VTR) measurements on elastomers were cut in the expulsion press fitted with an expulsion die (Part #23-00-00) specifically designed for American Society for Testing and Materials (ASTM) Standard Test Method [ASTM D814 (1991)]. The Type C tensile samples required for tensile testing of rubber materials were also cut in an expulsion press fitted with an
Expulsion Die (Part # 23-14-08) specifically designed for use in the ASTM D412-Method A. [ASTM (1987)]. Similarly, the Type IV samples required for tensile testing of plastics were cut in the expulsion press fitted with an Expulsion Die (Part # 23-14-23) specifically designed for ASTM D638 [ASTM (1991)]. The use of the press and dies permitted the cutting of multiple samples of uniform dimensions. When attempting to cut out the harder materials such as HDPE, PP, and Kel-F™ with the expulsion press, considerable difficulty was encountered. This problem necessitated machining the required “dog bone” samples of the materials to Type IV specifications. The individual samples were visually checked to assure that none had nicks or other imperfections prior to their use. A matrix was developed for labeling samples according to test method, sample number, and testing conditions. The samples were individually labeled with the use of ~0.3 cm steel letter and number stamp sets. Due to the limited space available, the tensile testing samples were labeled with ~0.16 cm steel letter and number stamps. The black EPDM and butyl rubber samples were individually labeled using indelible ink marking pens. As recommended by ASTM D618 (1990) and ASTM D1349-87 (1987), the plastics were conditioned at a standard temperature of 23°C (73.4°F) and relative humidity of 50% for at least 24 hours prior to the testing process. This was done by storing the cut samples in a desiccator filled with magnesium nitrate hexahydrate (500 g) and saturated with water. A humidity/temperature sensor was used to monitor these conditions. Procedures for generating this constant relative humidity environment are described in ASTM E104 (1991). During conditioning, the samples were stacked atop each other and separated from each other using metal spacers.

**Sample Irradiation**

The five liner materials and two seal materials were cut out for each of the property measurements and the respective radiation doses, temperatures, and time exposures. These were exposed to gamma radiation from an underwater ⁶⁰Co source at SNL. The samples were loaded into a metal basket in the same configuration as was used to condition the samples, i.e., the samples were stacked atop each other and separated by a metal spiral. The basket was then inserted into a water-tight stainless steel canister (volume ~4 L). The canister was sealed and lowered into the pool to a depth of 6 feet, purged with a slow steady flow (~30 mL/min) of dry air, and allowed to
come to thermal equilibrium at either ambient, 50, or 60°C. Once thermal equilibrium was obtained, the canister was lowered into its irradiation location in the pool and the exposure time was started to obtain the desired radiation dosage. The highest dose rate available at the Low Intensity Cobalt Array (LICA) Facility [Nelson et al (1996)] during these liner studies was \(-2\) kGy/hr. In subsequent seal studies, the LICA Facility received new \(60^{\text{Co}}\) sources resulting in dose rates of \(-7\) kGy/hr. Thus for liner irradiations where a gamma-ray dose of \(-1.4\) kGy was required, the samples were exposed for approximately 0.75 hours. For doses of \(-2.9\), 5.7, and 37 kGy, the corresponding longer exposure times were needed. After the samples received the calculated radiation dosage, the canister was removed from the pool and the samples were again placed in the conditioning chambers. No more than 24 hours typically elapsed between the time the samples had been exposed to radiation and when they were exposed to the simulant wastes.

Sample Exposure to Simulants

The general exposure protocol for specific gravity involved placing four specimens of each plastic material into a container, and exposing them to the specific testing conditions. The four specimens were bundled together using 19 cm nylon cable ties. Within each bundle, the specimens were separated through the use of \(-1.5\) mm metal pins as spacers. This allowed for the ready access of the waste simulant to all surfaces of each specimen. For relatively insoluble materials, ASTM D543 (1987) recommends about 10 mL/in² \((-1.6\) mL/cm²). A 2-L glass bottle was loaded with the four bundled test specimens and then filled with 1,600 mL of the test solution. After adding the liquid simulant waste, the plastic lid was attached to the jar and tightened. The jar(s) were placed in the respective environmental chambers maintained at 18, 50, and 60°C. The jar(s) were kept in these environmental chambers for 7, 14, 28, and 180 days. Similar procedures were followed for each of the other four testing procedures, i.e., dimensional testing, hardness testing, stress cracking tests, and tensile tests. In the case of stress cracking experiments, the samples were held in specially designed stainless steel specimen holders described in ASTM D1693 (1988). The samples held in the specimen holders were placed in the jars containing the aqueous waste simulant. For VTR measurements, each of three 1/2 pint \((-236\) mL) Mason Jars (Kerr Group, Inc., Los Angeles, CA, Part # 70610-3) was filled with
approximately 200 mL of the test solution. The EPDM and butyl rubber discs were loosely attached to the jars with metal bands. The jars were placed in an upright configuration (elastomer and band facing up) into the respective environmental chambers. The jars were then held at the respective test temperatures for one hour to equilibrate. After sealing and weighing them, the jars were placed in the chambers again in an inverted position and held at the specific test temperature for the required time period.

DISCUSSIONS

The purpose of a chemical compatibility testing program is to provide a scientifically defensible methodology for measuring the chemical compatibility of polymeric liner and seal materials with hazardous wastes. These polymeric materials are those which may be used in current and future container designs for the transportation of hazardous and mixed wastes throughout the DOE complex. The approach for developing such a testing program was to assess the current state of chemical compatibility testing technology and develop methods that might lead to satisfactory, comprehensive, and reliable chemical compatibility data for use by the U. S. DOE in its Office of Environmental Management.

The results of the first phase of this testing program have been presented by Nigrey and Dickens (1995). We will only summarize the results in this paper. The 1st phase involved the screening of five candidate liner and six seal materials to four simulant mixed wastes, respectively. The testing protocol involved exposing the respective materials to ~2.9 kGy of gamma radiation followed by 14-day exposures to the waste types at 60°C. Seal materials were tested using VTR measurements while materials suitable for liner applications were tested using specific gravity measurements. For these tests, screening criteria of ~1 g/m²/hr for VTR and a specific gravity change of 10% mentioned by Swope et al (1985) were used as the metric. Those materials which failed to meet these criteria were judged to have failed the screening tests and were excluded from the next phase of this experimental testing program.

The screening studies concluded that while all seal materials passed exposure to the aqueous simulant mixed waste, EPDM rubber and SBR had the lowest VTRs. In the chlorinated
hydrocarbon simulant mixed waste, only VITON passed the screening tests. In both the simulant scintillation fluid mixed waste and the ketone mixture simulant mixed waste, none of the seal materials met the screening criteria. Those materials with the lowest VTRs will be evaluated in the comprehensive phase of the program. For specific gravity testing of liner materials the data showed that while all materials with the exception of PP in chlorinated hydrocarbon simulant mixed waste passed the screening criteria, Kel-F™, HDPE, and XLPE were found to offer the greatest resistance to the combination of radiation and the four simulant mixed waste chemicals.

With the completion of the screening phase of the program several years ago, the comprehensive phase of this program has been in progress. Since all seal and liner materials passed the screening tests when exposed to the simulant Hanford Tank Waste, ten materials needed to be subjected to the test matrix. This would result in an extremely large sample set. In view of manpower and budget constraints, the comprehensive testing phase of the program was further subdivided into the testing of liner materials and seal materials. The results of liner testing will now be discussed.

The material properties that should be evaluated to assess the suitability of potential liner materials in mixed waste packaging designs are mass, density, and dimensional changes, hardness, modulus of elasticity, tensile strength, elongation, and stress cracking in polyethylene materials. Five liner materials were subjected to a matrix of four radiation doses, three temperatures, and four times in the simulant waste. The evaluation parameters used in this comprehensive testing phase consisted of the previously mentioned material properties. These properties were evaluated using standardized test methods such as those developed by the ASTM. For specific gravity changes, ASTM D792 (1991) was used. In evaluating dimensional changes, ASTM D471 (1991) was used. For hardness changes, ASTM D2240 (1991) was used. In evaluating stress cracking in polyethylene materials, ASTM D1693 (1988) was used. For evaluating the tensile property changes in liner materials, ASTM D638 (1991) was used. For evaluating elastomers, several additional standard test methods were used. For compression set, ASTM D395 (1989) was used. Finally for evaluating the tensile properties of rubber materials, ASTM D412 (1997) was used.

Before describing the results of this study, we will describe the general comprehensive testing strategy. This strategy is shown in Figure 1. Five liner materials (HDPE, XLPE, PP, Kel-F™,
and Teflon) were evaluated. These materials were subjected to four different protocols (Paths A-D). To determine the intrinsic properties of the materials, the “Baseline samples” (Path A) were prepared for each of the tests. In order to differentiate the effects on the materials by radiation and chemicals, one series of samples was only exposed to the simulant (Path B), while the other series of samples were exposed to both radiation and the simulant (Path C). The first series of samples are described as “Simulant Only” in the flow diagram. It should be noted that both series of samples were exposed for the four time periods (7, 14, 28, and 180 days) at three different temperatures (18, 50, and 60°C). For two testing protocols, tensile testing and stress cracking, where the effects of radiation and temperature alone could have significant impact on the properties, a series of samples described as “Rad Only” is shown in the flow diagram (Path D). What may not appear obvious from the flow diagram is the large number of samples being tested in this comprehensive testing phase of the program. In view of the large amount of data generated, we will only present the results of conditions where material properties have significantly changed. These conditions were at the highest gamma radiation dose (~37 kGy), the highest exposure duration (180 days) and the highest exposure temperatures (60°C).

RESULTS
Figure 2 shows the percent specific gravity (% S. G.) changes of five liner materials exposed to 37 kGy of gamma ray doses followed by exposure to the aqueous simulant waste at 60°C for 7, 14, 28, and 180 days. It should be noted that the scale for % S. G. Change is rather small, e.g., from 1 to 5%, and either positive or negative. The sign of the specific gravity indicates whether specific gravity has increased or decreased when compared to the pristine materials, i.e., the material’s specific gravity at ambient conditions. Therefore, changes in the magnitude and the sign of specific gravity values indicate changes in this property. The greater the absolute values of the changes the more the materials are affected by this set of environmental conditions. Since properly engineered packaging components are not expected to be affected by contents of the package, i.e., the mixed wastes, materials exhibiting the least changes in specific gravity should be selected as packaging components. The results in Figure 2 show that all materials, with the
exception of Teflon, exhibited specific gravity changes less than 2%. These results for are significantly different from those found for HDPE, XLPE, PP, and Kel-F™ exposed to the lower radiation doses and temperatures. Under the less aggressive conditions S.G. changed less than 1%. Teflon stood out in that increases in S. G. of more than 2% were observed. Increases in specific gravity (or relative density) suggest that either the mass increased or the volume of the samples decreased. As can be seen in Figure 3, the latter situation is the case for Teflon. Figure 3 gives the volume changes for five liner materials exposed to 37 kGy of gamma ray doses followed by exposure to the aqueous simulant waste at 60°C for 7, 14, 28, and 180 days. Since volume represents the product of length, width, and thickness, the evaluation of volume changes provides a concise picture of these environmental effects. The data shows that for HDPE and Kel-F™ minimal volume changes occurred. Teflon exhibits decreases in volume at all exposure times.

In Figure 4, we give the hardness changes of five liner materials exposed to 37 kGy of gamma ray doses followed by exposure to the aqueous simulant waste at 60°C for 7, 14, 28, and 180 days. As was seen in the previously discussed measurements, Teflon stands out in its response to a combination of high radiation and elevated exposure temperatures. Teflon showed decreases in hardness up to ~14%. This means that the material has become softer. For certain applications in transportation packagings this may or may not be desirable.

Figure 5 gives the tensile strength changes for five liner materials exposed to 37 kGy of gamma ray doses followed by exposure to the aqueous simulant waste at 60°C for 7, 14, 28, and 180 days. Consistent with all the previous measurements, all materials had small tensile strength changes while Teflon had a decrease in tensile strength of nearly 80%.

Based on the results presented in this paper, it is worthwhile to attempt to identify the one liner material which displayed the greatest chemical compatibility towards the simulant mixed waste under these conditions. In order to accomplish this, a ranking scheme needed to be developed. For this purpose, we chose to sum the property changes and derive an average value over the four exposure times. That material which was calculated to have the lowest average property change value, i.e., changed the least, was assigned an arbitrary value of one (1). The other materials were
then given values from two (2) to five (5) in the order of increasing average property change values. This can be seen visually by closely inspecting the results given in Figures 2 - 5. For example, in Figure 4, XLPE would appear to be first, i.e., assigned a value of 1, HDPE (2), Kel-F™ (3), PP (4), and Teflon (5). By adding the respective values for all four radiation doses and three temperatures, a total value for each measurement can be obtained. The ranking scheme developed by this process is given in Table 1. The material with the best overall response should have the lowest score in all the properties measured. This can be determined by adding the rankings for each material and choosing the material with the lowest total value. As can be seen in Table 1, this very simplistic approach has identified the chlorofluorocarbon Kel-F™ as the material which is most compatible with this simulant mixed waste under these conditions. However, the well-known engineering plastic, HDPE, could equally well be identified as being very compatible by virtue of its good performance when specific gravity and tensile strength are used as the metric. For these two measurements, HDPE could be ranked first by virtue of this materials low total value for these two properties. Since packaging designers may have other criteria for selecting materials, the data in Table 1 can be used in different ways.

Table 1. Material Ranking

<table>
<thead>
<tr>
<th>Property</th>
<th>HDPE</th>
<th>XLPE</th>
<th>PP</th>
<th>Kel-F</th>
<th>TEFLOW</th>
</tr>
</thead>
<tbody>
<tr>
<td>Specific Gravity Changes</td>
<td>26</td>
<td>31</td>
<td>32</td>
<td>31</td>
<td>60</td>
</tr>
<tr>
<td>Dimensional Changes</td>
<td>43</td>
<td>46</td>
<td>32</td>
<td>29</td>
<td>31</td>
</tr>
<tr>
<td>Hardness Changes</td>
<td>39</td>
<td>22</td>
<td>37</td>
<td>22</td>
<td>60</td>
</tr>
<tr>
<td>Tensile Strength Changes</td>
<td>21</td>
<td>32</td>
<td>26</td>
<td>31</td>
<td>55</td>
</tr>
<tr>
<td>Total</td>
<td>129</td>
<td>131</td>
<td>127</td>
<td>113</td>
<td>206</td>
</tr>
</tbody>
</table>

Since HDPE might be selected by some based on the data presented here, it is worthwhile to discuss the issue of stress cracking. Environmental stress cracking is a form of chemical attack in which a chemical that does not appreciably attack or dissolve a polymer in an unstressed state will cause catastrophic failure when the polymer is stressed in its presence. The stress cracking phenomenon is a recognized potential problem with some varieties of HDPE and other semicrystalline polymers. For this reason, a specific standardized test, ASTM D1693 (1988) has been developed. In Figure 6a, we give the stress cracking failures for HDPE exposed to -1.4, 2.9, 5.7, and 37 kGy of gamma ray doses followed by exposure to the aqueous simulant waste at 60°C for
7, 13, 28, and 180 days. Figure 6b, shows the stress cracking failures for XLPE subjected to the same conditions. These results show quite clearly that XLPE has superior stress cracking performance.

We now proceed to present some of the comprehensive testing results for the elastomers EPDM and butyl rubber. As was mentioned previously, compression set and vapor transmission rate was measured in addition to those of specific gravity, mass, dimensions, hardness, and tensile properties. These property measurements for EPDM have indicated that this elastomer is remarkably resistant to the effects of radiation and the aqueous simulant at these temperatures and exposure times. In certain instances, a slight “beneficial” effect of the aqueous simulant on irradiated EPDM was also observed. An example of this effect is shown in Figure 7, where EPDM samples exposed to four gamma radiation doses and then held at the three temperatures for 14 days (a) is compared with samples exposed to both radiation and the simulant under these conditions. A close inspection of the data in Figure 7b reveals that a slight increase in tensile strength was observed for EPDM samples exposed to both radiation and the simulant. For EPDM samples exposed to only gamma radiation (Figure 7a), EPDM exhibited lower tensile strengths. The cause of this effect is not understood at this time. The overall results from these testing results suggest that the type of aqueous mixed waste used in this study have minimal effects on EPDM. Similar testing on butyl rubber has been completed and the data is being analyzed. Preliminary analyses show that butyl rubber also appears to be resistant to the effects of radiation and aqueous mixed wastes based on all the above mentioned property measurements except tensile properties. The latter measurements show that butyl rubber begins to exhibit a loss in tensile strength and tensile stress after exposure to ~5.7 kGy and the aqueous simulant at all three temperatures and exposure times. These effects are shown in Figure 8. In Figure 8a, we compare the tensile strength of EPDM to that of butyl rubber (Figure 8a) under the same conditions of exposure time, exposure dose, and exposure temperature. The sudden loss of tensile strength for butyl rubber at ~5.7 kGy in Figure 8b is clearly not observed for EPDM rubber in Figure 8a. These results suggest that EPDM rubber has better mechanical properties under these
conditions. A more detailed analyses still needs to be performed to establish the overall performance of EPDM versus Butyl rubber.

CONCLUSIONS

We have described a Chemical Compatibility Program for the evaluation of transportation packaging components which may be used in transporting mixed waste forms. Consistent with the methodology outlined in this paper, we have performed the second phase of this experimental program to determine the effects of simulant Hanford Tank mixed wastes on packaging materials. This effort involved the comprehensive testing of five plastic liner materials in the aqueous mixed waste simulant. The testing protocol involved exposing the respective materials to 1.4, 2.9, 5.7, and 37 kGy of gamma radiation followed by 7, 14, 28, 180 day exposures to the waste simulant at 18, 50, and 60°C. From the data analyses performed, we have identified the fluorocarbon Kel-F™ (PCTFE) as having the greatest chemical durability after having been exposed to gamma radiation followed by exposure to the Hanford Tank simulant mixed waste. The most striking observation from this study was the extremely poor performance of Teflon under these conditions. Both EPDM and butyl rubber were found to be remarkably resistant to radiation and this simulant mixed waste under the above conditions.

REFERENCES


Kel-F, A trademark product for polychlorotrifluoroethylene (CTFE) formerly manufactured by 3M. Daikon America now manufactures this material under the tradename Neoflon.


Figure 1. Comprehensive Testing Strategy for Plastics and Elastomers

Figure 2. Specific gravity measurements

Figure 3. Volume measurements
Figure 4. Hardness measurements

Figure 5. Tensile strength measurements

Figure 6. Stress cracking measurements for HDPE (a) and XLPE (b)

Figure 7. Tensile strength changes for irradiated EPDM exposed at 18, 50, and 60° C for 14 days (a), and EPDM samples after exposure to ~ 0, 1.4, 2.9, 5.7, and 37 kGy gamma radiation followed by exposure for 14 days to the aqueous simulant mixed waste at 18, 50, and 60° C (b).
Figure 8. Tensile strength changes for EPDM (a) and butyl rubber (b) after exposure to 0, 1.4, 2.9, 5.7, and 37 kGy gamma radiation followed by exposure for 180 days to the aqueous simulant mixed waste at 18, 50, and 60°C.