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Long-term Performances of New and Regenerated Polyethylene Membranes Used as Vapor Barriers in Temperate Climates

Rasmussen, Torben Valdbjørn; Hansen, Tessa Kvist; Nielsen, Jens Kromann; Steenstrup, Frederik R.; Ottosen, Lisbeth M.; Petersen, Louise Green; Shashoua, Yvonne

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Long-term performances of new and regenerated polyethylene membranes used as vapor barriers in temperate climates

ABSTRACT

Long-term performances were assessed for nine polyethylene (PE) membranes used as vapor barriers and comprising virgin PE, 100% new PE, regenerated PE and multilayered virgin and regenerated PE. The membranes were evaluated and compared using standard laboratory tests described by the standard EN 13859-1. Materials were assessed using five types of chemical analyses and seven physicomechanical tests. Chemical and mechanical properties were determined before and after exposure to an ageing regime comprising 168 days at 70 °C in total. Although the suppliers described the membranes only by the general term polyethylene, the chemical analysis suggested that all were based solely on low-density polyethylene, LDPE. Suppliers of the membranes stated that regenerated polyethylene was sorted, washed, cleaned, and enhanced with additives, including stabilizers. The study suggested that this treatment produced membranes with bulk properties similar to those of virgin and 100% new LDPE. However, visual examination of all membranes containing regenerated polyethylene showed additional particulate inclusions. This suggested that it is currently difficult to make membranes from regenerated polyethylene without physical defects. This study indicated that the content of regenerated PE in membranes, evaluated in this study, were not a significant factor in its performance. Importantly, measurement of their resistance to water vapour before and after being exposed to an ageing regime, indicated that all the membranes evaluated would perform satisfactorily as vapor barriers in buildings.

Torben Valdbjørn Rasmussen is a senior scientist in the Department of Civil Engineering and Construction Management, BUILD, Aalborg University, Copenhagen, Denmark. **Tessa Kvist Hansen** is an assistant professor in the Department of Civil Engineering and Construction Management, BUILD, Aalborg University, Copenhagen, Denmark. **Jens Kromann Nielsen** is a professional leader in the Department of Plastics and Packaging Technology, Danish Technical Institute, Taastrup, Denmark. **Frederik R. Steenstrup** is a section leader in the Department of Plastics and Packaging Technology, Danish Technical Institute, Taastrup, Denmark. **Lisbeth M. Ottosen** is a professor in the Department of Civil Engineering, Technical University of Denmark, Lyngby, Denmark. **Louise Green Petersen** is a PhD student in the Department of Civil Engineering, Technical University of Denmark, Lyngby, Denmark. **Yvonne Shashoua** is a professor in the Department of Environmental Archaeology and Material Research, National Museum of Denmark, Kongens Lyngby, Denmark.

INTRODUCTION

Polyethylene (PE) membranes are used as vapor barriers. In the past, it was common for PE membranes to be made from pure (virgin or 100% new) PE, but today, an increasing number of membranes are offered that contain regenerated or a mixture of regenerated and pure polyethylenes. As part of the increased interest in sustainability in construction, it is expected that demand will increase for membranes made from regenerated PE or a mixture of pure and regenerated PE.

Virgin PE describes material that has been fabricated only once. 100% new PE describes material that has not been used after fabrication as well as cuts and residual material from the production of virgin PE. 100% new PE contains a proportion of the additives from the production of virgin PE. The quantity and the types of additives in 100% new PE varies. Regenerated PE is fabricated from collected, post-use plastic, also called recycled plastic. The plastic is sorted, cleaned and washed before it is melted, and additives are added. A combination of pure and regenerated PE are used to fabricate a layered membrane. The base layer comprises regenerated PE and thinner layers of 100% new PE or virgin PE are added to one or both sides of the base layer.

Vapor barriers in temperate climates are mounted on the warm side of the thermal insulated exterior building envelope, see figure 1. Knowledge and experience of pure PE vapor barriers has been acquired through years of use. However, knowledge and documentation of the performance and, in particular, long-term performance of vapior barriers containing regenerated PE membranesis lacking (Rasmussen et al., 2018).

Figure 1   Exterior timber stud wall, where the vapor barrier (shown in red) is mounted on the warm side of the thermally insulated exterior building envelope. The webs of the membrane must overlap on a solid surface and be joined together with adhesive or tape (shown in grey)

This paper identifies, evaluates and compares the compositions, durability and long-term performances of vapor barrier membranes comprising virgin PE, 100% new PE, regenerated PE and one membrane layered with virgin and regenerated PE. Nine different commercially-available membranes were selected for examination, and purchased. Purchased membranes were selected from those most commonly used by contractors. Samples taken from each membrane were assessed using chemical analyses and physicomechanical tests. Tests were performed to determine the material properties and stabilities of the membranes. Most of the tests were performed both before and after ageing. In addition, visual examination of membranes containing regenerated PE was performed. The ageing regime used comprised 168 days in total at 70 °C. Based on the tests and the ageing, the service life of the membranes, in relation to material properties, chemical content and physical stabilities, their effectiveness as vapor barriers in constructions with long service lifetimes, was estimated.

METHOD

Samples taken from each membrane were assessed using five types of chemical analyses and seven physicomechanical tests. All test methods were based on standardised methods described by the standard EN 13859-1 and in ISO or DIN standards. The exceptions were the chemical analyses that followed standard laboratory protocols performed with specialized equipment and visual examination. Tests were performed to determine the material properties and stabilities of the membranes. Most of the tests were performed both before and after ageing.

Materials

Nine different PE membranes from nine different vapor barrier systems were selected for the study. The different PE membranes comprised two fabricated from virgin PE, two from 100% new PE and five from regenerated PE. One of the five membranes of regenerated PE, according to its manufacturer, comprised a base layer of regenerated PE between two layers of pure PE. The tested membranes were selected from those most commonly used by contractors in Denmark. One roll of each membrane was purchased from retail building material businesses located randomly in Denmark to maintain independency from the suppliers. An additional five rolls of each of the membranes containing regenerated PE were purchased to conduct visual examination. Each roll contained between 250 and 500 m^2 of membrane. The properties of the membranes (identified by ID numbers) provided by the manufacturers are shown in Table 1. Membranes comprising virgin PE were identified as T7482-1 and T7482-2. Membranes comprising 100% new PE were numbered as T7482-3 and T7482-4. Membranes comprising regenerated PE were known as T7482-5, T7482-6, T7482-7, and T7482-9. One membrane, denoted as T7482-8 comprised three layers of PE, a base layer of regenerated PE with layers of virgin PE, one on each side.

Table 1.   Properties of membranes evaluated in the study as supplied by manufacturers.

ID	Service life	Membrane material	Thickness [mm]
T7482-1	> 30 years	Virgin PE	0.20
T7482-2	50 years	Virgin PE	0.12
T7482-3	50 years	100% new PE	0.20
T7482-4	> 15 years	100% new PE	0.20
T7482-5	> 20 years	Regenerated PE	0.15
T7482-6	> 15 years	Regenerated PE	0.20
T7482-7	> 20 years	Regenerated PE	0.20
T7482-8	> 30 years	Regenerated PE and pure PE	0.20
T7482-9	Not declared	Regenerated PE	0.20

Ageing

All tests were performed on unaged as well as aged samples, except for the chemical tests comprising Beilstein test, X-ray fluorescence spectroscopy (XRF) and the Loss on Ignition test which were only conducted on unaged samples. Ageing was performed by placing the samples for 84 days in a climate chamber at 70 °C and 90% relative humidity followed by 84 days at 70 °C at less than 10% RH.

Chemical Analyses

Five samples of each membrane were analysed to determine their chemical structures both before and after ageing. Beilstein, X-ray fluorescence spectroscopy (XRF) and Loss on Ignition tests were only conducted on unaged membranes. The analyses performed are shown in Table 2.

Table 2.   Chemical analyses of membranes

Analysis type	Performed on unaged samples	Performed on aged samples
Attenuated Total Reflection - Fourier Transform Infrared (ATR-FTIR) spectroscopy	X	X
Acid-Detection (A-D) indicator strips	X	X
Beilstein test	X	
X-ray fluorescence spectroscopy (XRF)	X	

Attenuated Total Reflection - Fourier Transform Infrared (ATR-FTIR) spectroscopy was used to identify the chemical structure of plastic present in each membrane and to evaluate their chemical stabilities. To investigate whether samples showed signs of chemical degradation over time, ATR-FTIR spectra of aged samples were compared with those of unaged samples (Rasmussen et al., 2020).

Acid-Detection (A-D) indicator strips were used to show the release of organic acids as gases from membranes which indicate a potential reduction in service life of the materials. A-D strips are paper-based indicators that use the pH-sensitive indicator bromocresol green to detect the presence of acids in all materials, including plastics. When A-D strips are placed in a closed container with the specimen, the strips change colour from blue at pH 5.4 to green (slightly acidic) and then to yellow at pH 3.8 (highly acidic) if acid gases are released. The color changes within 24 hours (Nicholson and O'Loughlin, 1996 and Rasmussen et al., 2020).

The Beilstein test (Beilstein, 1872) was used to identifythe presence of the plastic type polyvinyl chloride, PVC in samples. PVC is chemically unstable, can emit toxic plasticizers when it decomposes and is therefor undesirable in building materials designed for longterm use (Rasmussen et al., 2020).

X-ray fluorescence spectroscopy (XRF) was used to determine the elemental composition of samples. XRF is a non-destructive, surface measurement technique that can in principle detect and measure the content of all elements, however with a reduced sensitivity for the lightest elements (Beckhoff et al., 2006). A Bruker Tracer III-V, that detect elements with a higher atomic number than magnesium was used (Rasmussen et al., 2020).

Loss on Ignition was used to quantify the content of organic material in samples. Samples were annealed at a temperature of 550 ° C and weight loss measured. The material remaining is attributed to inorganic additives and impurities in membranes. Polyethylene is fully decomposed at this temperature, whereas mineral impurities will only be decomposed by much higher temperatures. PE is annealed to a weight percent of 0% at 480 ° C (Ali et al. 2005). Samples of 2-2.5 g were cut from each membrane, pre-dried at 105 °C and then annealed at 550 °C in a muffle furnace. The mass of the pre-dried samples (m_0) and the mass after annealing (m_g) were weighed to 4 decimal places. The loss on ignition is defined as weight loss on annealing and calculated as $((m_0 - m_g)/m_0) * 100$ (Rasmussen et al., 2020).

Physicomechanical Tests

Five samples of each membrane (except for the water vapor diffusion resistance test conducted on two samples of each membrane) were analyzed, using seven physicomechanical tests. The tests were performed both on unaged and aged samples of the materials as shown in Table 3.

Table 3.   Physicomechanical tests performed on samples

Test type	Test standard	Performed on unaged samples	Performed on aged samples
Tensile strength, both longitudinally and transversely the production line, as well as fracture elongation and Youngs modulus*	DS/EN ISO 527-3	X	X
Tear strength both longitudinally and transversely the production line**	DS/EN 12310-1	X	X
Water vapor diffusion resistance	DS/EN 1931	X	X

*The tensile strength was determined as the highest measured stress during the tensile test. Elongation at break was determind as the strain at break. Young's modulus was determined at 0.5-1.0% elongation, even though it is not part of the standard used ISO 527-3.
**The towing speed is 100 mm/min.

Visual Examination

Five complete rolls of each membrane containing regenerated PE were examined for cracks, larger fragments and weak areas. Rolles were opened to their full width and length, so they could be fully examined in a single session. The membranes were visually examined in detail, piece by piece.

Cracks, large fragments and weak areas in the individual membrane were cut out and assembled for further classification. The visual examination of membranes thus served to check and compare whether the sorting and purification

of all membranes comprising, regenerated PE and their combinations with pure PE, had been conducted to an acceptable degree under production.

Detailed information on the manufacturing process was not disclosed by the suppliers, but sorting, purifying, washing, and addition of stabilizers were believed to be of crucial importance in producing membranes comprising regenerated PE. Membranes of regenerated PE are extruded from molton granules to which stabilizers are added. Granules have been pre-sorted, cleaned, and washed. Membranes are extruded and folded before being rolled around a cardboard tube in lengths of typically 25 or 50 meters. Rolls were 1.0 m wide and the membranes folded in the middle. During visual examination, membranes were unfolded and pulled over a table, 2 m at a time. When the membrane was on the table, the foldline was examined first followed by the flat surfaces of the membrane itself.

RESULTS

Chemical Analyses

ATR-FTIR spectroscopy. ATR-FTIR spectra of samples of unaged, pure PE, virgin PE (T7482-1 and T7482-2) and 100% new PE (T7482-3 and T7482-4) before ageing were chemically identical to those of reference samples low-density PE (LDPE) run at the same time. All the pure PE samples exhibited characteristic peaks between 3000 and 2840 cm^{-1}, attributed to asymmetric and symmetrical stretching of CH_2 groups, peaks at 1469 cm^{-1} due to deformation in CH_2 groups and at 718 cm^{-1} from rocking vibration of CH_2 groups (Shashoua, 2008). No polymer types other than LDPE were identified in the pure PE samples.

ATR-FTIR spectra of samples containing regenerated PE (T7482-5, T7482-6, T7482-7, T7482-8, and T7482-9) agreed well with reference samples of LDPE. The difference between ATR-FTIR spectra of samples of pure PE and samples containing regenerated PE was that samples containing regenerated PE showed additional peaks between 1500 and 650cm^{-1} attributed to inorganic additives. No polymers other than LDPE were identified in the samples. All samples containing regenerated PE contained calcium carbonate, but T7482-9 contained measurably higher concentrations than the other samples containing regenerated PE.

A-D indicator strips. The results suggested that none of the samples released acid, either before or after ageing.

Beilstein test. The results suggested that none of the samples contain PVC.

XRF. No inorganic elements were detected in samples of pure PE. By contrast, all membranes comprising regenerated PE were found to contain additives based on calcium, titanium, and zinc in the form of calcium carbonate, titanium oxide and zinc oxide.

Loss on Ignition. Loss on ignition for the samples examined are shown in Figure 2. The mean of the two repeat measurements for each membrane is indicated by sample A and sample B.
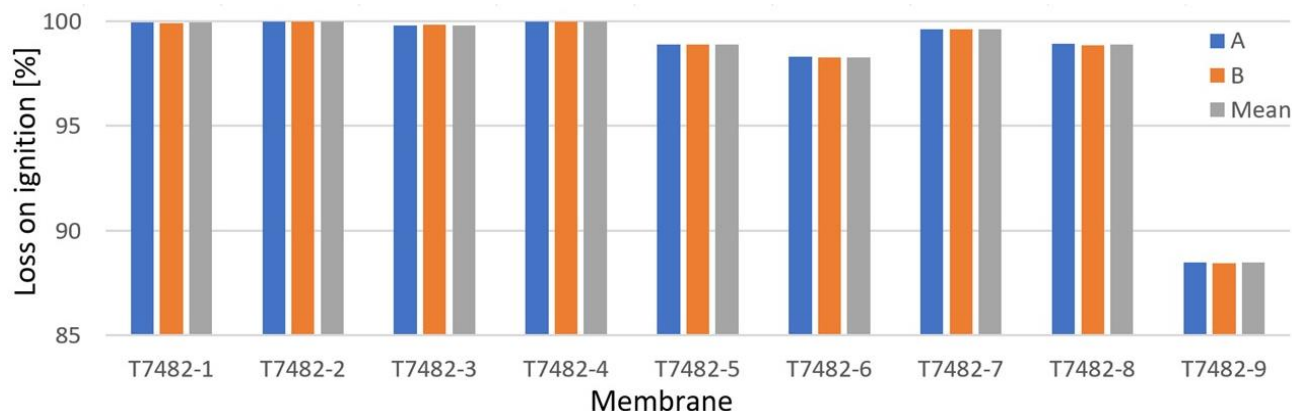
Figure 2   Loss on ignition for the samples examined.

Physicomechanical Tests

Tensile strength parallel to the direction of production as well as fracture elongation and Youngs modulus performed on unaged and aged samples is shown in Table 4.

Table 4.   Properties parallel to the direction of production

ID	Tensile strength [MPa]	Fracture elongation [%]	Youngs modulus [MPa]	Tensile strength* [MPa]	Fracture elongation*[%]	Youngs modulus* [MPa]
T7482-1	17.5	456	105	16.0	402	133
T7482-2	29.0	593	271	24.5	531	210
T7482-3	20.7	402	133	19.2	350	153
T7482-4	16.7	522	177	15.4	485	119
T7482-5	16.4	601	116	16.6	687	106
T7482-6	15.5	480	158	14.9	504	176
T7482-7	18.9	623	162	18.0	646	153
T7482-8	19.4	530	174	17.4	518	165
T7482-9	14.9	556	158	14.0	571	191

* Performed on aged samples.

Tensile strength at right angles to the direction of production as well as fracture elongation and Youngs modulus performed on unaged and aged samples is shown in Table 5.

Table 5.   Properties at right angles to the direction of production

| ID	Tensile strength [MPa]	Fracture elongation [%]	Youngs modulus [MPa]	Tensile strength* [MPa]	Fracture elongation* [%]	Youngs modulus* [MPa]
T7482-1	16.3	688	115	15.1	604	139
T7482-2	28.4	747	274	26.6	688	245
T7482-3	20.3	609	139	18.9	576	133
T7482-4	16.2	652	89	15.3	623	109
T7482-5	16.4	691	156	16.4	718	162
T7482-6	18.7	641	168	18.0	649	165
T7482-7	18.8	688	187	18.7	769	142
T7482-8	18.4	710	203	16.6	670	172
T7482-9	15.1	621	126	16.3	664	180

* Performed on aged samples.

Tear strength both parallel and at right angles to the direction of production performed on unaged and aged samples is shown in Table 6. The measurements are rounded to nearest 5 N, cf. standard 12310-1.

Table 6.   Tear strength

ID	Parallel [N]	Right angles [N]	Parallel* [N]	Right angles * [N]
T7482-1	55	60	60	60
T7482-2	85	85	90	90
T7482-3	80	80	90	90
T7482-4	60	55	60	60
T7482-5	85	80	90	90
T7482-6	115	115	115	115
T7482-7	115	115	130	130
T7482-8	110	110	115	115
T7482-9	75	75	85	85

* Performed on aged samples.

Water vapor diffusion resistance performed on unaged and aged samples are shown in Figure 3.
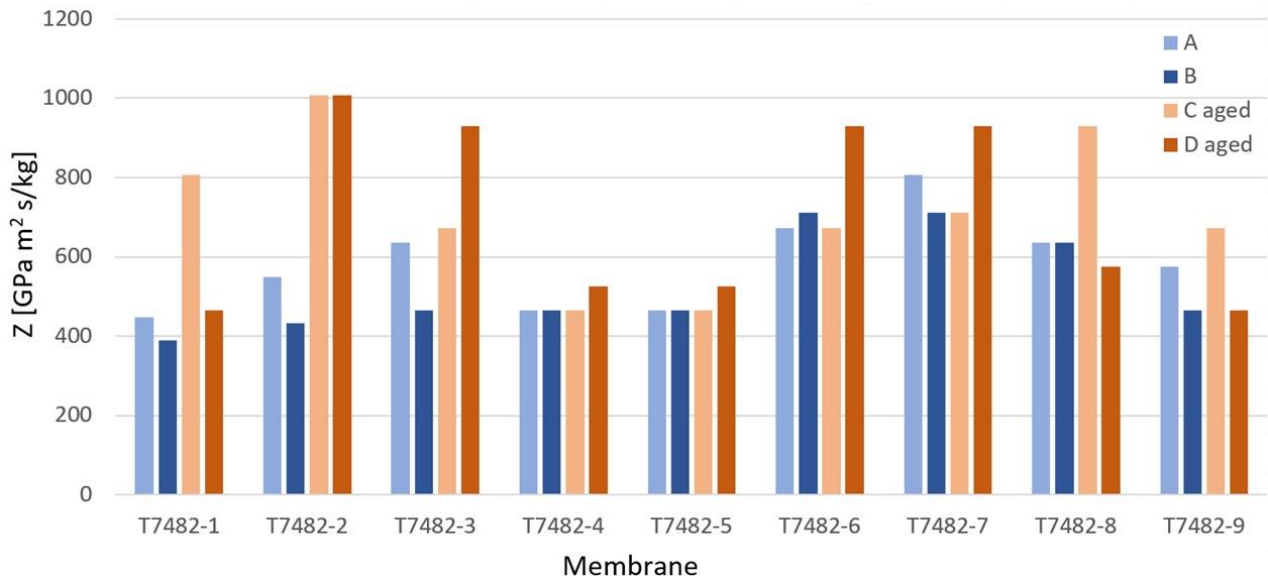
Figure 3 Water vapor diffusion resistance for the samples examined.

Visual Examination

Number of holes and cracks along the foldline for the individual membranes of PE are shown in Table 7.

Table 7. Examination of PE membranes

ID	Holes	Cracks in fold	Fragments	Length [m]	Area [m²]
T7482-5	0	0	Yes	125	250
T7482-6	3	2	Yes	250	500
T7482-7	0	0	Yes	250	500
T7482-8	15	0	Yes	250	500
T7482-9	0	0	Yes	250	500

DISCUSSION

ATR-FTIR spectroscopy was performed on samples taken from all the membranes included in the study. This was done to determine the types of plastics in the individual membranes and to assess their chemical stability upon aging. The method is commonly used to identify chemical compounds in plastics and their degradation products formed by aging. All the membranes, both those comprising pure and regenerated PE, were found to be based on LDPE. No other polymers were found.

Not all inorganic additives can be detected by ATR-FTIR spectroscopy because most absorb infrared radiation at wavelengths that are lower than 500 cm^{-1}, below the sensitivity for a diamond ATR crystal. An exception is calcium carbonate ($CaCO_3$). Calcium carbonate is often added to polyethylene and other types of plastic to increase their strength and reduce opacity with minimal increase in cost. Calcium carbonate strongly absorbs infrared radiation between 1450 and 600 cm^{-1}. Signs of chemical degradation on aging can be detected by the formation of carbonyl groups (C = O) in ATR-FTIR spectra. The formation of carbonyl groups reflects a change in chemistry that results in discoloration of PE. Spectra of the membranes examined showed no signs of carbonyl groups or of chemical degradation.

A-D indicator strips were used to examine the membranes for release of organic acids. Acid can contribute to the formation of chemical instability that reduces its service life. All membranes were tested before and after aging. The presence of acid in the membrane material or its production upon aging were determined. None of the membranes contained acid or produced acid before or after aging. A-D strips were blue before the test and in contact with the samples before and after

aging caused none of them to change color from blue to green or yellow.

The Beilstein test showed negative results, as none of the flames turned green during the test. The results suggest that none of the samples contain PVC. The results confirmed the results from ATR-FTIR spectroscopy showing that all membranes comprised PE as the only polymer.

XRF detected no inorganic elements in membranes of pure PE, virgin PE and 100% new PE. By contrast, the elements calcium, titanium, and zinc in the form of calcium carbonate, titanium oxide and zinc oxide were identified in all samples containing regenerated PE. It seems likely that these elements belong to the fillers added to many polymers to increase strength and reduce opacity. Titanium oxide is also used to impart white coloration and to reflect UV light from the surfaces of the material. Together with zinc oxide, titanium oxide protects the polymer from sunlight and degradation

Loss on Ignition measurements showed high agreement between repeat samples for each membrane. All the membranes with virgin and 100% new PE had lower content of inorganic material (highest loss on ignition) compared to the membranes with regenerated PE. Only membrane T7482-4 comprised 100% organic and no inorganic material. Membrane T7482-9 had a loss on ignition of approximately 88.5% and thereby was the membrane containing the highest quantity of inorganic material.

Tensile Strengths for all the membranes were relatively similar at about 15-16 MPa. The single exception was membrane T7482-2 that showed significantly higher tensile strength, although the membrane was also the thinnest of those studied. The variations between measurements before and after aging were small. Furthermore, it appears that the elongation at break at right angles to the direction of production for all membranes, both before and after aging, is between 550% and 750%, regardless of whether the PE membrane contains pure PE or regenerated PE. No effect of aging can be detected based on Youngs modulus.

Tear strength showed no significant change for membranes on aging. However, it should be noted that the average tear strength of the membranes in general is slightly higher after aging than before aging.

Moisture resistance number (Z) for PE membranes with a thickness of 0.15 mm is published as between 300 and 600 GPa m^2 s/kg (Gottfredsen & Nielsen, 2006). In the current study, none of the membranes showed moisture resistances below 300 GPa m^2 s/kg.

Visual examination of membranes containing regenerated PE showed that all contained fragments of less than 1 mm and single fragments up to 4 mm. Cracks and holes were found in membranes from several different rolls. For membrane T7482-6, three holes were found in the flat surface of the foil of the membrane and 2 holes were found in the membrane's foldline. The holes were between 10 and 20 mm long and 3 mm wide. The holes in the membrane's foldline were smaller than 3 mm. For membrane T7482-8, no holes were observed in the foldline. By contrast, 15 holes were found in the flat surface of the membrane with dimensions up to 18 mm in length and 12 mm wide.

CONCLUSION

This study has shown no general differences between the performances of membranes solely based on whether they are produced from pure PE, virgin PE, 100% new PE, regenerated PE or a combination of pure and regenerated PE. Importantly water vapor diffusion resistance measurements showed that all membranes examined, both before and after aging, performed satisfactorily as vapor barriers. The PE membranes examined were found to be based on LDPE alone. None of the membranes examined contained or produced acids on aging.

Suppliers of the examined membranes suggest that regenerated PE comprises collected PE, that is sorted, washed, purified and stabilized with additives. The study shows that it is possible to sort, wash and purify collected PE as well as to add additives in the form of stabilizers (preservatives) so that chemical and physical properties for regenerated PE membranes are similar to those of membranes produced from pure PE.

Visual examination of membranes containing regenerated PE showed that they all contain fragments smaller than 1mm and up to 4 mm. For two out of five of the examined types of membranes of regenerated PE, fractures and holes were observed in the membrane between 3 and 20 mm. Based on visual examination, it may be concluded that a higher degree of quality is needed to produce vapor barrier membranes of regenerated PE. Producing membranes containing regenerated PE to be used for vapor barrier without significant defects is clearly a challenge for some manufacturers and is an issue that needs to be addressed.

ACKNOWLEDGMENTS

REFERENCES

Ali, Z. I., Youssef, H. A., Said, H.M. and H. H. Saleh. 2005. *Thermal stability of LDPE, iPP and their blends*. Thermochimica Acta, 438(1-2): 70-75.

Beckhoff, B., Kanngießer, B., Langhoff, N., Wedell, R. and H. Wolff. 2006. *Handbook of Practical XRay Fluorescence Analysis*. Springer. ISBN 3-540-28603-9.

Beilstein F. 1872. Ueber den Nachweis von Chlor, *Brom und Jod in organischen Substanzen. Ber*. Dtsch. Chem. Ges. 5 (2): 620–21.doi:10.1002/cber.18720050209).

Godtfredsen and Nielsen. 2006. *Bygningsmaterialer - grundlæggende egenskaber*. Polyteknisk forlag. Lyngby. Danmark.

Nicholson, C. and O'Loughlin E. 1996. *The Use of A-D Strips for Screening Conservation and Exhibit Materials*. American Institute for Conservation. 24th Annual Meeting. June 10-16. Norfolk Virginia. https://cool.conservation-us.org/coolaic/sg/bpg/annual/v15/bp15-11.html.

Rasmussen, T. V., Hansen, T. K., Nielsen, J. K., Steenstrup, F. R., Ottosen L., M., Petersen, L. G., Hansen, M. H., Shashoua, Y. 2020. Material Properties (In Danish: Materialeegenskaber – Test af polyethylenmembraners egenskaber før og efter accelereret ældning). SBi 2020:06. BUILD, Aalborg University.

Rasmussen, T. V., Møller, E. B., Steenstrup, F. R., Nielsen, J. K., Ottosen, L. M., Petersen, L. G., Hansen, M. H. & Shashoua, Y. 2018. PE-membranes in the building envelope, a literature study (In Danish: PE-membraners levetid i byggeriet: et litteraturstudie). SBi-Report; No. 2018:11. Aalborg Universitet.

Shashoua, Y.R. 2008. *Conservation of Plastics*. Oxford: Butterworth-Heinemann.