Contract No:

This document was prepared in conjunction with work accomplished under Contract No. 89303321CEM000080 with the U.S. Department of Energy (DOE) Office of Environmental Management (EM).

Service Life Prediction of Polymeric Hose-In-Hose Systems Used to Transfer High Level Liquid Waste Between Operations – 22134

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ABSTRACT
An equation has been developed where burst strength has been established as a service life criterion for rubber hoses that are used to transfer radioactive liquid waste. The equation accounts for degradation due to caustic chemical, ionizing radiation, and ambient aging between waste transfer periods. The work presented below is a follow up to the presentation given at WM2021. These flexible hose systems have proven to be a viable and cost effective alternative to metal piping in several instances and can assist in timely processing and removal of legacy waste.

INTRODUCTION
An example of the Hose-In-Hose system is illustrated in Figure 1. The inner hose is the conduit for liquid waste and the outer hose is there as an engineering control for containment in case of leakage. Hose-In-Hose systems were first used at the Hanford facility using a product developed specifically for them and was priced accordingly. Savannah River Remediation (SRR) first used the Hose-In-Hose system in the mid 2000's to transfer low level liquid waste from H-Canyon to the Tank Farm. The current Hose-In-Hose system used at SRR is for intermediate transfers of high level waste between tank 13 and 15 and has been in operation for 6 years (2016 to the present). SRR uses an off the shelf industrial chemical hose.

Figure 1. Example of the Hose-In-Hose Transfer system used at Savannah River Remediation.

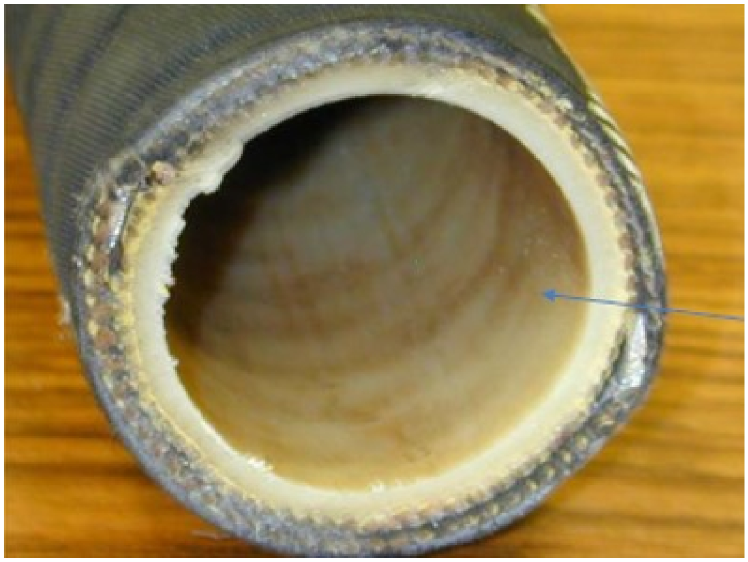

Figure 2. Cross sectional view of the SRR conduit hose.

Figure 2 shows a cross section of the conduit hose used at SRR. This hose is constructed with an inner liner made of compounded crosslinked polyethylene (XLPE). The black outer layer is composed of Ethylene Propylene Diene Monomer (EPDM) rubber. Between these two layers is a layer of reinforcement that includes two steel helical wires, a spiral-plied synthetic fabric and a resin that binds all three layers together. Synthetic fabric is also used within the outer layer of EPDM.

In comparison, a cross section of the conduit hose used at Hanford is shown in Figure 3. This hose is similar in construction to the hose used at SRR. There is an inner layer, a reinforcement layer, and outer layer. In place of a 2 mm XLPE inner layer the Hanford hose uses a 3mm EPDM rubber inner layer. The reinforcement layer contains two steel wires and reinforcing fabric. The outer layer is composed of EPDM rubber with a synthetic fabric used within that layer as well.

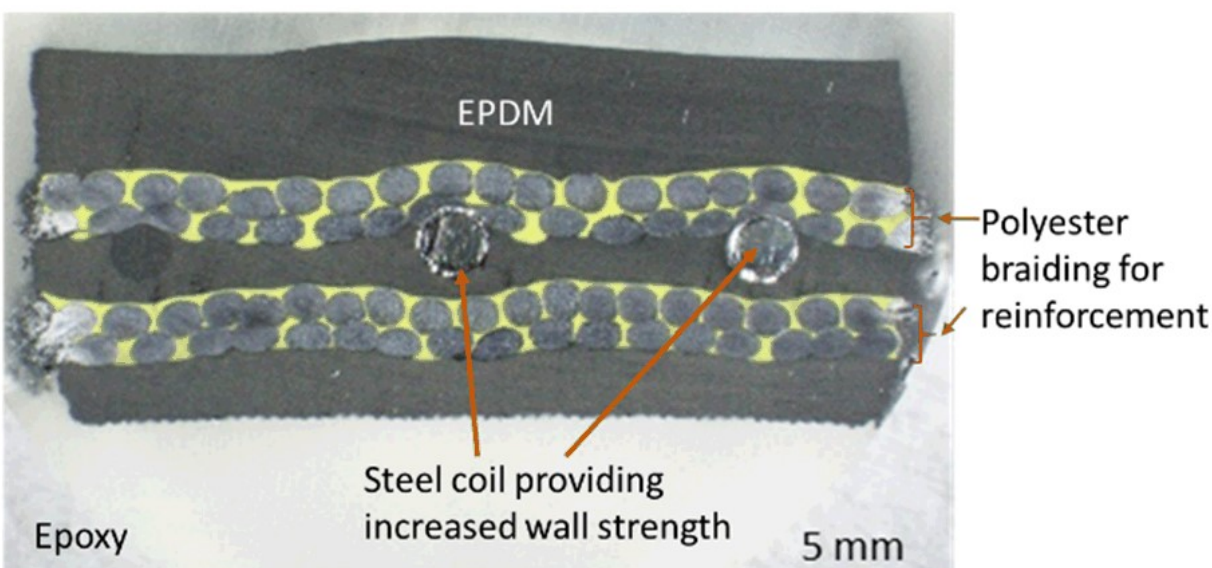

Figure 3. Cross sectional view of the Hanford conduit hose.

Initial characterization of the Continental ContiTech Viper™ hose for use at Savannah River was performed in the mid-2000s by Savannah River National Laboratory (SRNL). The purpose of that work was to find the limitations of this hose for use in radioactive environments. Some data gaps were identified at that time and in recent investigations. The purpose of this work was to fill in those data gaps, provide additional information, and assist SRR by making recommendations.

Among various objectives of this work was to provide burst pressure data after 0.10, 0.25 and 0.50 MGray of gamma total dose, test for any dose rate effects, and do Time Temperature Superposition (TTS) to characterize aging in the ambient environment in between periods of liquid waste transfer. It has been well documented that the rate of gamma dose can have a large effect on service life of polymers [1] [2] [3]. Diffusion Limiting Oxidation (DLO) can occur. At high gamma dose rates, the oxidation rate is faster, resulting in depletion of oxygen in the bulk of the polymer. Diffusion rates are not fast enough to replenish oxygen in the polymer and the rate of degradation decreases. This can result in predictions of polymer service life that are longer than the actual service life.

METHODS

Aging Methods

SRR provided SRNL with a calculation that determined the dose rate delivered to the surface of the conduit hose during transfer of liquid waste between tank 13/15. The dose calculated was 0.027 kGray/hr. (2,713 rad/hr.), with 99% of that coming from beta and 1% coming from gamma. A good bit of time was spent in trying to find a source of beta radiation to use in accelerated aging tests, this included commercial e beam generators. No readily available beta irradiation methods were discovered due to limitations in source capacity and very high dose rates.

Gamma Irradiation was used in earlier studies at SRNL and are an industry standard to determine material degradation due to ionizing radiation. In 2008 SRNL dosed XLPE samples obtained from the hose manufacturer and whole hose samples in the SRNL Co-60 Gamma Irradiator at a rate of 4.00 kGray/hr. (400,000 rad/hr.) to total dose levels of 0.50, 1.00, 2.50, and 3.00 MGray (50, 100, 250, and 300 Mrad). High dose rate and high total dose were used to find the limits of the hose within a reasonable irradiation time period. The possibility of dose rate effects was noted at that time.

The Co-60 Gamma Cells at SRNL were available and comparisons to previous work in 2008 could be made. Accelerated gamma aging was performed at dose rates of 0.05 and 1.00 kGray/hr. (5,000 and 100,000 rad/hr.). These dose rates were chosen to determine if dose rate affects were occurring that would result in overestimating service life of the hose due to DLO. One reference [3] indicates that dose rate effects are not seen below a rate of 0.05 kGray/hr. This is also on the same order of magnitude as the calculated dose rate provided by SRR that the in service hose experiences during transfer (0.027 kGray/hr.). The Co-60 source in the irradiator had not been replaced in a while and 1.00 kGray/hr. was the maximum dose rate attainable.

Time Temperature Superposition (TTS) studies were performed on both XLPE samples from the vendor and whole hose samples. This involved aging samples in convection ovens at temperatures of 130, 110, 90, and 70^{O} C for various lengths of time. Time lengths were chosen based on the tensile strength of XLPE at these temperatures [4]. It was assumed that hose rupture would be caused by a break in the XLPE inner liner.

Test Methods

Various tests were used to characterize degradation of the hose. These included the following tests on XLPE inner liner samples obtained from the vendor: tensile strength; % elongation at failure; durometer hardness, Fourier Transform Infrared (FTIR) spectroscopy, solvent swell and crosslink density (xlink density). In addition, whole hose burst strength was tested on gamma and thermally aged hose samples. This article will focus on two of the most important: tensile strength of XLPE (ASTM D638), and whole hose burst strength (ASTM D380).

RESULTS

Tensile Strength of XLPE

Samples of whole hose were tested for burst strength after gamma irradiation in 2008 and results are plotted in Figure 4 along with tensile strength of XLPE samples obtained from the vendor in 2008 and irradiated at SRNL. In addition, some of these tensile samples were immersed in 25% NaOH at 200^{O} C for 30 days after irradiation. Linear equations best fit the XLPE tensile strength data and an exponential equation best fits burst strength data that goes up to a high total dose of 3.00 MGray. From this set of data from 2008 it appears that the tensile strength of the XLPE inner liner has a significant effect on burst strength of the hose. Early in this study it was decided to use Burst Strength of the whole hose as part of the criterion for determining service life limitations. Bursting of the whole hose accounts for contributions from all components of the composite industrial hose which includes inner layer, reinforcing layer, and outer layer

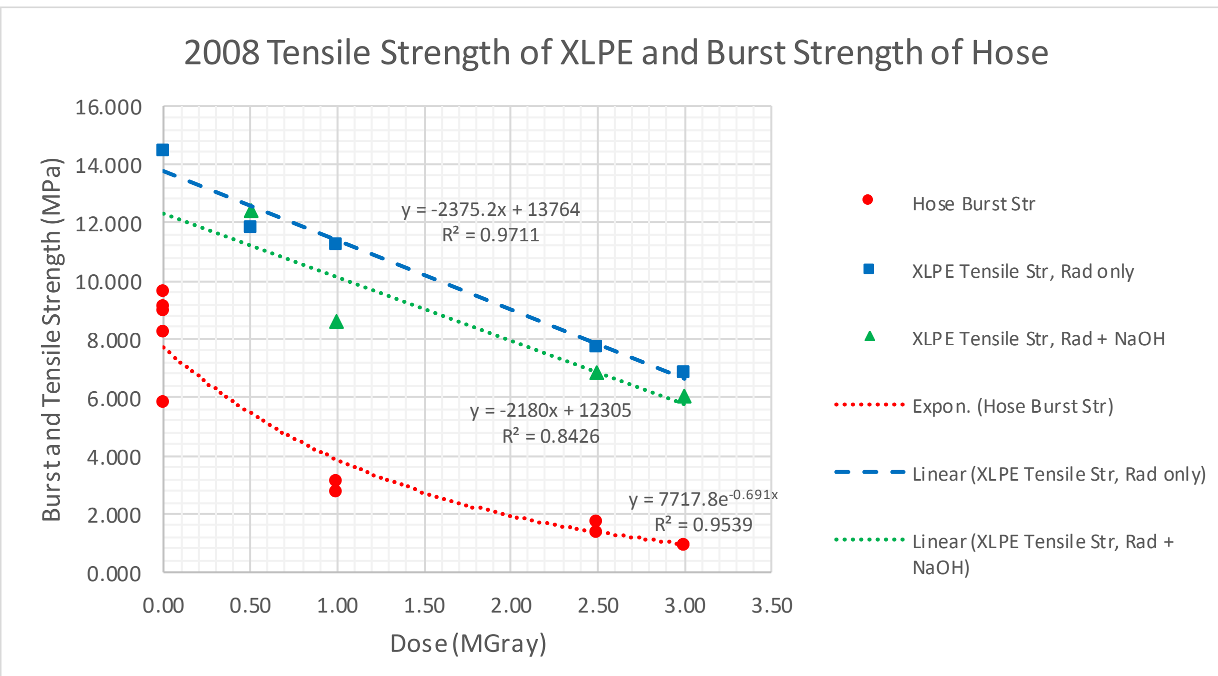

Figure 4. Burst strength of whole hose and tensile strength of XLPE samples tested in 2008. Dose rate used was 4.00 kGray/hr.

The testing in 2008 helped SRR qualify these composite polymer hoses for use as potential replacement for flexible steel hoses as flexible jumpers for a limited service life of 6 months. Further testing was recommended to fill in data gaps identified in the previous testing and for evaluation of the current HIH system between tank 13 and 15.

The industry standard recommended hose design ratio [5] for minimum burst pressure to maximum working pressure is 4:1. Since the maximum working pressure of the hose, as recommended by the manufacturer [6], is 1.38 MPa (200 psi) the minimum burst pressure should not go below 5.52 MPa (800 psi). As seen in Figure 4, burst strength of 5.52 MPa was reached at 0.50 MGray of dose in testing that occurred in 2008. Testing of this current work focused on filling in the gap of information between 0 and 0.50 MGray.

Figure 5 shows a graph of tensile strength versus total dose of XLPE vendor samples obtained in 2020 along with tensile strength of XLPE vendor samples from 2008. The tensile strength of XLPE samples in 2020 appear to be different than the samples in 2008. Tensile strength of the 2020 XLPE as is (before irradiation) is about 20% lower than the 2008 XLPE samples as is. After 0.50 MGray of total dose 2020 XLPE samples are about 27% lower in tensile strength than 2008 samples. This along with other differences observed (ex. FTIR analysis) indicates there may have been a change in the XLPE inner liner of the hose. There is not much difference in tensile strength between samples that were irradiated at 1.05 kGray/hr. dose rate and those irradiated at 0.05 kGray/hr. This is true for samples tested after 0.10, 0.25, and 050 MGray total dose. It appears that dose rate effects do not affect tensile strength of the 2020 XLPE liner.

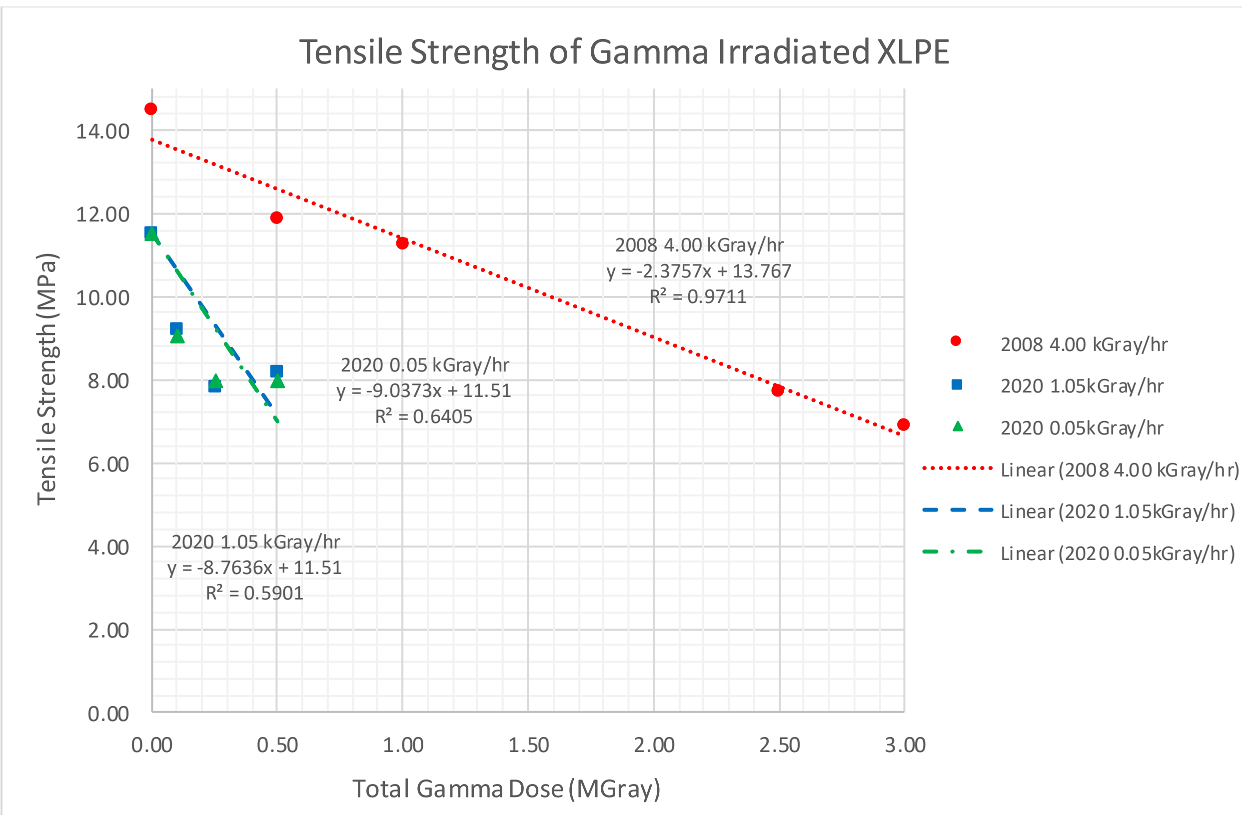

Figure 5. Tensile strength at failure for 2008 and 2020 XLPE inner liner samples.

Figure 6 shows a graph of % elongation at failure versus total dose of the same samples tensile tested in Figure 5. % elongation behavior of the 2020 XLPE samples appear to be different than the % elongation behavior of the 2008 samples. % elongation of 2020 samples as is are about 290% (or 2.9x) greater than the 2008 samples. This is a large difference, the 2020 XLPE is quite a bit more flexible than the 2008 XLPE. In addition, dose rate effects are observed in % elongation of 2020 samples that were irradiated at

1.05 kGray/hr. and 0.05 kGray/hr. There is a greater decrease in % elongation seen at the lower dose rate; this is consistent with (DLO).

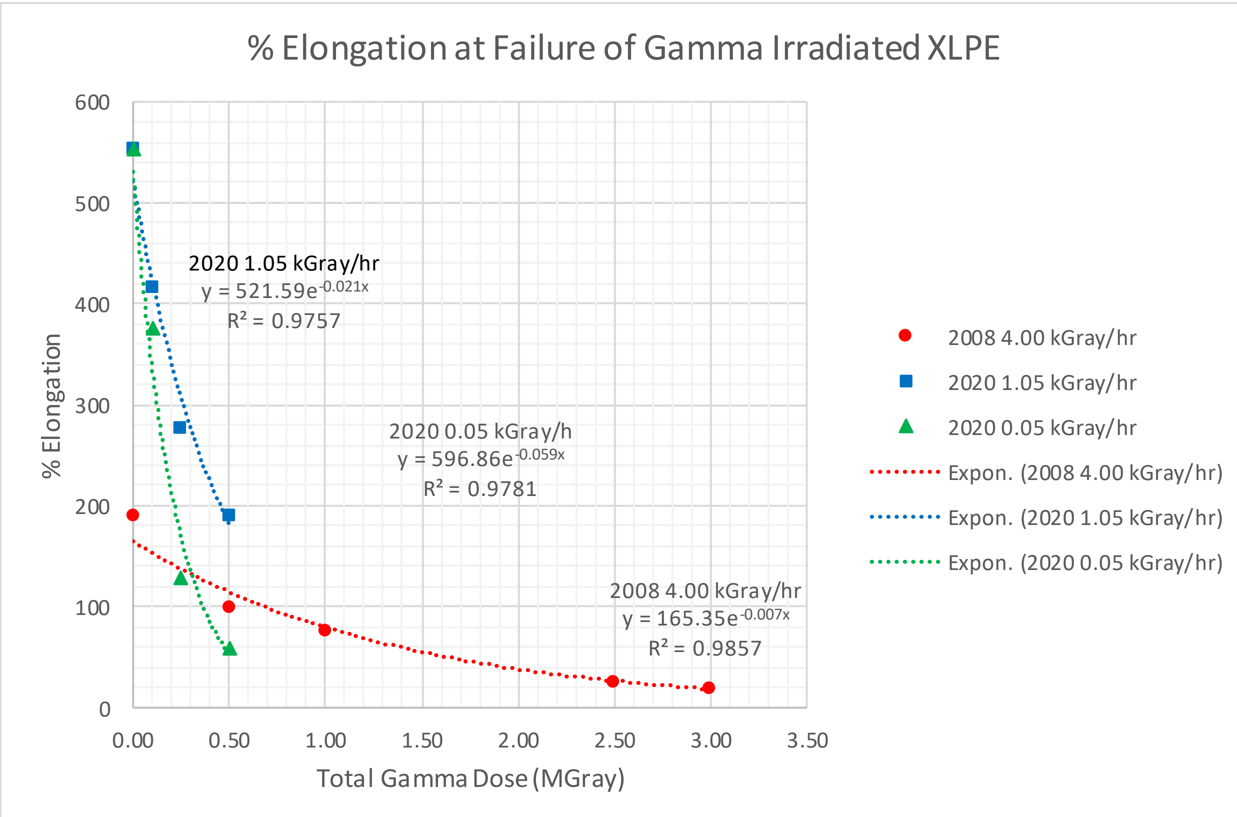

Figure 6. % elongation at failure for 2008 and 2020 XLPE inner liner samples.

Tensile strength testing was also done on XLPE samples that were thermally aged at elevated temperatures. Unfortunately, machine error occurred during this testing and these results are not presented.

Burst Strength of Gamma Aged Hose Samples

In 2008 high dose levels were chosen to determine the limitations of the hose. As previously mentioned, the recommended minimum allowable burst strength should be 4 times greater than the maximum working pressure [5]. Since the maximum working pressure of the hose is 1.38 MPa (200 psi) the minimum allowable burst pressure is 5.52 MPa (800 psi). Figure 7 shows burst strength data generated in 2008 and 2020. In theory, by extrapolating 2008 burst strength pressure data, 5.52 MPa burst strength will occur after aging the hose to 0.50 MGray total dose. Burst testing of 2020 hose was done after irradiating to levels of 0.10, 0.25, and 0.50 MGray to fill in the data gap between 0 and 0.50 MGray

It appears from Figures 7 and 8 that any possible changes made to the hose after 2008 have resulted in higher burst strengths at the lower total dose levels tested in 2020. Figure 8 is simply an enlargement of the data between 0 and 0.50 MGray. After 0.10 MGray total dose the tested burst strength of 2020 hose is 15% higher than the 2008 extrapolated value (8.25 versus 7.20 MPa), at 0.25 MGray 2020 burst strength is 24% higher (8.05 versus 6.50 MPa) than the extrapolated 2008 value, and at 0.50 MGray 2020 burst strength is 38% higher (7.60 versus 5.50 MGray).

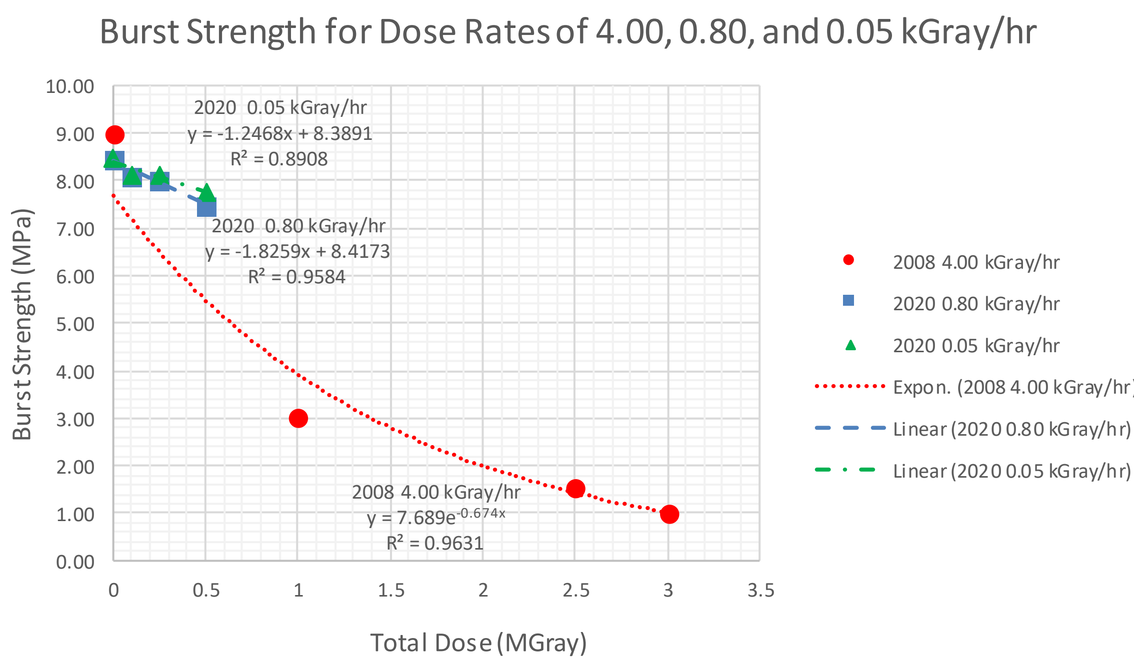

Figure 7. Burst strength of gamma irradiated hose samples in 2008 and 2020.

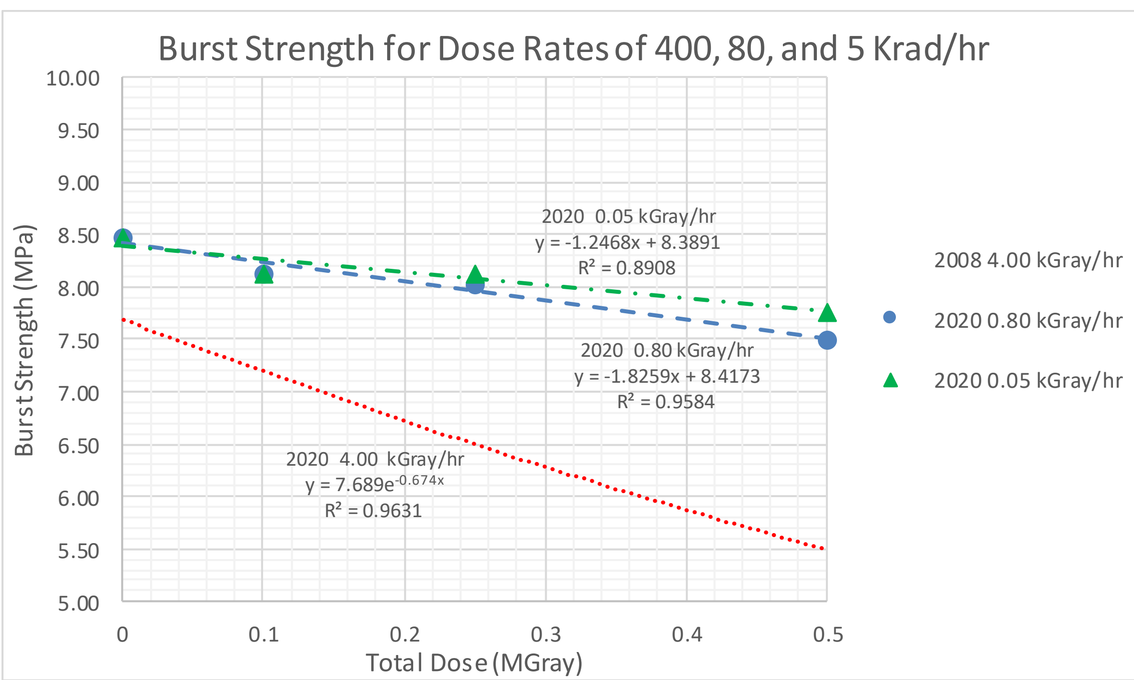

Figure 8. Enlargement of Figure 7 between 0 and 0.50 MGray total dose.

It is believed that one reason for the increase in burst strength is due to an increase in toughness of the XLPE inner layer in 2020. Toughness is used to describe materials that undergo plastic deformation under stress, it is the amount of energy per unit volume that a material can absorb before rupture and is determined by measuring the area under the stress strain curve of the material. Even though tensile strength at rupture is lower for 2020 XLPE samples the % elongation is larger for 2020 samples and therefore the area under the stress strain curve is larger. This is illustrated in Figures 9 and 10. One representative sample plot was selected from the three samples that were tested for each condition.

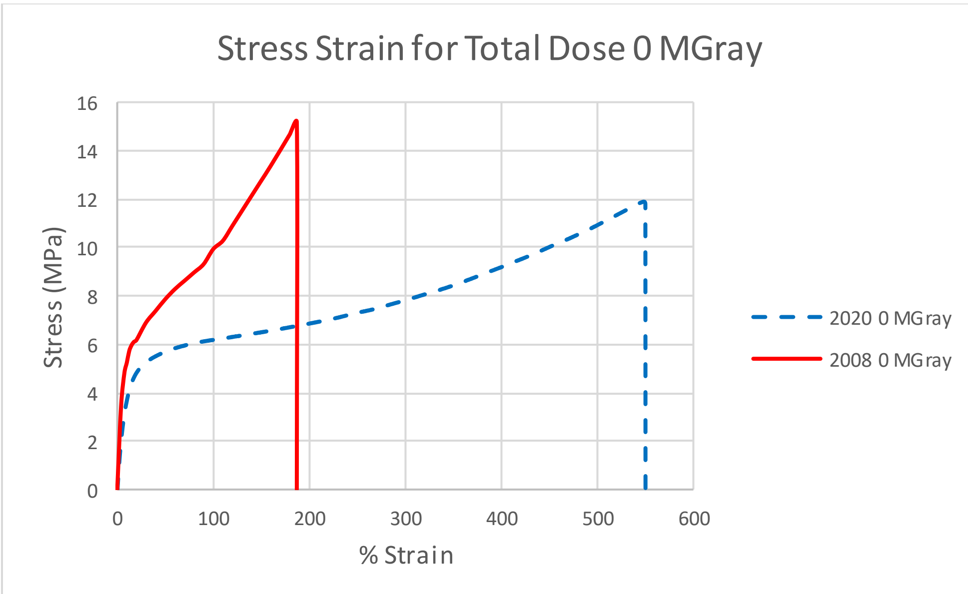

Figure 9. Representative stress strain curves of 2008 and 2020 XLPE vendor samples provided. 0 MGray of total dose.

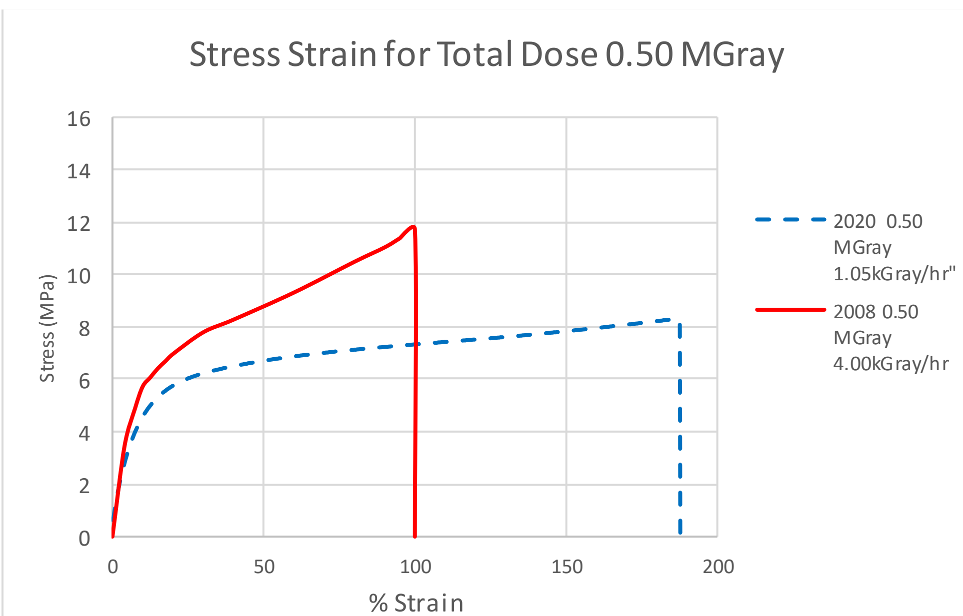

Figure 10. Representative stress strain curves of 2008 and 2020 XLPE vendor samples provided. 50 MGray of total dose.

Burst Strength of Thermally Aged Hose Samples

Time Temperature Superposition is a common accelerated aging method used to characterize degradation of polymeric materials [7] [8]. Figure 11 shows the results of burst strength testing of hoses that were aged in ovens at 130^{O}, 110^{O}, 90^{O}, and 70^{O} C for the time periods shown on the graph. Estimates for temperatures and time lengths were taken from literature data for thermal degradation of XLPE [4]. Degradation of hose samples occurred slower than expected for samples aged at 90^{O} and 70^{O} C.

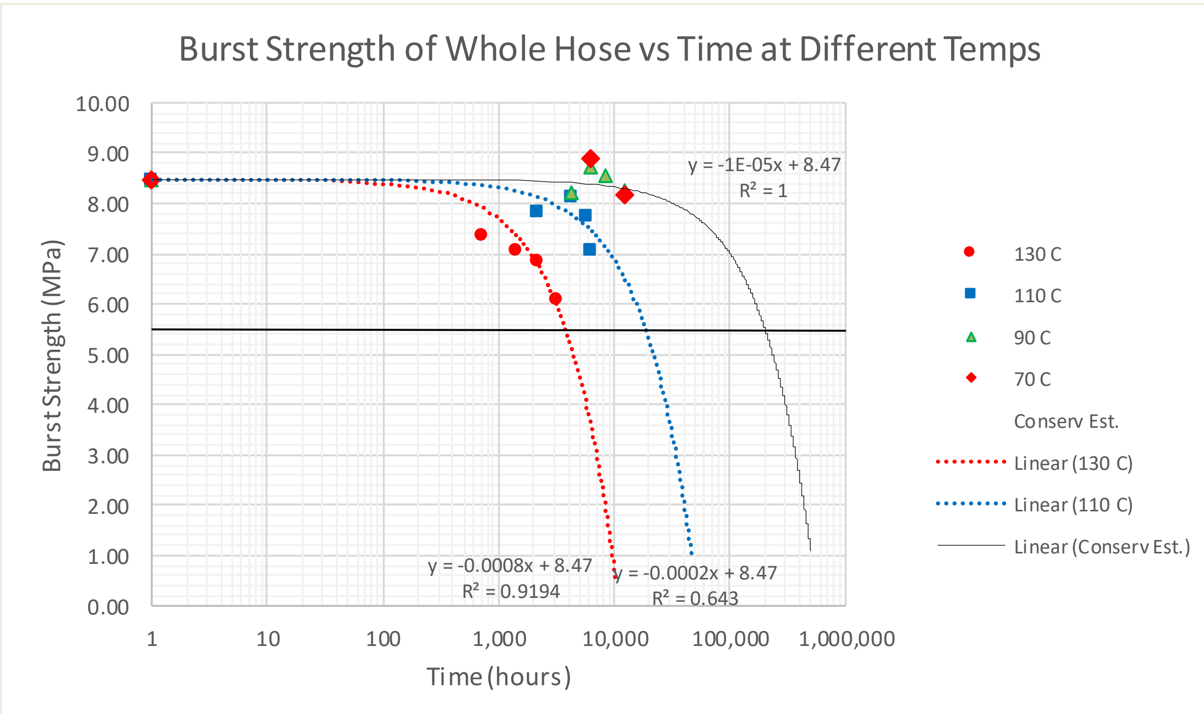

Figure 11. Burst testing of hose samples at various times and temperatures.

The objective of TTS is to use a shift factor (a_T) to fit the data at higher temperatures down to the lowest temperature tested, in this case 70^{O} C. A master curve can be created at the lowest temperature where the data points will extend down to low burst strengths. Determining what the master curve would look like at ambient temperatures (like 30^{O} C) can be done by using an Arrhenius plot of the log of the shift factors (a_T) versus 1000/T (K).

After burst testing of samples aged 17 months (12,504 hrs.) at 90^{O} and 70^{O} C it was determined that there was not a large enough downward trend at these two temperatures to be able to shift all four curves down to 70^{O} C. Two more sample groups remained in the 70^{O} C oven and the next removal date was reset for one of the groups to 36 months. Two additional sample groups were replaced in the 90^{O} C oven with one to be removed at 36 months as well. The removal date for the last of the 70^{O} and 90^{O} C samples will be determined after the 36 month samples are burst tested. One additional sample group was placed in the 130^{O} C oven as well to obtain one data point below the 5.52 MPa burst strength criterion. This will be removed after 7 months (5,088 hrs.).

The curve labeled 'Conserv Est.' is a best guess at coming up with a master curve so that some estimation of decrease of burst strength properties can be determined and therefore account for degradation of the hose at ambient conditions between transfers. Explanation of the use of burst strength for service life prediction is discussed in the next section 'Mode of Failure and Service Life Criterion'.

MODE OF FAILURE AND SERVICE LIFE PREDICTION

Hoses that were burst were also cut in two in the length direction and the mode of failure was discovered. This is illustrated in the photos of Figure 12, which are higher magnifications of the same picture. As the Figure shows, after burst testing there is an indentation line that appears on the surface of the XLPE inner liner, the indentation is identified as 'Fracture line' in the photos. Just looking at the inner surface of the XLPE it is difficult to determine if this indentation goes through the entire cross section of the XLPE liner. Upon closer inspection of the hose cross section at the cut line these indentations or fractures go through the entire thickness of the XLPE inner liner. This is seen in the two bottom enlargement photos of the top photo.

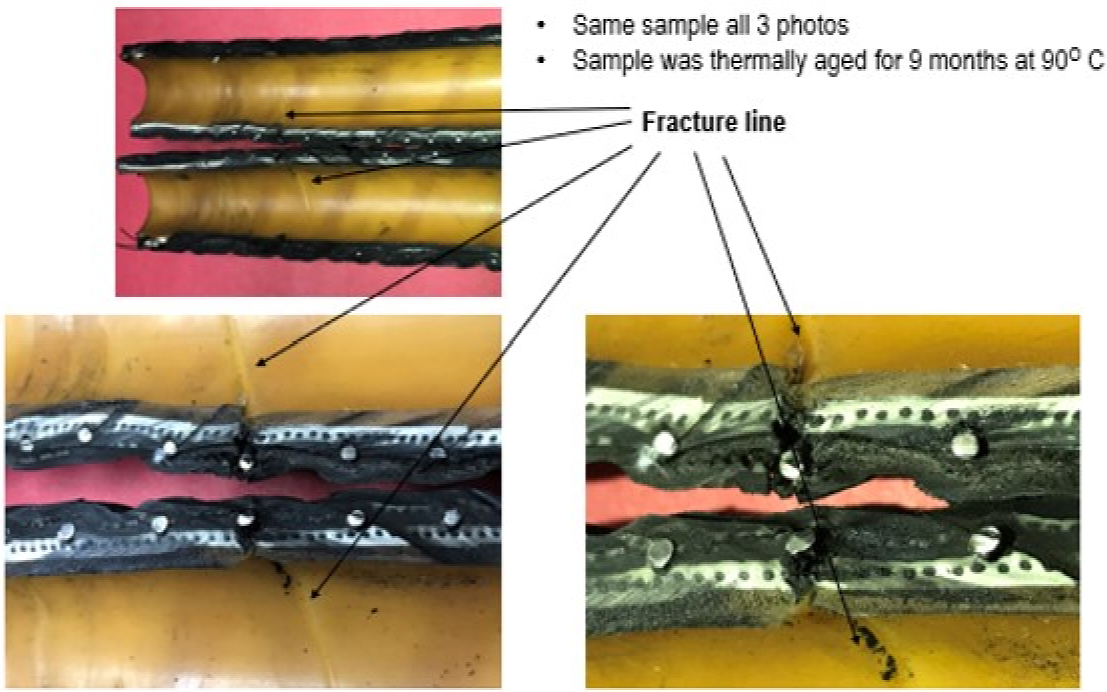

Figure 12. Illustration of failure mode for hose aged at 90° C for 9 months. This was mode of failure in every test group burst tested

Three hose samples were removed at each time and temperature, therefore, burst testing was done in triplicate. One hose sample from each condition was then split in two. This failure mode was observed on every hose split in two after burst testing. The indentation or fracture line seen at the surface of the XLPE liner follows along the same path as a helical wire. Failure is due to impingement of the XLPE liner on the high modulus metal wire when the hose is under pressure from the aqueous hydraulic fluid. The metal wires and braided fabric help prevent large elongations of the XLPE liner.

Service life criterion is derived by using burst strength data from gamma aged and thermally aged whole hose samples. Burst strength was chosen as the criterion because it accounts for contributions from each component of the composite Viper™ hose; the XLPE inner liner, the reinforcing layer (braided fabric and helical metal wire), and the EPDM outer covering. It is the interaction of all these components that contribute to preventing burst failures in the hose. The high modulus (stiff) reinforcing layer restrains the XLPE from going to large elongations under pressure.

The effect of hose degradation by radiation plus caustic NaOH solution during transfers and the effect of degradation under ambient conditions are additively combined into the equation below:

$$y = 1228 + \left((-5.0032)x_1\right) + ((-0.0020)x_2)$$

where: y = burst strength; not to go below 800 psi (converts to 5.52 MPa)
x_1 = total dose (Mrad is used) during transfer.
x_2 = time (hours) between transfers when hose is in ambient conditions.
1228 psi = burst strength with no aging or the y intercept of the two linear components (converts to 8.47 MPa).

The equation above reflects the equation used at the Savannah River Site and is based on English units, SI units are shown in parenthesis. The criterion of 5.52 MPa (800 psi) as the minimum allowable burst pressure is taken from the Association for Rubber Products Manufacturers (ARPM) Hose Handbook [5]. The recommended hose design ratio for the minimum burst pressure to maximum working pressure is 4:1. Since the 1" ID Viper™ conduit hose is rated for a maximum working pressure of 1.38 MPa (200 psi) the minimum burst pressure criterion was set to 5.52 MPa (800 psi). When x_1, the combined total dose from all intermittent transfers and x_2, the time that the hose sits in ambient conditions between transfers (hours) are plugged into the equation the resulting calculated burst pressure should not go below the 800 psi (5.52 MPa) criterion.

The first term of the equation above ({-5.0032} x_1) reflects the decrease in burst strength due to radiation and caustic chemical. The slope of -5.0032 was determined by taking the average of the burst data at 0.80 and 0.05 MGray/hr. dose rates (data points at 0.10, 0.25, and 0.50 MGray total dose) for 2020 hose in Figure 8 and decreasing them by 10%. The basis for doing this is illustrated in Figure 4 data for the 2008 hose testing. In 2008 XLPE samples were tensile tested after gamma irradiation. Additional samples not tensile tested after gamma irradiation were then immersed in 25%NaOH at 93^{O} C for 30 days and then tensile tested. It was found that there was about a 10% drop off in tensile strength from the XLPE samples that saw both irradiation and exposure to the 25% NaOH at 93^{O} C for 30 days versus only irradiation. These results are shown in Figure 4. The assumption is made that a 10% drop off in tensile strength of the XLPE liner will result in 10% decrease in hose burst strength.

Testing by Florida International University (FIU) showed similar caustic chemical effects on burst testing of hose used in the Hanford HIH system. The Hanford hose was subjected to 25% NaOH at a temperature of 54^{O} C for 12 months and there was a 6% drop off in burst strength from the as is unaged hose [9]. 12 months (8,760 hrs.) exposure of SRR liquid waste would result in a total dose of 0.238 MGray delivered to the SRR hose based on the calculated dose rate of 0.027 KGray/hr. provided by SRR. Conclusion #2 in the next section shows that there is a total dose limit of 0.25 MGray imposed on the SRR HIH, this is very similar to the total dose delivered if SRR liquid waste were used in the FIU experimental pumping equipment for 12 months It should be noted that in the FIU study at a temperature of 77^{O} C and exposure time of 12 months the drop off in burst strength went up to 29%. In addition, the inner layer of the Hanford hose is Ethylene Propylene Diene Monomer (EPDM) rubber.

The second term of the equation above, ({- 0.0020} x_2), reflects the decrease in burst strength due to the time between transfers or degradation in ambient conditions over time. This is simply a conservative estimate of the linear slope for decrease in burst strength with time by looking at the trends for burst strength of hose removed from the 130^{O} and 110^{O} C ovens. This is shown in Figure 11, note that Figure 11 is a semi log plot, if the x axis was not on a log scale the curves would be linear.

CONCLUSIONS

1) A criterion equation has been developed that attempts to account for caustic, ionizing radiation, and time/temperature effects. It is based on data obtained from burst strength testing of hoses that have seen accelerated aging under gamma radiation, accelerated aging at elevated temperatures, and exposure to caustic chemical. The equation is shown below:

$$y = 1228 + \left((-5.0032)x_1\right) + ((-0.0020)x_2)$$

where: y = burst strength; not to go below 800 psi (converts to 5.52 MPa)
x_1 = total dose (Mrad is used) due to irradiation plus caustic effects during transfer.
x_2 = time (hours) between transfers when hose is in ambient conditions.
1228 psi = burst strength with no aging or the y intercept of the two linear components (converts to 8.47 MPa).

2) Assumptions are made in using the criterion equation above that is based on accelerated aging conditions. The author's judgement of whether the assumption will lead to an overestimation of service life and no failure would occur is indicated as conservative. If the assumption is judged to lead to an underestimation of service life and therefore possible premature failure could occur, this is indicated as nonconservative.

Assumptions include the following: degradation caused by directly ionizing radiation (electrons from beta) is like indirectly ionizing radiation (photons from gamma), conservative; use of 5.52 MPa as the criterion burst strength when 1.38 MPa is the working pressure of the hose, conservative; estimation of 0.002 as the slope of the burst strength versus time TTS master curve, conservative; there is no consideration of synergistic effects between caustic chemical, ionizing radiation, and temperature, nonconservative; environmental stress cracking is not significant (nonconservative). It is believed that there are enough conservative estimations and assumptions made so that the equation will not underestimate service life.

3) Data is now available that allows increases in total dose and time limits for the current HIH system installed in 2016. The total dose limit increases from 0.10 MGray to 0.25 MGray . The upper time limit goes from 6 years to 10 years.

4) Dose rate effects were observed in some physical properties (like % elongation) but not in the criterion property of burst strength. This is likely due in part to the restriction that the reinforcing layer puts on hose elongation in the radial direction.

5) It is highly recommended that some type of forensics testing be done on the current HIH system in place between tanks 13/15 when the system is replaced. One reason would be to see how reasonable the equation is for predicting service life. If nothing else a visual inspection for discoloration of the XLPE inner liner and microscopic inspection for cracking should be made on the old hose that has been cut in two. Test method development is currently underway to develop a durometer hardness test on curved hose sample and an FTIR method to determine oxidation levels throughout the cross section of the XLPE inner liner.

6) It is also recommended that the Time Temperature Superposition studies be completed as described in this paper. This will provide a much better estimate of the decrease in burst strength attributed to time between transfers under ambient conditions.

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