Incorporation of postconsumer polyurethane foam into a polymer/clay aerogel matrix

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ABSTRACT: The feasibility of incorporating ground recycled polyurethane (PU) foam into clay/polymer aerogels was demonstrated, and a range of compositions were prepared and characterized to determine the effect of variation in the formulations on density and mechanical properties of the resulting materials. This study followed a modified combinatorial approach. Initially, experiments were performed in water using either sodium exchanged montmorillonite or laponite clay, poly(vinyl alcohol) (PVOH) solution as the polymer binder, and the recycled PU foam. Freezing and freeze-drying the aqueous gels produced aerogels, which were characterized through density and mechanical testing, scanning electron microscopy, and thermal gravimetric analysis. The study was expanded by exploring alternative binder chemistries, including the use of an alginate polymer in place of the PVOH or adding a polyisocyanate as a crosslinking agent for PVOH. The effect of recycled PU foam content, clay type and level, and binder type and level on the mechanical properties of the aerogels were determined and will be discussed herein. © 2015 Wiley Periodicals, Inc. J. Appl. Polym. Sci. **2015**, *132*, 42586.

INTRODUCTION

Insulation for appliances, such as refrigerators, is typically manufactured from closed-cell polyurethane (PU) foams, a family of products that are quite effective throughout the life of the appliances. Presently, only certain parts of appliances (primarily metals, refrigeration gasses, and foam blowing agents) are recovered and/or recycled once they have come to the end of their useful lives. The PU foam insulation is currently not one of the reused components; the work described herein is an effort to create a new option for recycling of this large volume material into new low-density hybrid inorganic/organic aerogel products.

Clay/polymer composites have been an area of extensive research over the last decade. These composite materials rely upon exfoliation of layered, smectite clays, providing homogeneous dispersion of filler within a polymeric phase, to generate a network which can transfer physical loads, vitrifies the polymer via hydrogen bonding, and/or modification of polymer crystallization and morphology.[1,2] An alternative method of forming clay/polymer composites takes advantage of the high dispersability of smectite clays in water. The structural features of highly compressible "gel skeletons" produced by vacuum sublimation of frozen thixotropic clay gels in water or benzene were first described by Weiss,[3] Hoffman,[4] and Norrish.[5]

Processing parameters for clay aerogels and the definition of a structure in which clay particles are linked edge-to-face much like a "house of cards" owing to opposite surface and edge charges that exist in clays were reported by Van Olphen.[6] The effects of process parameters, such as clay concentration and freezing rates, upon the size and shape of resultant clay aerogels was investigated by Nakazawa.[7] Schiraldi and coworkers have reported the effects of integrating polymers into these freeze dried clay aerogel systems.[8–16]

Household refrigerator recycling operations take the closed cell foam insulation recovered from discarded refrigerators and grind it into a fine powder under reduced pressure to break open the cells. This reduced pressure milling process allows recovery of the blowing agent contained within the foam cells, thus preventing its uncontrolled release; some of these agents are ozone depleting and/or have high global warming potential. In the present work, ground, recycled PU foam is combined with clay and a polymeric binder, and then freeze-dried into aerogel structures. This approach could allow reuse of the PU insulating materials, with minimal negative environmental impact. Because of the ease of tuning polymer/clay/binder formulations in such aerogels, a wide range of mechanical properties could be expected from the new compositions.

EXPERIMENTAL

Materials

Sodium montmorillonite (MMT; Southern Clay Inc., PGW grade) was used as the clay mineral in preparation of the specific aerogel compositions. Poly(vinyl alcohol) (PVOH; MW 195,000 g/mol, Sigma-Aldrich), ammonium alginate (Fisher Scientific), Dispercoll® U53 (anionic high molecular weight PU dispersion) (Bayer MaterialScience LLC) was used to reinforce the matrix of clay platelets and bind the ground postconsumer PU foam or "fluff" (supplied by JACO Environmental of Dana Point, CA. As supplied, the PU fluff has a density of 184 kg/m³ and is >99% pure with no detectable residual blowing agent). Bayhydur® 302 (1,6-hexamethylene diisocyanate-based polyisocyanate modified to be water dispersible (isocyanate equivalent weight of 243 g/eq. NCO; available from Bayer MaterialScience LLC) was used to crosslink PVOH in some formulations. Water was deionized (DI) using a Barnstead ROpure low-pressure, reverse osmosis system.

Aerogel Preparation

The desired quantities of clay were weighed and combined with 100 mL DI water in a Waring Laboratory model blender (22,000 rpm free range speed) for 1–3 min to ensure homogeneity. The desired amount of ground PU foam was then added to the clay/water gel and once again mixed in the blender for 1 min. Separately, the specific polymer binder for a particular aerogel was dissolved in DI water. The clay/foam gels were then combined with the polymer/water solutions using a hand mixer to minimize the amount of bubbles captured by the viscous gel. The resulting gel suspensions appeared homogeneous, with no evidence of phase separation. The gel suspensions were added to 15 dram polystyrene vials, capped, submersed in an ethanol/solid carbon dioxide freezing bath (-70°C), and then freeze dried using a SP Scientific Virtis AdVantage 2.0 Bench Top Lyophilizer. The shelf temperature and condenser coil in the lyophilizer were set to temperatures of -70°C and -85°C, respectively. After a vacuum of 40 μbar was achieved, the temperature of the shelf was increased to 25°C in order to sublime the water from the frozen gel suspensions. After 72 h, all samples were removed from the freeze drier, and the resultant polymer/clay aerogels were characterized.

For clay-free aerogel compositions, the same procedure was followed, except that only the low sheer mixer was used.

Example Ground Polyurethane Foam Aerogel Preparation

In order to produce a 1% PU foam/5% MMT clay/5% PVOH aerogel composition (percentages are based on the wet composition by weight), 50 mL of deionized water was combined with 5 g of MMT clay in a high-shear blender and mixed for 1–3 min, periodically checking the dispersion to ensure homogeneity. Once homogenous, 1 g of PU foam was added, in several aliquots, mixing until homogeneous. Separately, a 10 wt % PVOH solution in deionized water was prepared. To the aforementioned PU foam/clay solution, 50 mL of the recently created 10 wt % PVOH solution was added. The newly created PU foam/clay/PVOH solution was added to a plastic container and mixed under low-shear conditions. The mixture was blended for an additional 1–3 min to ensure homogeneity.

Preparation of PVOH Aerogel Crosslinked with Bayhydur® 302

As described above, desired quantities of clay were weighed and combined with 100 mL DI water in a Waring Laboratory model blender (22,000 rpm free range speed) for 1–3 min to ensure homogeneity. The desired amount of ground PU foam was then added to the clay/water gel and once again mixed in the blender for 1 min. Separately, 10 g of PVOH (227 meq -OH) were dissolved in 100 mL of water. To the 10 wt % PVOH solution, 2.761 g (11.4 meq NCO) of Bayhydur® 302 was added, along with three drops of dibutyltin dilurate as a catalyst. Immediately upon addition of Bayhydur® 302 and the catalyst, the solution became cloudy-white and increased in viscosity. The clay/foam gels were then combined with the crosslinked PVOH suspension using a hand mixer to minimize the amount of bubbles captured by the viscous gel. The resulting gel was frozen and subsequently lyophilized in the aforementioned manner.

Aerogel Mechanical Characterization

Once freeze dried, the resultant cylindrical aerogels (approximately 20 mm in diameter and 45 mm in height) were tested using an MTS model 2525/ReNew upgrade package 5565 mechanical tester equipped with a 1000 N load cell. As is standard with mechanical testing carried out on aerogels, their properties under compression were tested. Bulk densities of the aerogels were obtained through careful dimensional measurement (height and diameter) using digital calipers along with determination of the mass. In order to prepare the aerogels for compression testing, each monolith was cut to a height of 20 mm using a band saw. The ultimate strain for the tester was set to a value of 0.75, i.e., 75% of the total linear distance. Five samples were tested for each composition.

RESULTS AND DISCUSSION

A series of aerogels containing different levels and ratios of ground recycled PU foam/montmorillonite clay/poly(vinyl alcohol) (MMT/PVOH) were produced and characterized. The compositions, densities, and compressive properties of each formulation are given in Table I.

As is evident from Table I, hybrid inorganic/organic aerogels having a range of densities (0.058–0.19 g/cm³) and compressive moduli (0.35–21.9 MPa) can be achieved over the design space evaluated in this work.

The first trend that can be noted here is that the moduli for each group correlated with the concentration of PVOH. As seen in both Figures 1 and 2, the 10% PVOH loading possessed a significantly higher modulus than the two lower levels of PVOH binder content (7.5% and 5.0%). Next, it was observed that the aerogel which possessed the highest modulus was 1% foam/5% MMT clay/10% PVOH (21.9 MPa). This is not surprising as it contained the highest loading of clay and polymer, and the smallest loading of PU foam. In this system, the polymer has an affinity for the MMT clay platelets due to electrostatic attraction. The PU foam, in contrast, would not be expected to interact similarly with the PVOH; the foam appears to act as a diluent, negatively impacting those aerogels produced with high

Table I. Sample Compositions, Densities, Compressive Moduli and Specific Compressive Moduli ($n = 5$) for Aerogels Comprising PU Foam/MMT Clay/PVOH

PU foam	MMT	PVOH	Average density (g/cm^3)	Average modulus (MPa)	Average specific modulus (MPa*cm^3*g^{-1})
2%	2.50%	5%	0.0579	0.35 ±0.07	6.0±1.3
3%	2.50%	5%	0.0747	0.92 ±0.19	12.3 ±2.6
4%	2.50%	5%	0.0679	0.42 ±0.08	6.2 ±1.2
5%	2.50%	5%	0.105	1.99 ±0.51	18.9 ±4.8
1%	2.50%	7.50%	0.0831	1.57 ±0.56	18.9 ±6.8
2%	2.50%	7.50%	0.0795	1.90 ±0.41	23.9 ±5.1
3%	2.50%	7.50%	0.0818	1.98 ±0.28	24.2 ±3.5
4%	2.50%	7.50%	0.0939	2.01 ±0.72	21.4 ±7.6
5%	2.50%	7.50%	0.104	2.34 ±0.90	22.4 ±8.6
1%	2.50%	10%	0.128	5.71 ±1.33	44.6±10.4
2%	2.50%	10%	0.113	5.22 ±2.00	46.2 ±17.7
3%	2.50%	10%	0.112	4.25±1.27	38.0±11.3
4%	2.50%	10%	0.129	4.17±1.02	32.2 ±7.9
5%	2.50%	10%	0.098	4.21 ±0.80	42.7 ±8.1
1%	5%	5%	0.0757	1.00 ±0.33	13.1 ±4.3
2%	5%	5%	0.104	4.76±1.12	45.9±10.8
3%	5%	5%	0.108	3.57 ±0.65	32.9 ±6.0
4%	5%	5%	0.113	2.46 ±0.74	21.8 ±6.5
5%	5%	5%	0.127	2.93 ±0.47	23.0 ±3.7
1%	5%	7.50%	0.116	2.96 ±0.53	25.5 ±4.6
2%	5%	7.50%	0.124	4.07±1.00	32.9 ±8.0
3%	5%	7.50%	0.129	7.89±1.12	61.1 ±8.7
4%	5%	7.50%	0.151	4.89±1.12	32.5 ±7.5
5%	5%	7.50%	0.154	7.96±1.62	51.7±10.5
1%	5%	10%	0.152	21.9 ±5.5	143 ±36
2%	5%	10%	0.174	16.1 ±5.7	92.5 ±32.9
3%	5%	10%	0.148	13.5 ±5.1	91.0 ±34.1
4%	5%	10%	0.186	16.1 ±3.1	86.6±16.7
5%	5%	10%	0.194	14.0 ±3.9	72.2 ±20.3
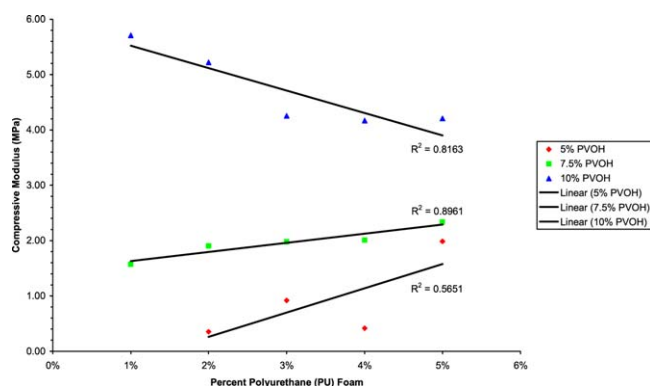
Figure 1. Average compressive moduli for the aerogels containing 2.5% MMT clay as a function of PU foam and PVOH binder level. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary. com.]
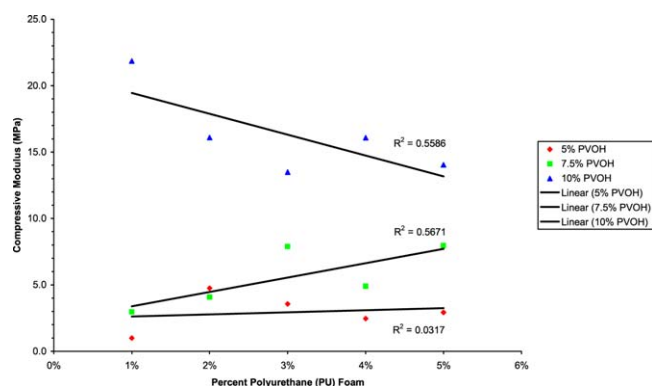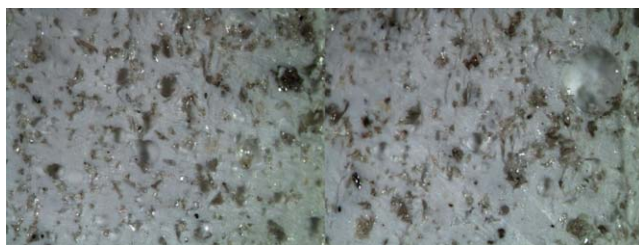

Figure 2. Average compressive moduli for the aerogels containing 5% MMT clay as a function of PU foam and PVOH binder level. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary. com.]

Figure 3. 12P488 optical microscopy images of 5% foam/5% MMT clay/10% PVOH aerogels at 64× magnification. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

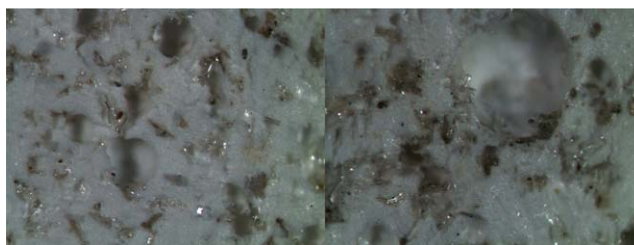**Figure 4.** 12P488 optical microscopy images of 5% foam/5% MMT clay/10% PVOH at 120× magnification. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

concentrations of solids, and behaving as a benign additive in lower polymer concentration aerogels.

Observations based on Figures 3 and 4 indicated that the PU foam (seen as dark regions) was not homogenously distributed throughout the aerogel matrix. The PU foam (as a nonreinforcing filler) forms aggregates which do not interact strongly with the matrix and ultimately reduce the compressive modulus of the aerogels.

Scanning electron micrographs of an aerogel containing 5% foam/5% MMT clay/10% PVOH are shown in Figures 5–7. The images clearly display the locations of foam particles that were trapped in the clay/polymer matrix. It is believed that the areas where it appears there is a void in the sample indicates the location of a particle of the PU foam. The inability to see the foam particles in the SEM images suggest that the PU foam particles were likely removed during cutting and preparation of the samples for microscopy. This may be a result of poor adhesion between the PU foam particles and the matrix.

Exploration of alternative binder chemistries proved more fruitful. Alginate, also referred to as alginic acid, is an anionic polysaccharide found in the cell walls of brown algae. It is extremely hydrophilic, capable of absorbing 200–300 times its dry weight in water.[8] In its dry form, alginate is a yellowish-orange granular solid, with a sweet smelling aroma. Chemically, its structure is a linear block copolymer of β-D-mannuronate and α-L-guluronate. Commercially, alginate is extracted from three types of seaweed: the giant kelp *Macrocystis pyrifera*, *Ascophyllum nodosum*, and various types of Laminaria. Alginate is also produced by two types of bacteria: Pseudomonas and Azotobacter. When the dry powder is mixed with water, it forms a homogenous gel and therefore finds use as a thickening agent in drinks, ice cream and cosmetics; alginate is self-extinguishing in the presence of a flame, and thus finds use in applications requiring fire-proof materials.

Alginate was used as a binder to produce a series of PU foam containing/clay/alginate and neat PU foam/alginate bound aerogels. Compositions, densities and compressive properties of the resulting samples are shown in Table II, Figures 8 and 9. The series included samples containing PU Foam ranging from 1% to 5%, at a constant level of 5% MMT, as well as an analogous set of samples with 1–5% PU foam in the absence of any MMT.

As in the PVOH bound systems, samples having the highest polymer binder levels also showed the highest moduli. The highest modulus achieved with alginate as a binder was an impressive 13.1 MPa from a sample containing 5% PU foam/5% MMT clay/10% alginate at a density of 0.164 g/cm³. Interestingly, the foam/MMT clay/alginate systems show an inverse trend in moduli with respect to the previously described foam/MMT clay/PVOH system. The MMT clay appears to play the role of compatibilizer between alginate polymer and PU foam, such that the resultant aerogels possessed higher moduli with
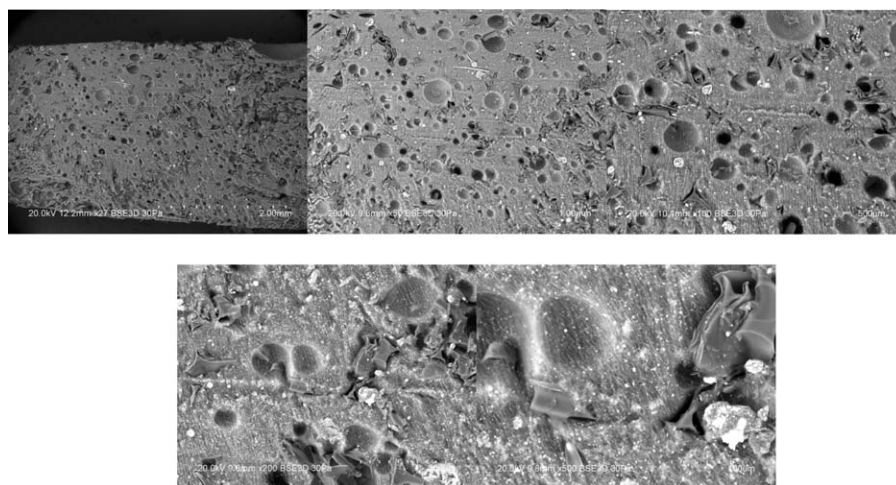

Figure 5. Scanning electron micrographs showing the locations of the foam particles parallel to the direction of cross-sectional slice for the sample containing 5% foam/5% MMT clay/10% PVOH.
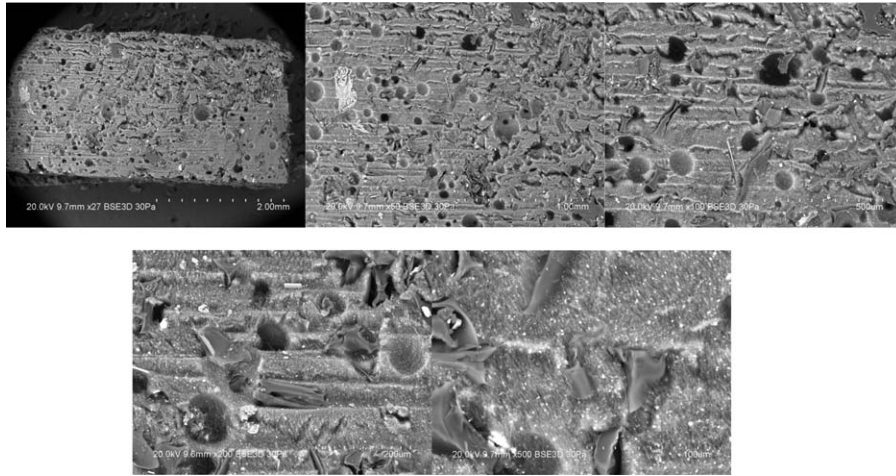
Figure 6. Scanning electron micrographs showing the locations of the foam particles perpendicular to the direction of cross-sectional slice for the sample containing 5% foam/5% MMT clay/10% PVOH.
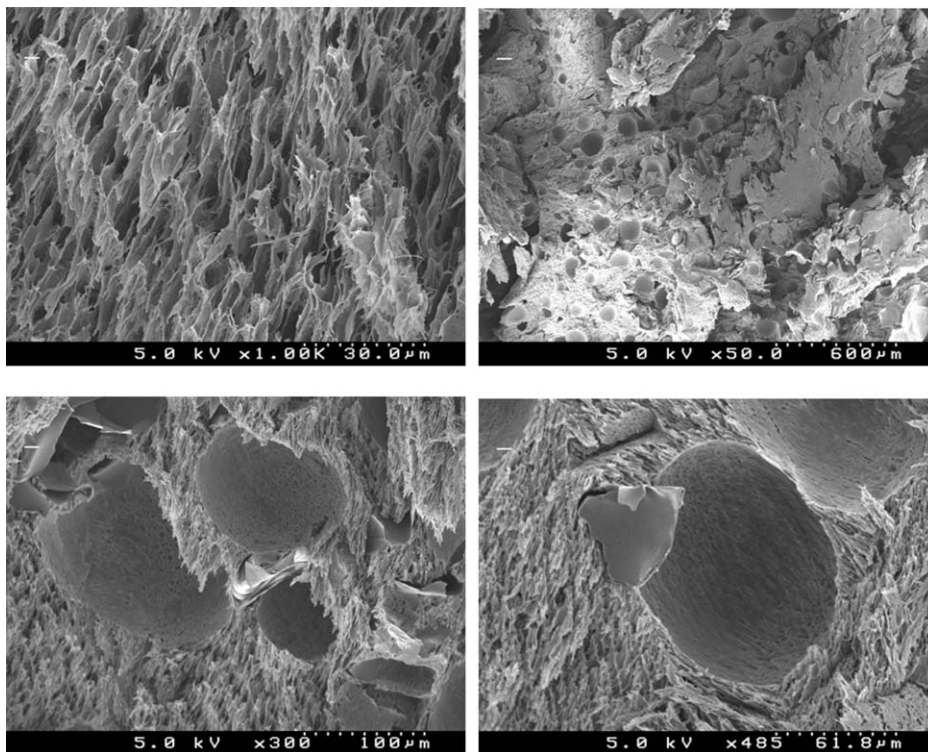

Figure 7. Scanning electron micrographs of the internal structures for the aerogel containing 5% foam/5% MMT clay/10% PVOH. Close examination shows the crevasses left by the foam particles.

Table II. PU Foam/MMT Clay/Alginate Aerogel Compositions and Mechanical Properties ($n = 5$)

| PU foam	MMT	Alginate	Average density (g/cm^3)	Average modulus (Mpa)	Average specific modulus (MPa*cm^{3}*g^{-1})
1%	-	7.50%	0.0772	3.07 ±0.55	39.7 ±7.2
2.5%	-	7.50%	0.0831	2.64 ±0.56	31.8 ±6.8
5%	-	7.50%	0.113	2.21 ±0.38	19.5 ±3.4
1%	-	10%	0.0954	5.24 ±2.21	54.9 ±23.1
2.5%	-	10%	0.119	4.32 ±1.59	36.3 ±13.4
5%	-	10%	0.131	3.69 ±1.23	28.2 ±9.4
1%	5%	7.50%	0.114	5.75 ±1.57	50.5 ±13.7
2.5%	5%	7.50%	0.125	8.77 ±2.57	70.1 ±20.6
5%	5%	7.50%	0.141	10.1 ±1.9	71.5 ±13.5
1%	5%	10%	0.132	9.51 ±0.87	72.0 ±6.6
2.5%	5%	10%	0.143	12.5 ±2.6	87.6 ±18.5
5%	5%	10%	0.164	13.1 ±3.2	80.1 ±19.5
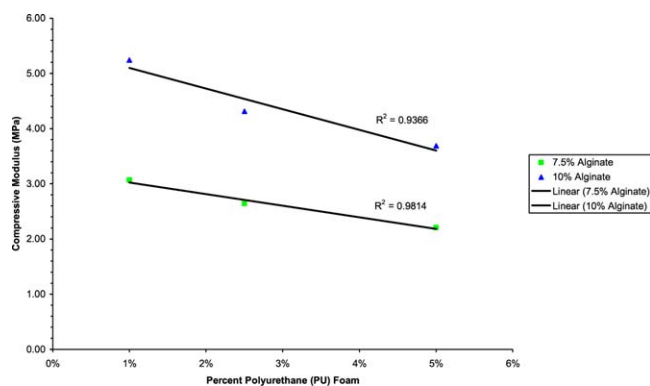
Figure 8. Average compressive moduli for the aerogels as a function of PU foam and alginate binder level (no MMT clay). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Figure 9. Average compressive moduli for the aerogels as a function of PU foam and alginate binder level (5% MMT clay). [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

increased loading of PU foam. In the absence of the MMT clay, these samples showed the same trend as the PVOH bound samples, with increasing levels of PU foam resulting in decreasing modulus. The ability of clay to serve as a synergist in systems with limited aerogel properties/limited water compatibility has been demonstrated before.[14]

A series of six PU foam/MMT clay/Dispercoll® U53 aerogels were produced and tested. The compositions, densities and compressive properties of these aerogels are given in Table III and Figure 10. This was the first group in a series of PU binders that we evaluated in order to determine if a PU binder could give increased modulus in the aerogels by helping to compatibilize the PU foam with the aerogel matrix.

Although these PU systems were able to produce aerogels, the moduli achieved were significantly lower than what we observed with the PVOH or alginate binders at similar clay and PU foam loadings. Presumably, this is due to the lower modulus of the more flexible PU binder versus the alginate or PVOH system. However, it should be noted that as expected, interaction between the PU foam and PU binder did result in a higher compressive modulus. This is exemplified by the approximately 3.4-fold and 5-fold modulus increase upon increasing the PU foam level from 1% to 5% in the 7.5% and 10% PU binder containing systems, respectively (see Table III and Figure 10). This is significantly higher than what is observed using the alginate or PVOH binders, where increasing PU foam over the
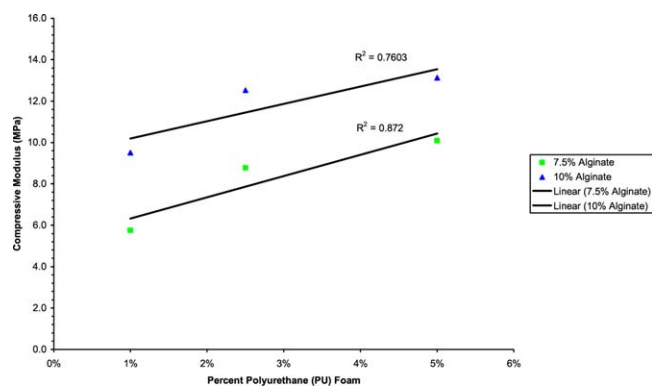
same range results either in a decreased, or in the best case, a more modest increase in modulus.

In order to explore the effect of crosslinking of the PVOH binder on the mechanical properties of the resulting aerogels, a series of 6 PU foam/MMT clay/crosslinked PVOH-aerogels were produced and tested. The compositions, densities and compressive properties of these samples are given in Table IV and Figure 11. The crosslinking of PVOH was performed with Bayhydur® 302 a 1,6-hexamethylene diisocyanate based polyisocyanate modified to be water dispersible. Since this particular polyisocyanate was water dispersible, we attempted the crosslinking reaction of hydroxyl groups on the PVOH in water. We adjusted the stoichiometry to provide 1 eq. of NCO for approximately every 20 eq. of OH side groups on the PVOH. In order for this reaction to occur effectively at room temperature, three drops of dibutyltin dilurate catalyst, which is known to accelerate the reaction of the OH groups with the NCO groups, were added. Immediately following the addition of the catalyst, the solution became cloudy-white, indicative of a chemical reaction occurring.

Compression testing shows that the crosslinking reaction using Bayhydur® 302 did in fact lead to increased modulus. In samples produced with MMT clay and 10% PVOH binder alone, increasing levels of PU foam gave decreasing modulus (lines 10–14, Table I), with the highest value (5.71 MPa) seen at 1% PU foam. In striking contrast, the crosslinked samples showed

Table III. Sample compositions, Densities, Compressive Moduli, and Specific Compression Moduli ($n = 5$) for Aerogels Featuring PU Foam/MMT clay/Dispercoll® U53

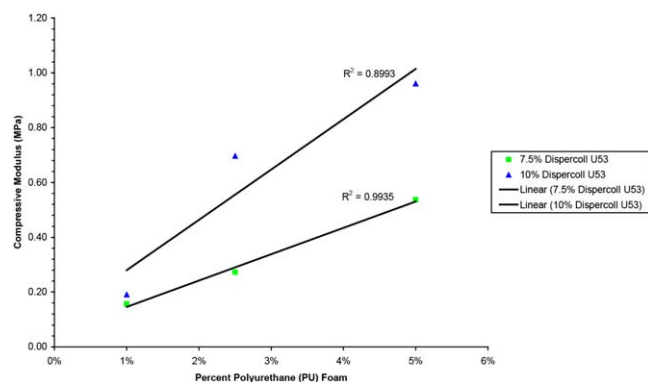| PU foam	MMT	Dispercoll® U53	Average density (g/cm^3)	Average modulus (MPa)	Average specific modulus (MPa*cm^3*g^{-1})
1%	5%	7.50%	0.137	0.16 ±0.02	1.15 ±0.14
2.50%	5%	7.50%	0.138	0.27 ±0.10	1.98 ±0.75
5%	5%	7.50%	0.144	0.54 ±0.02	3.73 ±0.16
1%	5%	10%	0.148	0.19 ±0.06	1.29 ±0.41
2.50%	5%	10%	0.171	0.70 ±0.18	4.07 ±1.05
5%	5%	10%	0.178	0.96 ±0.24	5.40 ±1.34

Figure 10. Average compressive moduli for the aerogels containing 5% MMT clay as a function of PU foam and Dispercoll® U53 level. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary. com.]
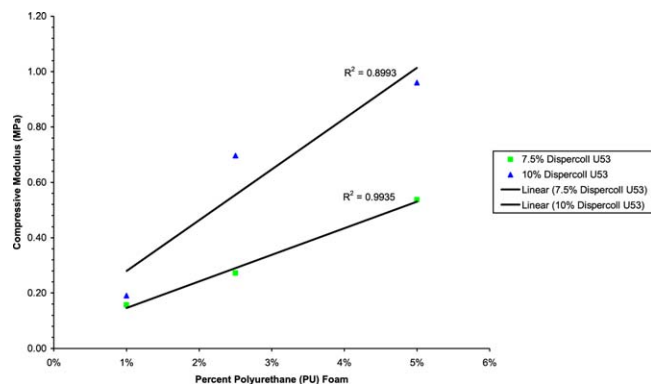

Figure 11. Average compressive moduli for the aerogels containing 2.5% MMT clay as a function of PU foam and binder (PVOH crosslinked with Bayhydur® 302) levels. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

increasing modulus with increasing levels of PU foam. In the crosslinked sample a modulus of 6.77 MPa was observed for the sample containing 5% foam/2.5% MMT clay/10%PVOH, compared with 4.21 MPa for the analogous sample prepared without any added crosslinking agent. Although it is apparent that this method of crosslinking was successful, it is also likely that the process could be improved upon. Since this reaction occurs in water, a side reaction between the isocyanate and water may have produced a primary amine and CO_2. Further reaction between the nascent primary amine and another polyisocyanate molecule would produce a urea linkage, effectively decreasing the crosslink density that could be achieved in the absence of this side reaction with water.

The results of thermogravimetric analysis (TGA), Figure 12, for the 5% PU foam/5% MMT clay/10% PVOH show that the sample contained 2.75% water. This value is confirmed by the initial downward slope (blue curve labeled "water evaporation") on the TGA curve corresponding to approximately 100°C. The onset of degradation of the PVOH occurred at 241.4°C, which is within the margin of error for the literature value. Also, 50.9% of the sample was found to be PVOH. Degradation of the PU foam is likely to be responsible for the continued mass loss after the degradation of the PVOH was completed. At the maximum temperature of the test, we continued to see slow mass loss, presumably due to slow degradation of the PU foam.

CONCLUSIONS

The incorporation of PU foam into clay/polymer aerogels was successfully demonstrated and the resulting low-density hybrid organic/inorganic aerogel compositions were characterized. The compressive moduli of these systems were strongly influenced by the choice and level of the clay, the binder, and the level of PU foam incorporated.

The highest modulus aerogels were achieved with low levels of PU foam and high levels of PVOH binder. In these systems, we believe the PVOH binder has an affinity for the MMT clay platelets through electrostatic attraction, resulting in maximum reinforcing effect on the matrix. In contrast, the PU foam is not expected to interact with the PVOH binder and may disrupt the interaction of the PVOH and clay therefore reducing the compressive modulus of the resultant aerogel.

Alginate/MMT clay systems displayed rather interesting properties, which were in diametric contrast to the mechanical characteristics displayed by the MMT clay PVOH systems. The alginate appeared to interact with the MMT clay and PU foam such that the resultant aerogels possessed higher moduli with increased loading of PU foam. This observation is interesting and particularly useful if the goal is to maximize the recycled PU fluff content, while producing high compressive modulus materials.

Table IV. Sample Compositions, Densities, Compressive Moduli, and Specific Compression Moduli ($n = 5$) for Aerogels Featuring Foam/MMT Clay/PVOH Crosslinked with Bayhydur® 302

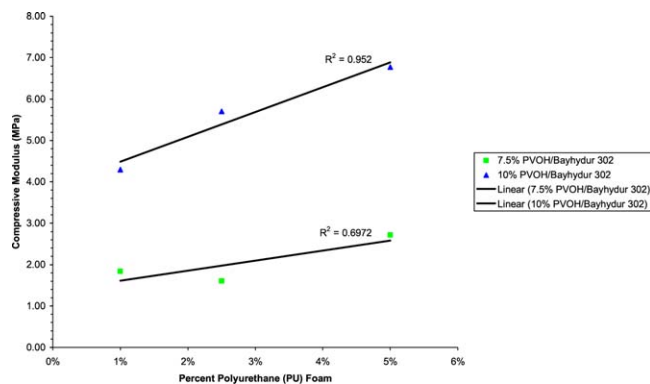| PU foam	MMT	PVOH	Bayhydur® 302	Average density (g/cm³)	Average modulus (MPa)	Average specific modulus (MPa*cm³*g⁻¹)
1%	2.50%	7.50%	2.76%	0.120	1.84 ±0.24	15.3 ±2.0
2.50%	2.50%	7.50%	2.76%	0.156	1.60 ±0.65	10.3 ±4.1
5%	2.50%	7.50%	2.76%	0.157	2.72 ±0.96	17.4 ±6.1
1%	2.50%	10%	2.76%	0.153	4.29 ±1.70	28.0 ±11.1
2.50%	2.50%	10%	2.76%	0.163	5.70 ±1.84	34.9 ±11.3
5%	2.50%	10%	2.76%	0.172	6.77 ±1.67	39.3 ±9.7

Figure 12. Thermogravimetric analysis (TGA) for the decomposition properties of the 5% foam/5% MMT clay/10% PVOH aerogel sample. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

Finally, PU binders were evaluated, in the belief that they would interact with the PU foam in such a way as to produce an aerogel that was reinforced by the PU foam. While these PU binders did form aerogels upon freeze-drying, they produced much lower modulus systems, presumably due to the flexible nature of the PU binder. However, there was evidence that the desired reinforcing effect of the PU foam, which we expected to achieve through increased compatibility of the PU foam and the PU binder, was achieved.

Introducing crosslinking in the PVOH binder via formation of PU linkages by reaction of the PVOH binder with the polyisocyanate, Bayhydur® 302, was effective at increasing the modulus of the aerogels. However, if the crosslinking had taken place after the PVOH aerogel had been freeze-dried, it is likely that the compressive modulus could have been increased even further, by avoiding the nonproductive (in terms of crosslink formation) reaction of the polyisocyanate with water.

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