LAMINATED CERAMIC POLYMER COMPOSITES FOR VENTILATED FAÇADE APPLICATIONS

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ABSTRACT

This paper reports on the viability of polymer - ceramic laminations to improve the integrity of ceramic tiles in the case of rupture or breakage. The study focuses on the suitability and advantages of using different thermoplastic film formulations heat laminated to ceramics and natural stone, demonstrating excellent adhesion between the films and the ceramic. Mechanical performance in terms of impact resistance and tile integrity after breakage is studied as well. Furthermore the durability of such systems when exposed to environmental conditions like humidity, water exposure at various pH values, and UV radiation is investigated. The ceramic - polymer composite integrity upon breakage has been also tested on industrial scale tiles to confirm scalability of the technology. Finally we discuss the influence of the lamination process variables which should aid the process design and industrial implementation of the technology.

The results obtained so far within our extensive development program demonstrate that the thermoplastic polymers evaluated perform extremely well in terms of adhesion to ceramics and natural stone and can be used to improve significantly the integrity of the tiles. The improvements in mechanical performance of the laminates versus the unmodified ceramic should allow the potential to reduce the thickness of the tile used in these types of application. The key advantages of using thermoplastic polymer films over wet-chemistry based polymer systems like epoxy are the greater ease of handling and the time efficiency of the heat lamination process.

Finally, the concept of ceramic-polymer lamination can be used to generate or design multilayer systems with optimized performance for new applications. A patent is pending to protect these technology developments.

1. INTRODUCTION AND VALUE PROPOSITION

The use of ceramic tiles in ventilated façade systems [1] has experienced significant growth in recent years. Such systems exhibit excellent heat and acoustic insulation which align with today's trend towards sustainable construction methods. Despite these apparent advantages, given that the tiles are not directly adhered to the building walls, but suspended using mechanical clips, doubts about the system security due to the possibility of falling fragments in the case of fracture or breakage exist. To reduce this risk a suitable tile thickness has to be used and in some cases reinforcement with in-situ reactively cured epoxy and a polyester mesh is used. This in turn brings the added complications of system complexity, industrial hygiene, curing time and ceramic pre-drying amongst others.

2. CERAMIC – POLYMER LAMINATION AND ADHESION

Porcelain stoneware ceramic and natural stone tiles were heat laminated on a laboratory scale with various designed thermoplastic film formulations with a typical film thickness of 500 microns. The ceramic tiles were prepared by the ITC and their composition is mainly based on silicon oxide (67%) and aluminum oxide (20%) with an internal porosity of 10% and a thickness of 8 and 6mm without ribs. The composition of the commercial granite samples is similar at silicon oxide (72%), aluminum oxide (14%) and potassium oxide (4%). The commercial marble samples are on the other hand basically composed of calcium carbonate with small amounts of magnesium carbonate.

The heat lamination of the thermoplastic films to the various tiles was carried out by compression on a Collin hot press, model P300P. The ceramic tile and the thermoplastic film were placed over a steel frame with the desired final thickness as shown in Figure 1, avoiding direct exposure of the tile to extensive pressure resulting in breakage and securing good contact between tiles and polymer films. Lamination conditions were 180ºC and 1bar during 300 seconds followed by 600sec of cooling the press to 50ºC.

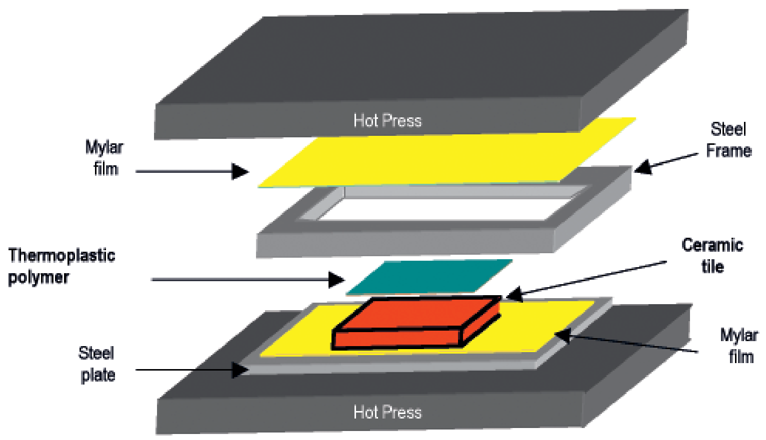

Figure 1. Scheme of laboratory ceramic tile heat lamination

Adhesion between ceramic tiles and thermoplastic film samples was measured using a modified 180º peel test [2] (Specimens had a bonded length of 60 mm and a free end of 80 mm). The selected film samples were made of LDPE as reference, of ethyl-acrylic acid (EAA) copolymer and maleic-anhydride (MAH) grafted polyethylene, as shown in Chart 1. The data confirms that excellent adhesion can be achieved via heat lamination between ceramic and functional polyethylenes.

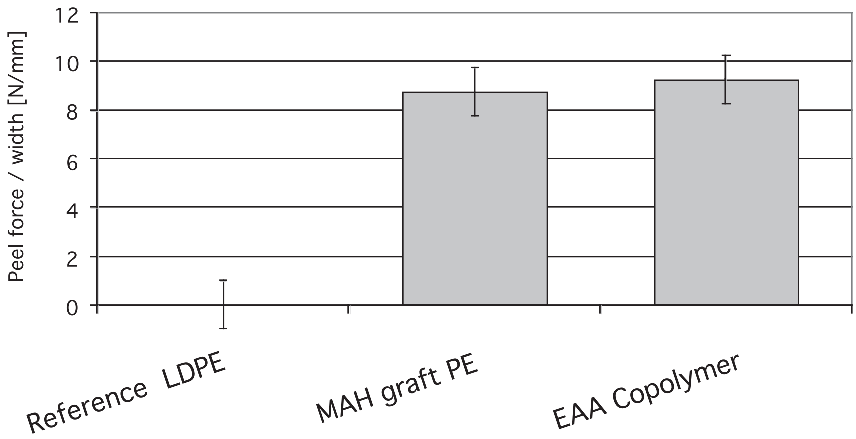

Chart 1. Adhesion of functional polymer film to ceramic tile in 180º peel test

3. INTEGRITY OF CERAMIC – POLYMER COMPOSITE

In case the ceramic tile breaks its integrity is a key parameter in its safety performance. Integrity here is defined as the individual fracture pieces of the tile not falling apart but being kept in place. In case of unmodified tiles there is little to prevent this happening except preventing tile breakage itself by increasing the tiles thickness. The lamination with polymer film offers a solution by keeping the fractured pieces in place. The performance enhancement provided by the thermoplastic film is thus based on the adhesion between polymer and ceramic and the rigidity and strength of the film to keep the fracture pieces in place.

To test for integrity, various sample tiles (14x14cm) laminated according to the procedure in section 2 were impact fractured by placing tiles horizontally on a testing support (Figure 2). A hardened steel ball of 46g was dropped from 63.5cm onto the tile following a testing norm for safety glass, ISO 3537 [3].

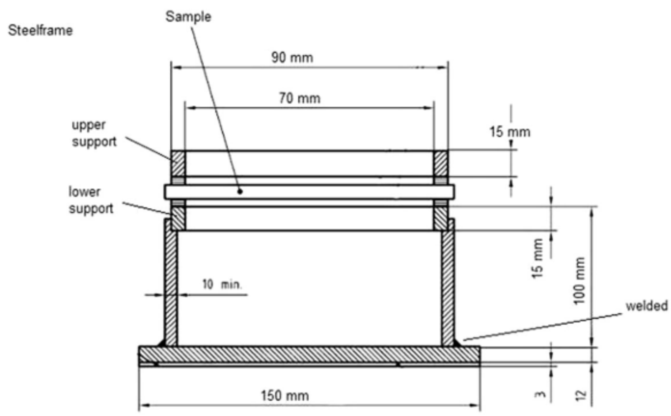

Figure 2. Supporting frame for ball drop test

The fractured tiles were judged for integrity based on adhesion and the overall stability/rigidity of the fractured composite by comparison. A subjective qualitative scale rating of 0 = poor to 3 = excellent was used for comparison of the two parameters and as well the overall performance. The samples tested were unmodified tiles, tile composites with a epoxy-AB system and composites with various functional polyethylenes, i.e. EAA polyethylene copolymer, MAH grafted polyethylene, and proprietary film formulations thereof. The results of the tests are shown in Table 1. It is seen that the thermoplastic film formulations perform as well as the more complex and difficult to apply epoxy system and that the integrity of the broken tile is significantly improved over the unmodified tiles. The thermoplastic film can be applied in a few minutes and the composite is ready for use whereas the epoxy system requires a significantly longer curing times.

Material	Rigidity (film)	Adhesion (film)	Integrity (tile + film)
EAA Copolymer 1 / tile	3	3	3
EAA Copolymer 2 / tile	3	3	3
MAH grafted PE / tile	3	3	3
Epoxy resin 1 / tile	3	3	3
Epoxy resin 2 / tile	3	3	3
Tile without polymer	0	0	0

Table 1. Integrity analysis of tile polymer composites

In the final application ventilated façades in general and the proposed polymer composite in particular are exposed to various adverse environmental influences over time, amongst the most important being rain (water) and UV radiation. The integrity of the composites upon exposure to these influences over time is described in the following..

4. INTEGRITY AFTER WATER IMMERSION AT VARIOUS PH

Composite substrates prepared as described before were objected to water immersion tests at various pH values, at 2, 7, and 10, respectively to simulate rainy conditions on exterior walls of buildings. The pH 7 solution was obtained by adding acetic acid and sodium acetate to deionized water. The pH was controlled with a pH meter. The other two pH solutions (pH 2 and 10) were purchased from Merck KgaA and used without further purification. After the predefined immersion times, the samples were let to dry at 25ºC for 1h, and tested for adhesion and integrity following the previously described procedure. As before, thermoplastic films containing EAA copolymer and MAH grafted PE were evaluated. Exposure times of the composites in the pH controlled water bath were 1h, 2h, 3h 4h, 6h, 8h, 10h, 14h and at maximum 24h.

Table 2 reports adhesion and integrity data as a function of time. Integrity was judged as before after breaking the tile in the ball drop test described earlier. Both films perform very well in the course of this demanding test for the adhesive bond with the MAH grafted polyethylene film showing slightly better results.

Additional data generated after immersion in water at pH7 for 24 hours and shown in Table 3 confirm the good performance of the developed thermoplastic films. The difference between two EAA Copolymer polyethylene films seen in Table 3 underlines the importance of the particular structure of the extruded film on the final performance and the implicated optimization potential.

The data demonstrate that water immersion does not negatively influence the performance of the composite system and that the system therefore should withstand exposure to rain on the external walls of buildings.

	MAH-grafted PE				EAA Copolymer film			
	pH = 2		pH = 10		pH = 2		pH = 10	
Time (h)	Adhe-sion	Integrity	Adhe-sion	Integrity	Adhe-sion	Integrity	Adhe-sion	Integrity
0	3	3	3	3	3	2	3	2
2	3	3	3	3	3	3	3	3
6	3	3	3	3	3	3	3	3
14	3	3	3	2	3	2	3	2
24	3	3	3	3	3	2	3	2

Table 2. Adhesion and integrity of tiles evaluated as a function of water immersion time at two pH values.

Extruded Films	seco t = 0 h		pH = 7	
	Adhesion	Integrity	Adhesion	Integrity
EAA Copolymer PE film 1	3	3	2	2
EAA Copolymer PE film 2	3	3	2	2
MAH grafted PE film	2	3	3	3

Table 3. Adhesion and integrity tests of tile polymer composite before and after immersion in water for 24 hours at pH 7

5. INTEGRITY AFTER UV-RADIATION AND HUMIDITY EXPOSURE

On the external wall of buildings the ceramic tiles are also indirectly exposed to UV radiation and humidity which can negatively affect performance. Advantageous for the application is that the polymer layer is laminated to the backside of the ceramic and thus not directly exposed to the radiation. To mimic this type of exposure, the prepared composites were arranged in an UV weathering chamber (Atlas Ci500 Xenon Weather-Ometer) with the ceramic face directed towards the UV radiation source. The duration of the testing was so far 500hours (and is carrying on). The chamber conditions are set at a radiation wavelength of 240nm, a relative humidity of 60%, a laminate sample temperature of 60ºC and a chamber temperature of 40ºC [1]. To simulate rain conditions, the samples were sprinkled with water following wet and dry cycles with 112 minutes of dry cycle followed by 18 minutes of wet cycle.

The data obtained until now in this ongoing study are summarized in Table 4. As seen, after 500 hours of exposure no performance loss in terms of integrity and adhesion has been recorded.

Film	UV radiation			
	t = 0 hours		t = 500 hours	
	Adhesion	Integrity	Adhesion	Integrity
EAA-copolymer PE	3	3	3	2
MAH-grafted-PE	2	3	3	3

Table 4. Integrity of composites after 500h of UV radiation and humidity exposure.

6. INTEGRITY IMPACT TESTING OF INDUSTRIAL SCALE COMPOSITE

Today no specific normative applicable to ventilated façades exists at National or European level. The functional benefits of these constructive systems are therefore evaluated by means of a European Technical Approval (ETA). In order to determine the impact behavior of ceramic-polymer system, criteria described in the guideline "Kit's for External Wall Claddings. Part I Ventilated cladding elements and associated fixing devices" have been employed. This guideline is currently in the development phase and will be the base for the future European Technical Approval applicable to ventilated façades [1].

Concerning security requirements, the guideline states that constructive elements are to be designed to avoid danger and injuries to people. Such dangers can clearly be caused by the presence of sharp broken edges falling off tiles or loosened fragments. In accordance with the guideline, the evaluation of industrial scale composite tiles is based on the method specified in norm ISO 7892:1988 [4]. The developed test exposes the conveniently placed tiles to impacts with steel balls of 0.5 and 1 kg at increasing levels of impact energy of 1, 3 up to 10 Joules. The tiles are mounted on a metallic substructure which has the same setup as in the final external ventilated façade application.

Commercial porcelain stoneware ceramic tiles with dimensions of 600x600x10mm have been used, with and without lamination. The tiles were fixed to the steel frame using a visible clip fixing. The latter is one of the most common cladding systems used in the industry. In addition, the gaps between the aluminum frame and the ceramic tile were filled with polyurethane foam to avoid mobility and vibration of the pieces. The setup is shown in Figure 3.

The fracture imparted to the tile at 3 Joules with no film laminate protection is shown in Figure 4. As seen, some edges of the broken structure are no longer sustained by the cladding system and easily could fall off at impact energies of equal or larger than 3 Joule.

A direct comparison between these unmodified tiles and the tiles laminated with thermoplastic films is shown in Figure 5 and 6. The results confirm very clearly the superior performance of the composite in terms of integrity and safety after impact up to the maximum energy applied here of 10 Joules. This demonstrates the benefits thermoplastic laminates in ventilated façades applications.

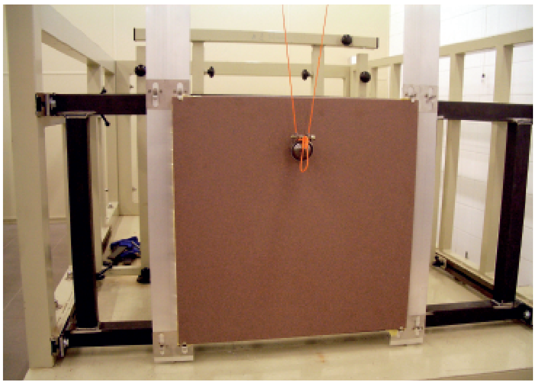

Figure 3. Test scheme

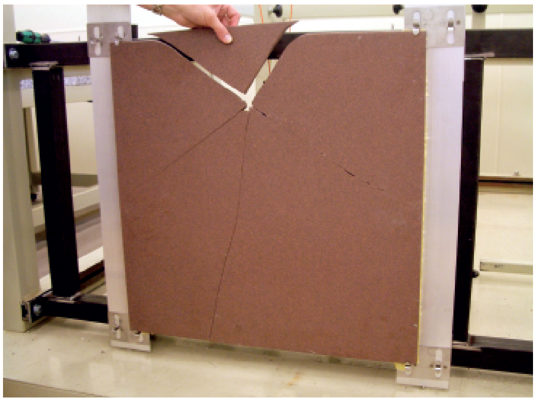

Figure 4. Breakage with loose piece at 3 Joules

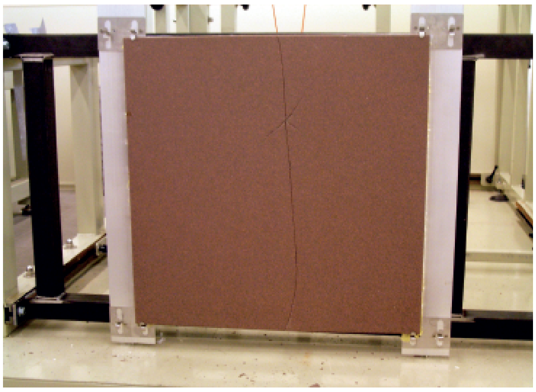

Figure 5. Film laminated Tile broken at 10 Joule

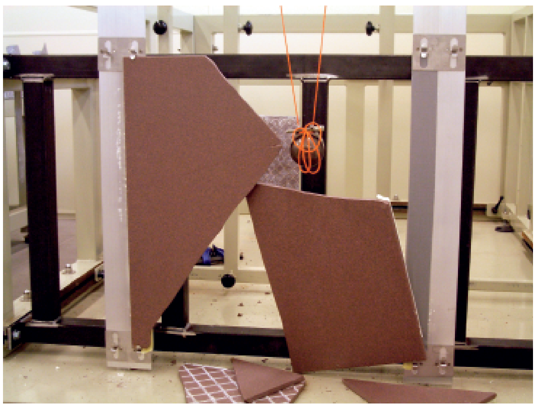

Figure 6. Ceramic tile broken at 10 Joule

System	Impact Energy		
	1 Joule	3 Joules	10 Joules
Ceramic tile	No damage	Breakage with loose-ning	Breakage with multiples loosening
Ceramic tile + film EEA-PE	No damage	Fissure without loose-ning	Fissure without loosening
Ceramic tile + film MAH-gr-PE	No damage	Fissure without loose-ning	Fissure without loosening

Table 5. Impact test of industrial scale tiles with and without polymer lamination

7. DISCUSSION OF INDUSTRIAL LAMINATION PROCESS

Industrial processes of heat lamination are applied commonly in the plastics industry. Each application typically requires a particular process solution due to the variety in substrate material, thickness, flexibility and surface morphology. Here we discuss two relevant examples of existing heat lamination process solutions as guidance. Subsequently some key parameters for the heat lamination of ceramic tiles with thermoplastic films are discussed followed by process design considerations.

Hot roll and belt lamination use heat and pressure as their means of bonding. As shown in Figure 7, the adhesive film and substrate are drawn onto heated rollers where the materials are pressed together. The heat activates the adhesive film, creating a bond when pressed against the substrate material. Hot roll and belt lamination of adhesive films allow for continuous in-line lamination. The proposed films are composed of thermoplastic polymers that allow them to be reheated and molded in subsequent operations. The web substrate to be laminated in these applications is typically flexible but the roller setup feeding the substrate can be easily modified to transport ceramic tiles.

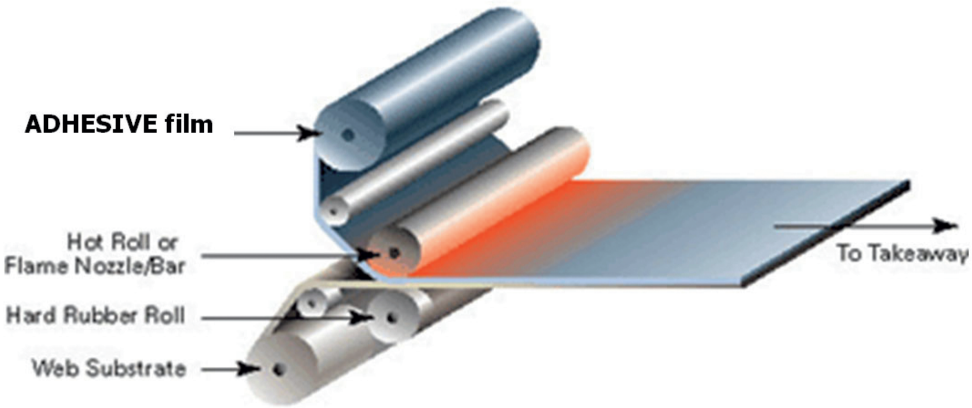

Figure 7. Hot Roll/Belt Lamination

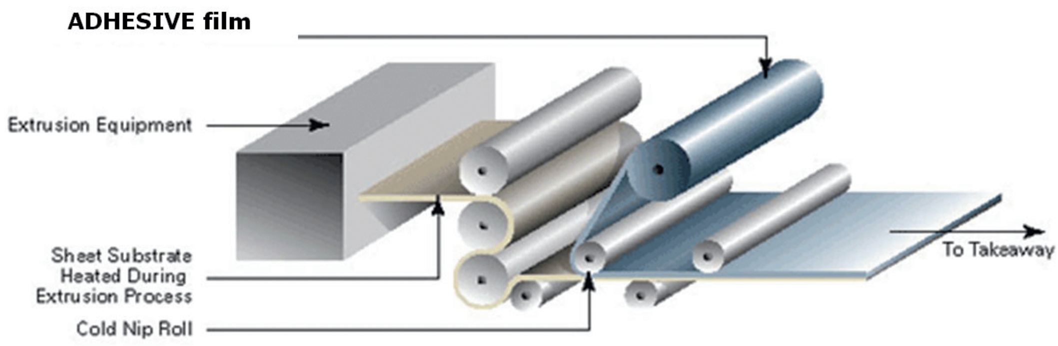

Figure 8: Sheet (or tiles) Extrusion.

In the sheet extrusion heat lamination process a flexible or rigid polymer sheet or profile is extruded as shown in Figure 8. The polymer sheet still carries residual heat from the extrusion process (temperatures > 150ºC), which is used to activate the adhesive film brought into intimate contact with the substrate by pressure rolls (which could also be heated if needed). The strength of the bond depends upon material selection and processing conditions. The extruder can be thought of as the firing oven of the ceramic tile production highlighting one of the key advantages of using the heat lamination technology: the heat of firing the ceramic tiles can be used directly to heat activate the adhesive film. This is should make the implementation of a lamination station into the process of ceramic tile production easily possible.

Based on this discussion about heat lamination processes some recommendations can be made for the heat lamination of ceramic tiles. The key parameters influencing the adhesion in heat lamination of ceramic tiles are temperature of the ceramic tile, hot roll temperature, nip roll pressure and line speed. These parameters were additionally evaluated in initial studies using a bench scale hot roll lamination station (Hot Roll Laminator; ChemInstruments, Fairfield, Ohio).

The thermoplastic polyethylene films discussed in this work generally have a heat activation temperature between 80 and 130ºC, which have to be considered as lower limits for process temperature settings. Good adhesion to substrates is typically obtained when setting the hot role temperature above 170ºC; in our studies 193ºC was used. Additionally, to promote good adhesion the ceramic tiles were heated well above the polymer activation temperature, aiding the heat mediated adhesive bond formation.

The pressure in the nip between role and ceramic tile has typically to be optimized. At low pressures, the wetting of the ceramic surface with the adhesive film is not sufficient resulting in poor bonding. On the other hand, too high pressures can lead to undesired decreases in the film thickness. Therefore, depending on film rigidity and polymer properties the optimal pressure has to be identified in straightforward experiments.

The line speed influences the contact time for the wetting of the tile with the adhesive film under a given nip pressure. Shorter contact times at higher line speeds decrease the time for bond formation which can result in insufficient adhesion. If high line speeds are needed and bonding times are short, increasing temperature and pressure can allow for shorter bonding times until optimal results are achieved.

Another interesting industrial batch lamination process, adopting a vacuum bag principle, can laminate two or more pieces of ceramic through heating under vacuum conditions. This is a process widely used in the architectural glass laminate industry. An illustration of the concept can be seen in Figure 9.

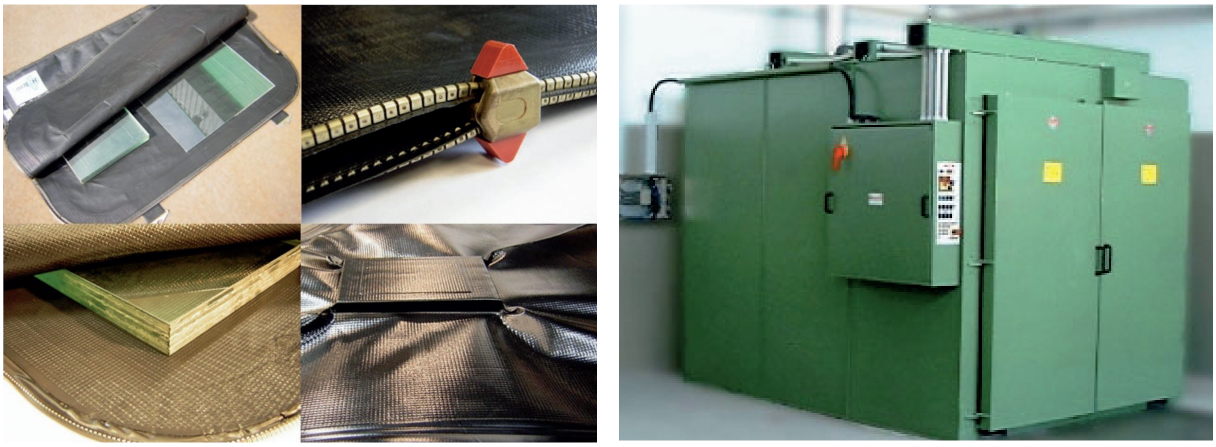

Figure 9. Vacuum lamination for glass sandwiches. Left: vacuum bags. Right: oven [6].

8. CONCLUSIONS

The data generated in this project clearly shows that the integrity and hence security of tiles used in ventilated façade systems can be enhanced when such tiles are laminated to thermoplastic polymers/films. Additionally, other benefits versus solutions used commercially today, such as epoxy or PU resins, potentially exist and are summarized as the key value propositions below:

- Precision thickness control
- Uniform coverage
- Simple, clean and easy handling and storage
- Space- and energy-savings potential
- Minimal adhesive waste
- Potential to reduce system weight

REFERENCES

[1] ETAG 12: 2006 Draft. Guideline for European Organization for Technical Approval of kits for external wall claddings – Part I: Ventilated cladding elements and associated fixing devices.

[2] ASTM D903-98 (2004), Standard test method for peel or stripping strength of adhesive bonds.

[3] ISO 3537: 1999. Road vehicles. Safety glazing materials. Mechanical test.

[4] ISO 7892: 1988. Vertical building elements. Impact resistance test. Impact bodies and general test procedures.

[5] ISO 1872-2: 2007 Plastic – Polyethylene (PE) moulding and extrusion materials – Part 2: Preparation of test specimens and determination of properties.

[6] Simtech, FLEXLAM®, http://www.simtech.be/eng/produits/produits.aspx?numpage=1&IdGamme=4 [Accesed: 2007-09-13].