EFFECTS OF SIMULANT MIXED WASTE ON EPDM AND BUTYL RUBBER

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SUMMARY

We have developed a Chemical Compatibility Testing Program for the evaluation of plastic packaging components which may be used in transporting mixed waste forms. In this program, we have screened 10 plastic materials in four liquid mixed waste simulants. These plastics were butadiene-acrylonitrile copolymer (Nitrile) rubber, cross-linked polyethylene, epichlorohydrin rubber, ethylene-propylene (EPDM) rubber, fluorocarbons (Viton® and Kel-F™), polytetrafluoroethylene (Teflon®), high-density polyethylene, isobutylene-isoprene copolymer (Butyl) rubber, polypropylene, and styrene-butadiene (SBR) rubber. The selected simulant mixed wastes were (1) an aqueous alkaline mixture of sodium nitrate and sodium nitrite; (2) a chlorinated hydrocarbon mixture; (3) a simulant liquid scintillation fluid; and (4) a mixture of ketones. The screening testing protocol involved exposing the respective materials to ~3 kGy of gamma radiation followed by 14-day exposures to the waste simulants at 60°C. The rubber materials or elastomers were tested using VTR measurements while the liner materials were tested using specific gravity as a metric. For these tests, screening criteria of ~1 $g/hr/m^2$ for VTR and specific gravity change of 10% were used. Those materials that failed to meet these criteria were judged to have failed the screening tests and were excluded from the next phase of this experimental program. We have completed the comprehensive testing phase of liner materials in a simulant Hanford Tank waste consisting of an aqueous alkaline mixture of sodium nitrate and sodium nitrite. From the data analyses performed, we have identified the chlorofluorocarbon Kel-F™ as having the greatest chemical durability after having been exposed to gamma radiation followed by exposure to the aqueous alkaline simulant mixed waste. The most striking observation from this study was the extremely poor performance of Teflon under these conditions. We have also completed the comprehensive testing of two elastomers, EPDM and Butyl rubber, in simulant Hanford Tank waste. In the evaluation of these two elastomeric materials, we have concluded that while both materials exhibit remarkable resistance to these environmental conditions, EPDM has a greater resistance to this corrosive mixed waste.

INTRODUCTION

The purpose of hazardous and radioactive materials packaging is to enable these materials to be transported without posing a threat to the health or property of the general public. To achieve this aim, regulations in the United States 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 by the U.S. Department of Transportation (DOT, 49 CFR 173) and the U.S. Nuclear Regulatory Commission (NRC, 10 CFR 71). Based on these national requirements, a Chemical Compatibility Testing Program (Nigrey 1995) was developed for the U.S. Department of Energy's National Transportation Program in the Transportation Systems Department at Sandia National Laboratory.

In this paper, we present the results of Part B of the second phase of this testing program. The first phase screened five liner materials and six seal materials towards four simulant mixed wastes. This work was presented at PATRAM'95 (Nigrey and Dickens 1995a). Part A of the second phase involved the comprehensive testing of five candidate liner materials to an aqueous Hanford Tank simulant mixed waste. This work was also presented at PATRAM'95 (Nigrey and Dickens 1995b). Part B involved similar testing on elastomeric materials, ethylene-propylene (EPDM) and butadiene-acrylonitrile (butyl) rubber. The comprehensive testing protocol involved exposing the respective materials to a matrix of four gamma radiation doses (~1, 3, 6, and 40 kGy), three temperatures (18, 50, and 60°C), and four exposure times (7, 14, 28, and 180 days). Following their exposure to these combinations of conditions, the materials were evaluated by measuring six material properties. These properties were specific gravity, dimensional changes, hardness, vapor transport rates, compression set, and mechanical properties. Among the mechanical properties measured were tensile strength, ultimate elongation, and tensile stress also referred to as the 100% modulus.

EXPERIMENTAL

Materials

The selected materials were six elastomers having known chemical resistance to a large number of classes of chemicals. The elastomers were butadiene-acrylonitrile rubber, ethylene-propylene rubber (EPDM), epichlorohydrin rubber, isobutylene-isoprene copolymer rubber (Butyl), styrene-butadiene rubber (SBR), and Viton® rubber. Of the six elastomers to be subjected to comprehensive testing in the Hanford Tank simulant, only EPDM and butyl rubber have been studied so far in this multi-year program.

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 stirrer. To this hot (~70°C) stirred solution, 17 g (0.107 moles) cesium chloride and 16 g (0.0952 moles) strontium chloride were added. Finally, 32 g (0.301 moles) of sodium carbonate was added to the solution. To the resulting mixture was added another 400 mL of deionized water 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.

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 and dies manufactured by Testing Machines Inc., Amityville, NY. The use of the press and dies permitted the cutting of multiple samples of uniform dimensions. The individual samples were visually checked to assure that none had nicks or other imperfections prior to their use. As recommended by the American Society of Testing and Materials (ASTM D618-61), the elastomers were conditioned at a standard temperature of 23°C and relative humidity of 50% for at least 24 hours prior to the testing process.

Sample Irradiation

The above mentioned samples were exposed to gamma radiation from an underwater ^{60}Co source at SNL. These 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 ~ 2 meters, purged with 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 was ~7 kGy/hr but the array used to irradiate these materials had dose rates of 0.95 kGy/hr. Thus for irradiation where a gamma-ray dose of 1.43 kGy was required, the samples were exposed for approximately 1.5 hours. For doses of ~3, 6, and 40 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.

Sample Exposure to Chemicals

The general exposure protocol for all tests involved placing the specimens of each elastomer material into a container, and exposing them to the specific testing conditions. Care was taken to ensure that sufficient simulant waste was present to expose the entire surface area of all the samples. After adding the liquid simulant waste, the plastic lid was attached to the jar and tightened. The containers were placed in the respective environmental chambers maintained at 18, 50, and 60°C. The containers were kept in these environmental chambers for 7, 14, 28, and 180 days.

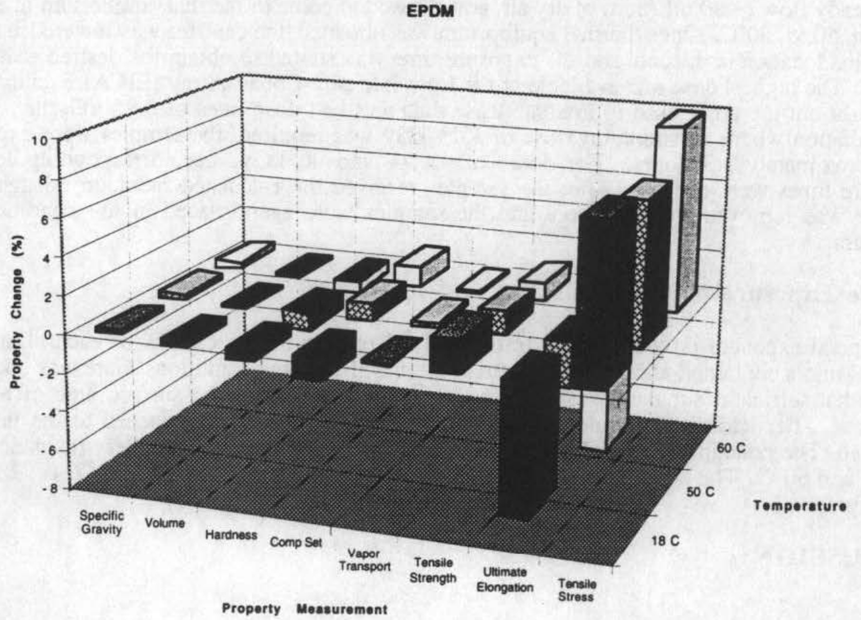
DISCUSSION

The material properties that should be evaluated to assess the suitability of potential elastomeric materials in mixed waste packaging designs are mass, dimensional and density changes, hardness, compression set, Vapor Transport Rates (VTR), tensile strength, elongation, and tensile stress (100% modulus). Since the measurement of <u>all</u> these material properties was expected to be costly and time-consuming, screening tests with relatively severe exposure conditions such as high temperatures (60°C) and high radiation levels (~3 kGy) were implemented to quickly reduce the number of possible materials for full evaluation. The screening criteria used were density changes for liners and vapor transport rates for elastomers. From this screening study, all of the selected liner and seal materials were found to have passed the screening criteria in the aqueous simulant mixed waste. This then resulted in a need to test these materials by exposure to a matrix of four radiation doses, three temperatures, and four times in the simulant waste. The properties of the materials were evaluated using standardized test methods such as those developed by the American Society for Testing and Materials (ASTM). For specific gravity changes, ASTM D792 was used. In evaluating dimensional and mass changes, ASTM D543 was used. For hardness changes, ASTM D2240 was used. In evaluating compression set, ASTM D395-Method B was used. For VTR measurements, ASTM D814 was used. Finally, for evaluating tensile properties, ASTM D412-Method A was used.

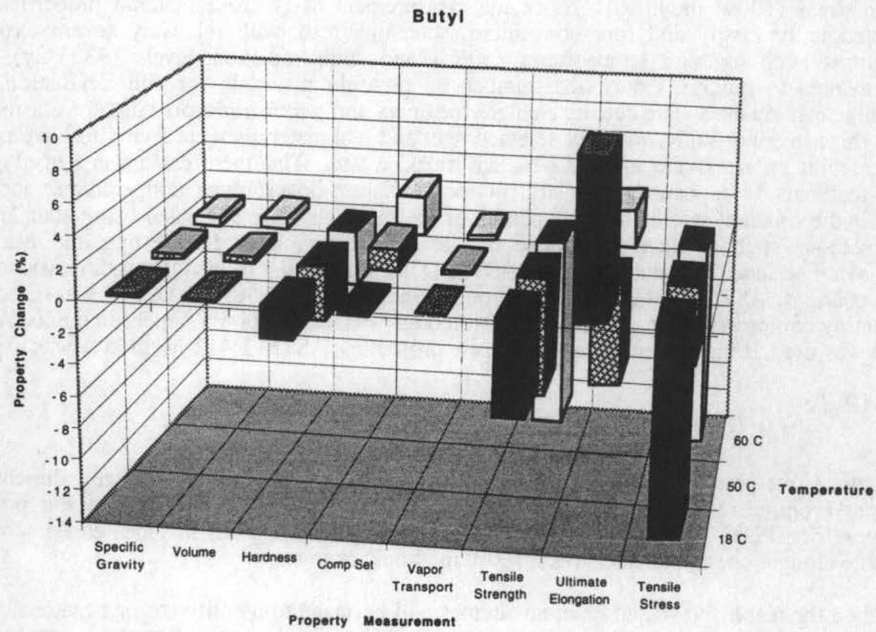
RESULTS

In Figure 1, we present the results of six measurements: specific gravity changes, dimensional changes (volume changes), hardness changes, compression set, VTR, and tensile property changes for EPDM and butyl rubber. Note that tensile properties include tensile strength, ultimate elongation, and tensile stress (100% modulus) changes.

Based on the results presented here, an attempt will be made to identify the one material which displayed the greatest compatibility towards the corrosive simulant mixed waste under these the extensive amount of data generated in this program and the limited space available in this paper, a summary of the experimental data is shown in Fig. 1. The values given in the 3-D bar graphs were obtained by calculating an average value from the individual values at the four gamma doses and four exposure times. These results therefore represent average values at the exposure conditions. The material with the lowest property change represents the most compatible material.



(a)



(b)

Figure 1. Comprehensive testing results for EPDM (a) and butyl rubber (b) Note: These results represent average values after exposure to four gamma radiation doses and the aqueous simulant waste over the four exposure times at 18, 50, and 60°C.

With this simple approach, a close inspection of Figure 1a & 1b shows that EPDM had the best response at these environmental conditions. For specific gravity, volume, hardness, compression set, vapor transport, and tensile strength changes in EPDM, some properties decreased about 1% and increased about 2% for others. In the case of ultimate elongation, decreases of nearly 10% were observed. However, the tensile stress (100% modulus) of EPDM increased by nearly 7% at the three temperatures. These results indicate that in general EPDM has become somewhat stiffer after exposure to these environmental conditions.

In Figure 1b, the response of butyl rubber at these environmental conditions is given. Similar to EPDM rubber, most of the properties changes were below 3%. However, the hardness, tensile strength, ultimate elongation, and tensile stress decreased from 4% to nearly 14%, respectively. These results indicate that butyl rubber has become softer and lost some of its strength. However, it should be restated that the results given in Figure 1 represent average values from the four radiation doses and four exposure times. Results observed at specific radiation doses and exposure times may be somewhat different than the averages. If specific values are of interest, the authors should be contacted.

CONCLUSIONS

We have developed a chemical compatibility program for the evaluation of plastic packaging components which may be incorporated in packaging for transporting mixed waste forms. From the data analyses performed to date in this study, we have identified the thermoplastic, polychlorotrifluoroethylene, as having the greatest chemical compatibility 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 poor performance of polytetrafluoroethylene under these conditions. In the evaluation of the two elastomeric materials, EPDM and butyl rubber, we have concluded that while both materials exhibit remarkable resistance to these environmental conditions, EPDM has a greater resistance to this corrosive simulant mixed waste.

ACKNOWLEDGMENTS

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