

# FOAMS WITH HIGH RECYCLED CONTENT ENABLED BY DYNAMIC CROSSLINKING

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## Abstract

Foams produced from crosslinked ethylene vinyl acetate (EVA) provide a balance of processing performance, mechanical properties, and durability in technically demanding applications. Crosslinked EVA foams do not melt, flow, or dissolve to enable the use of conventional reprocessing or recycling methods. As a result, recycling crosslinked EVA waste has long posed a challenge, and most crosslinked polymers and foams accumulate as plastic waste. We recently demonstrated technology that uses dynamic crosslinking to transform crosslinked EVA foam waste into new polymers with thermoplastic behavior. This new, recycled-EVA based polymer can be melt-processed using conventional techniques including foam compression molding. Here, we show that foams containing up to 50% recycled EVA foam waste can be produced by our dynamic crosslinking technology. We produced foams with density and hardness in the range for footwear by blending the recycled EVA-based polymer with virgin resins and conventional foaming additives in a scalable process. These findings highlight a scalable pathway for integrating recycled foam waste into new products, supporting closed loop circularity.

## Introduction and Current State of Technology

Ethylene–vinyl acetate (EVA) copolymers are widely used in footwear midsoles due to their excellent balance of cushioning, durability, and lightweight properties.<sup>1,2</sup> To achieve these performance characteristics, EVA is typically crosslinked using peroxides, forming a permanent, covalent, three-dimensional network that resists melting and flow.<sup>3</sup> While this structure provides effective mechanical properties, it also creates a significant barrier to recycling because conventional melt-processing techniques cannot be applied.<sup>4</sup> As a result, manufacturing scrap and end-of-life EVA foams accumulate as waste, with limited options such as downcycling or incineration—approaches that are energy-intensive and misaligned with circular economy principles.<sup>5,6</sup>

Current recycling approaches for EVA foams typically involve grinding scrap and blending it into virgin EVA resin as a filler. While this enables partial reuse of waste, recycled content is generally limited to 10–15 wt% to avoid significant deterioration in mechanical properties such as tensile strength, compression set, and abrasion resistance.<sup>7–9</sup> For example, Bianchi et al. reported that EVA foams containing 10 wt% recycled material exhibited lower performance compared to virgin EVA, despite environmental benefits.<sup>7</sup> These limitations underscore the need for innovative strategies that enable high recycled content without compromising performance, motivating the dynamic crosslinking approach explored in this work.

Dynamic covalent networks, or vitrimers, have emerged as a promising solution for recycling thermoset polymers.<sup>10,11</sup> Vitrimers incorporate exchangeable bonds that enable topological rearrangements under heat or stress, allowing reprocessing while maintaining network integrity.<sup>12</sup> Previous work demonstrated that crosslinked EVA can be converted into a vitrimer via mechanochemical processing with a transesterification catalyst and hydroxyl source.<sup>13,14</sup> In particular, we demonstrated in lab scale that EVA foams from footwear

scrap can be converted to new polymers. The material produced by this vitrimerization process exhibits four key properties: thermoplastic behavior, elasticity, reprocessability, and foamability.<sup>13-15</sup>

## Objective

The objective of this work is to validate the vitrimerization process for recycling crosslinked EVA foam at a pilot scale and to demonstrate its applicability in producing high-performance footwear midsoles with elevated recycled content. Prior proof-of-concept studies established vitrimerized EVA as melt-processable, elastic, reprocessible, and foamable.<sup>13-15</sup> This study aims to demonstrate that recycled EVA vitrimers can be produced at pilot scale and can be used to make foams with properties appropriate for footwear applications.

## Technology Approach

### (I) Vitrimer production

To better understand the effects of different vitrimer formulations on foamed EVA products, five different vitrimer compositions were prepared to compound with EVA resin. The amounts were chosen based on previous studies.<sup>14,16</sup> Each composition includes scrap recycled material, virgin EVA as a diluent with defined melt flow index (MFI), foaming agent, peroxide, and catalyst system for crosslinking (Table 1).

Due to the processing challenges associated with compounding 100 parts per hundred resin (PHR) of vitrimer precursor directly into a foamable EVA formulation, additional dilution with virgin EVA is necessary. Without this, excessive pressure buildup in the die can lead to clogging of the extruder. Thus, each vitrimer composition was compounded with a commercial virgin EVA resin to reduce the viscosity of the blend. Higher MFI resins (25 and 55) have lower viscosity which results in a lower extrusion die pressure. Conventional foaming and curing agents were added to initiate foaming and crosslinking.

EVA foam scrap with a small amount of virgin EVA resin was added in a 3.8 L kneader mixer and preheated to approximately 74 °C prior to material addition. Due to the low bulk density of the chunks, EVA foam was introduced in three stages to achieve a total charge of 2.5 kg (two additions of 1 kg followed by 0.5 kg), with intermediate mixing to promote densification.

Vitrimerization catalysts were added, and the mixing continued until the mixture reached a target temperature of 125 °C, after which the material was transferred to a TYP-50 co-rotating twin screw extruder for pelletization. For this study, four different formulations of vitrimerization catalysts were studied. A structured experiment was conducted to measure the effects of the relative concentrations of catalyst and cocatalyst (Table 1). While composition 6 has more vitrimer than 2-5, the relative amounts of the catalyst system are reported based on the ratio to vinyl acetate, which remains relatively constant.

Table 1. Foamable vitrimer formulations.

Composition	Vitrimer	25 MFI EVA	55 MFI EVA	EVA Scrap	Catalyst	Cocatalyst
1	-	100	0	0	High	None
2	A	50.0	7.5	42.5	Low	Low
3	B	50.0	7.5	42.5	High	Low
4	C	50.0	7.5	42.5	Low	High
5	D	50.0	7.5	42.5	High	High
6	C (conc.)	32.0	10.2	57.8	Low	High

## (II) Foamable Blends – Extrusion blending of vitrimer with foaming additives

The six compositions were subsequently compounded with 1.6 PHR of peroxide, 1.4 PHR of foaming agent, and virgin EVA. Composition 1 was compounded on an Xplore microcompounder and compositions 2 – 6 were compounded on an 18-mm twin-screw extruder. For all samples, the EVA was dry blended with foaming additives and passed once through the extruder. The extrudate was then water-cooled and pelletized. Due to high viscosity of composition 4, the temperature was increased and screw speed was decreased to keep the pressure below the upper threshold of the extruder (Table 2).

Table 2. Extruder conditions for foam composition compounding.

Composition	Melt Temperature (°C)	RPM
1	108	100
2	111	200
3	105	250
4	118	150
5	107	250
6	117	250

## (III) Foams containing vitrimer – Molding and Characterization

The compositions of EVA with foaming additives were foamed using a Luxor foam press in a 115 mm x 65 mm mold (Table 3). Pellets of each composition were pressed into a 10 mm thick sheet using a Carver press at 110 °C using approximately 5000 lbs of pressure. The sheet was then cut into small slabs and 65-70 g of the slabs were loaded into the preheated mold after coating the mold with Selsil mold release spray.

Table 3. Compression molding conditions for foam production.

Composition	Mold Time (min)	Mold Temperature (°C)	Mold Pressure (tons)
1	10	170	25
2	7:50	165	25
3	7	165	25
4	8	165	25
5	7	165	25
6	5:45	165	25

Foams were characterized using expansion ratio, density, and hardness per the following methods:

#### *Expansion ratio*

Sample molds were pressed with 100 mm imprint (Figure 1). The imprint was measured after expansion to determine the expansion ratio. For example, an imprint after expansion measuring 150 mm corresponds to an expansion ratio of 1.50.



Figure 1. Example of compression molded foam with 100 mm imprint.

#### *Density*

Density of two cutouts of each foam (duplicate) was measured according to ASTM standard D792.

#### *Asker C Hardness*

Foamed samples were tested directly for hardness according to ASTM standard D2240.

### **Results and Discussion**

The hardness, expansion ratio, and density of each foam are reported in Table 4. These tests all provide critical information for application in footwear midsoles. Asker C hardness was chosen over other hardness tests due to its relevance for soft materials, like foams.<sup>17,18</sup>

Table 4. Characterization of foams.

Composition	Vitrimer	Total Recycled Content (PHR)	Vinyl Acetate Content (PHR)	Expansion Ratio	Hardness (Asker C)	Density (g/cc)
1	-	0.0	28	1.30 ± 0.01	61.6 ± 1.2	0.372 ± 0.004
2	A	42.5	26.4	1.50 ± 0.01	45.0 ± 1.2	0.224 ± 0.003
3	B	42.5	26.4	1.51 ± 0.01	43.6 ± 1.1	0.230 ± 0.004
4	C	42.5	26.4	1.46 ± 0.01	49.7 ± 1.2	0.256 ± 0.005
5	D	42.5	26.4	1.48 ± 0.01	46.2 ± 1.3	0.286 ± 0.001
6	C (conc.)	57.8	25.8	1.42 ± 0.03	50.7 ± 3.3	0.311 ± 0.004

The target expansion ratio was approximately 1.5, and this was achieved with all samples. This indicates substantial volume increase while maintaining expected foam geometry and surface characteristics. This level of expansion reflects effective gas retention and curing, which are critical for lightweight applications.<sup>14,16,19</sup> An Asker C hardness of 40–50 for vitrimerized EVA foam represents a successful balance between softness and structural integrity.<sup>20</sup> The hardness results also indicate that changes in hardness are not highly dependent on VA content, but more so with vitrimer content.

With the exception of Composition 6, which contains more vitrimer, these vitrimer-based EVA foams hold densities below 0.30 g/cc, representing a lightweight material. As such, the target density range for footwear foam was met. In this, we also discovered that a higher expansion ratio resulted in a lower density, which was achieved with a lower amount of cocatalyst. Low density is preferred for both footwear performance and for transportation and handling, and enhances thermal insulation and energy absorption.<sup>21,22</sup>

Inverse correlations were observed between density, hardness, and expansion ratio, which are consistent with expectations for these foams (Figure 2). As more gas is trapped inside the foam, this softens the material and decreases density. These trends were found to be linear, which provides a useful design lever for tailoring properties in foam design.

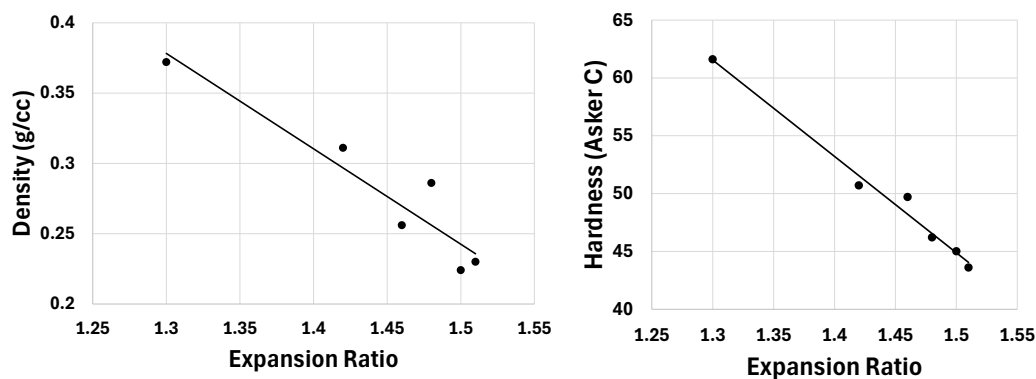


Figure 2. Density correlation with expansion ratio (left) and hardness (right). Both hardness and density decrease with increasing expansion ratio.

One critical finding in foams produced from EVA scrap was that small particles of rubber scrap from recycled content in the foam formulations generated defects in the foams (Figure 3). It appears that the rubber was not chemically modified, dispersed, or melted during the vitrimerization process; it leaves behind a contaminant that causes a small hole. Rubber scrap from recycled material was found to impede uniform foaming

expansion due to limited penetration of additives into the scrap. When foaming was activated in the press throughout the material, the rubber scrap acted as a localized barrier, blocking expansion and leaving small voids adjacent to the scrap particles. This was confirmed by the rubber scrap visibly present in the center of each void, and submersion of the isolated scrap in water resulted in sinking (density >1 g/cc), consistent with rubber and not EVA. This effect is presumed to artificially decrease the bulk density slightly.

In principle, rubber contamination may be eliminated early in the process by using machinery dedicated to EVA. It is also theoretically possible to sort the rubber particles from the scrap during the recycling process by submersion in water. The rubber will sink while the EVA will float, which then can be collected.

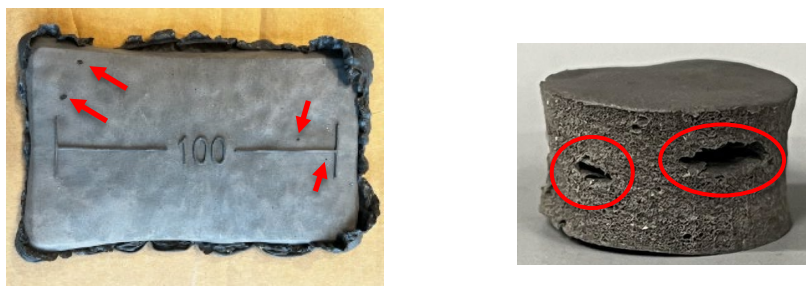


Figure 3. Holes present from rubber scrap blocking the foaming process. Size of holes in context of overall foam (left) and holes visible in a cut circular specimen (right).

## Conclusions and Recommendations

Foams produced from virgin EVA blended with vitrimer content and incorporating >50 PHR recycled material were successfully extruded and compression molded. Characterization revealed a property profile well-suited for footwear midsoles, with opportunities to refine the feedstock regarding the presence of rubber scrap. Importantly, the approach outlined here offers a promising route toward closed-loop recycling in footwear manufacturing.

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