The Impact of Bitumen Roofing Production Waste (BTw) on Physical Mechanical Properties of Concrete

Marija Vaiciene¹, Jurgita Malaiskiene², Alessandra Mobili³, Francesca Tittarel³,⁴

¹ Vilnius College of Technologies and Design, Civil Engineering Faculty, Antakalnio str. 54, Vilnius, Lithuania
² Vilnius Gediminas Technical University, Institute of Building Materials, Laboratory of Composite Materials, Linkmenu str. 28, Vilnius, Lithuania
³ Department of Materials, Environmental Sciences and Urban Planning (SIMAU), Università Politecnica delle Marche - ISTM Research Unit, via Brecce Bianche 12, 60121, Ancona, Italy
⁴ Institute of Atmospheric Sciences and Climate, National Research Council (ISAC-CNR), Via Gobetti 101, 40129, Bologna, Italy

m.vaiciene@vtdko.lt

Abstract. This article presents how concrete properties would change if part of a coarse aggregate (granite crushed stone) were replaced with bitumen roofing production waste (BTw). BTw is a huge ecological problem because these wastes are generated in large quantities when replacing old bitumen-based roof tiles. Wastes are also produced during the production of bituminous roof coatings. Usually BTw are stored in landfills or it is attempted to use/dispose them in the production of asphalt concrete. There are very few works which analyse the impact of BTw on the properties of cement materials, although the impact of these wastes on the properties of cement materials could be beneficial because BTw consist of aggregate, granules, bitumen and fibers. In order to use BTw, standard concrete samples were first formed, then 5/16 granite fraction was replaced with BTw in amounts of 2%, 4% and 6% by weight. The amounts of limestone Portland cement, fine aggregate (sand), water and superplasticizer in the concrete mixtures were constant. The new generation of superplasticizer based on polycarboxylates was used in mixtures. The following concrete properties were identified and analyzed: density of the mixture, flowability, density of concrete samples, water absorption, compressive strength, forecasted frost resistance, and microstructure studies were conducted as well. The results of the studies showed that BTw can be used in small amounts, i.e. up to 6%, then the density of the samples slightly decreased (by 2.4%) and water absorption increased (by 0.7%). Compressive strength, after replacing 2% granite crushed stone, decreased by 2.4%. However, gradual addition of the amount of BTw resulted in more closed pores that improved the frost resistance of the concrete. When 6% of bulk filler was replaced with BTw, closed porosity, compared to control samples, increased by 54% and forecasted frost resistance - by 26%. Microstructure analysis showed that with 6% BTw a dense cement stone structure was formed, showing the hydrates of portlandite and CSH.
1. Introduction
One of the current problems in the field of construction and demolition waste management is the recycling/recovery of old and new bituminous roofing materials [1]. Because it can take up to 300 years for asphalt shingles to fully decompose and the volume of asphalt shingle waste produced is so high, it is important that businesses and consumers be aware of viable asphalt shingle recycling methods. In the United States, bituminous roofing waste amounts to approximately 11 million tons a year and only 5% are recyclable [2–6], and this kind of waste is also increasing in Europe. These bituminous roofings are made using bitumen binder or polymer-modified bitumen, the exposed surface impregnated with slate, schist, quartz, vitrified brick, stone, mica, sand, limestone, or other filler [7]. Since they are close to asphalt, they are usually used for the second time in road construction. Authors [8] state that the use of recycled materials such as reclaimed asphalt pavement and recycled asphalt shingle is widely accepted as among the most commonly used sustainable strategies for asphalt concrete pavement due to its ability to partially substitute virgin asphalt binder and aggregate in asphalt concrete mixtures. Scientists [9] investigated the composition of recycled asphalt shingles in different states and sought to use them in warm asphalt mixtures. They found that, depending on composition, from 3% to 20% of bituminous waste can be used for roads. Then road rutting resistance is greatly improved and other properties remain similar. However, [10] the considered waste raises the mixture's embrittlement temperature from about 10°C to 19°C, depending on the preparation temperature of the mixture (120–200°C).

Many scientists [11-13] state that using approximately 5 wt.% of bituminous roofing waste reduces the amount of binder needed and improves the properties of asphalt concrete.

Analyzing the composition of bituminous roofing waste, it is believed that it can be used in small quantities for the production of cementitious hard concrete or lightweight concrete. However, very little research has been done and published in this field direction. Typically, this waste is shredded to a size of 1 mm, and its typical composition consists of 40–70% of filler granules, 20–40% of asphalt, more than 20% of limestone dust or mineral filler and 1–25% of fibrous base material [1]. The use of this waste makes it possible to reduce the density of the concrete, but by adding more, the compressive strength of the concrete is reduced. Waste containing bitumen usually increases the frost resistance of cement concrete [14–15], [1]. Scientists [16] found that fine-shredded bituminous roofing waste accelerates cement hydration, and that the compressive strength remains analogous to the control sample when 5% of sand is replaced by waste.

The main goal of our work is to investigate the production of bituminous roof production waste and evaluate its impact on the physical and mechanical properties of cement concrete. In this work, the possibility to utilize bituminous roof production waste in cement concrete is analysed.

2. Materials and methods
The following raw materials are used to form concrete samples: limestone Portland cement CEM II/A-LL 42.5 N, which conforms the requirements of EN 197-1. Chemical composition of limestone Portland cement is presented in Table 1.

| Chemical composition of limestone Portland cement. |
|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|-----------------|
| CaO             | SiO₂             | Al₂O₃           | Fe₂O₃           | MgO             | K₂O             | Na₂O            | SO₃             | Cl              | L.O.I.           |
| 63.42           | 20.61            | 5.45            | 3.36            | 3.84            | 0.96            | 0.12            | 0.80            | 0.001           | 2.18             |

Mineral composition of the limestone Portland cement: C₃S - 57.26 %, C₅S - 15.41 %, C₃A - 8.68 %, C₄AF - 10.15 %.
According to the granulometry results (Figure 1), the size of the limestone Portland cement particles varies from 0.3 to 140 µm. Particle size distribution is as follows: 50% of the Portland cement particles are 7 µm and smaller in size, the other 40% particles have a size of 7 µm to 50 µm, the remaining 10% have a particle size greater than 50 µm.

The microstructure of limestone Portland cement is shown in Figure 2. In the image, it can be seen that the particles of Portland cement are irregular in shape, their size varies within a wide range. Such properties are characteristic of Portland cement, the majority of which consists of a clinker obtained by burning limestone in a rotary kiln and grinding it in a ball mill. This type of Portland cement has an open matrix, and the products made from such binders are characterised by a porous microstructure. The materials produced from such binders have a higher gas penetration and are easier to penetrate by liquids.

Fine aggregate: natural sand conforming to EN 12620 requirements. Fraction - 0/4. Data on the physical properties of sand are given in Table 2.

| Characteristic          | Tests results |
|-------------------------|---------------|
| Particles density, kg/m³| 2500          |
| Water absorption, %     | 0.57          |
| Bulk density, kg/m³     | 1575          |
Coarse aggregate: granite crushed stone of 5/8 and 11/16 fractions. The characteristics of the coarse aggregate are given in Table 3.

**Table 3. Physical properties of granite crushed stone.**

| Characteristic              | Fraction | Tests results |
|-----------------------------|----------|---------------|
| Bulk density, kg/m³         | 5/8      | 1300          |
| Bulk density, kg/m³         | 11/16    | 1410          |

Waste: bituminous roofing production waste (BTw), which properties are presented in Table 4.

**Table 4. BTw properties.**

| Characteristic            | Fraction 5/16 |
|---------------------------|---------------|
| Bulk density, kg/m³       | 1013          |
| Particles density, kg/m³  | 2276          |
| Water absorption, %       | 0.65          |

All mixtures used a superplasticizer based on polycarboxylic ether polymers. Density is 1.04 g/cm³, pH value at 20°C temperature (20% solution) is 6.5. Superplasticizer conforms the requirements of EN 934-2. Water is according to EN 1008.

The composition of BTw is provided in Table 5. Granulometric composition is shown in Figure 3. The image of used bituminous roofing production waste is presented in Figure 4. The X-ray analysis of BTw is presented in Figure 5.

**Table 5. Composition of BTw.**

| fiberglass, % | Bitumen, % | CaCO₃, % | SiO₂, % | Polymers, % | Bulk minerals (dolomite, hematite, feldspars), % |
|---------------|------------|----------|---------|-------------|-----------------------------------------------|
| 2             | 27         | 30       | 4       | 1           | 36                                            |

**Figure 3. BTw granulometric composition**
Figure 4. The image of used BTw

Figure 5. X-ray analysis of BTw (Q - quartz, C - calcite, H - hematite, D - dolomite, O - feldspar (orthoclase))

It can be seen from the X-ray analysis, that BTw consists of minerals such as quartz, calcite, feldspar (orthoclase), dolomite, hematite.

Four concrete mixtures with a designation codes K0, K2, K4 and K6 are mixed during the research. K0, K2, K4 and K6 compositions are given in Table 6. Additionally, 2% of BTw is added to the concrete mixture K2, 4% - to the mixture K4 and 6% - to the mixture K6. The amount of waste is calculated as a percentage by weight (0%, 2%, 4% and 6%) of the 5/16 granite crushed stone fraction. The water/cement ratio in the mixture is 0.55. The amount of superplasticizer is 0.6% by cement weight. Density of the mixtures is determined according to EN 12350-6.

Table 6. Compositions of concrete mixtures.

| Mark | Cement, kg/m³ | Sand, kg/m³ | Granite crushed stone 5/16, kg/m³ | Water, kg/m³ | Plasticizer kg/m³ | Waste, kg/m³ | Waste, % | W/C |
|------|---------------|-------------|----------------------------------|--------------|-------------------|--------------|---------|-----|
| K0   | 300           | 980         | 1000                             | 165          | 1.8               | 0            | 0       | 0.55|
| K2   | 300           | 980         | 980                              | 165          | 1.8               | 20           | 2       | 0.55|
| K4   | 300           | 980         | 960                              | 165          | 1.8               | 40           | 4       | 0.55|
| K6   | 300           | 980         | 940                              | 165          | 1.8               | 60           | 6       | 0.55|
100×100×100 mm-sized samples of the prepared mixture were stored for 1 day under normal conditions, then for 27 days in water at 20°C±2°C. Production and hardening of concrete samples for strength determination is according to LST EN 12390-2. Compressive strength of concrete samples according to LST EN 12390-3. Density is determined based on LST EN 12390-7. Water absorption of samples is established according to the methodology described in literature [17].

The X-ray diffraction (XRD) analysis of the phase composition of BTw was carried out upon applying diffractometer DRON-7. In order to obtain X-ray radiation Cu Kα spectrum (λ = 0.1541837 nm), a graphite monochromator was used. The parameters of the tests were following: voltage - 30 kV; current - 12 mA; the range of the diffraction angle - from 4 to 80°, the detector movement step - 0.02°; the duration of the intensity measuring in a step - 0.5 s. Phase identification was carried out by decoding the XRD patterns according to ICDD diffraction databases.

ZEISS 1530 SEM, Carl Zeiss, Oberkochen, Germany, equipped with an EDAX probe, Schottky emitter, with two different secondary electrons detectors, the in-lens and the Everhart-Thornley and operating at 10 keV are performed in order to investigate the microstructure of concrete. The microstructure of the limestone Portland cement is examined with a scanning microscope SEM (EVO LS 25, Zeiss, Germany). The particle size of the limestone Portland cement is tested with a CILAS 1090 DRY analyser.

The forecasted frost resistance is determined by the $K_f$ coefficient which is obtained based on [18–19]. By determining the total and open porosities ($P_a$) based on their difference, the closed porosity of the concrete ($P_u$) is calculated. The coefficient $K_f$ is then calculated by the formula:

$$K_f = \frac{P_u}{P_a \cdot 0.09} \quad (1)$$

Then, according to Figure 6, the quantity of freeze-thaw cycles is forecasted for the concrete [19].

![Figure 6. The influence of frost resistance factor $K_f$ on frost resistance of concrete](image)

### 3. Results and discussions

The data in Table 7 show that the density of concrete mixtures in all batches remains similar, ranging from 2340 to 2370 kg/m³. The flowability class of all concrete mixtures is F3 (from 420 to 450 mm).

In the course of the work, the average values for physical and mechanical strip properties are calculated. Average density and compressive strength of concrete samples made from mixtures K0, K2, K4, and K6 are shown in Figures 7 and 8. The obtained data show (Figure 7) that when concrete is mixed with different amount of BTw the density of the samples varies from 2267 to 2322 kg/m³. Control samples and samples with 2% and 4% BTw have similar densities. The highest density is obtained for concrete samples made of a mixture K0 and the lowest - for K6. With a maximum waste content of 6%, the density decreases by 2.4% compared to control samples. The reduction in the
density of concrete is determined by the lower density of BTw particles compared to granite crushed stone, as found by authors in [20] and [21] for other types of wastes used as replacement of natural aggregates.

Table 7. Density and flowability of concrete mixtures.

| Mark | Mix density, kg/m³ | Mix flowability, mm |
|------|-------------------|---------------------|
| K0   | 2360              | 420                 |
| K2   | 2370              | 440                 |
| K4   | 2363              | 450                 |
| K6   | 2340              | 420                 |

Figure 7. Dependence of concrete density on BTw amount

The results in Figure 8 show that as the amount of BTw increases, the strength of the concrete samples decreases. The average compressive strength of the samples prepared from the control mixture is 37.3 MPa. When 2% BTw is added in the mixture, the compressive strength decreases by 2.4%, and at 4% of BTw, it is reduced by only 1.3% compared to the control samples. In this case, the strength of the samples changes only within the margin of error. By further increasing the amount of waste to 6%, the compressive strength begins to decrease significantly - by 12.6%. The reduction in strength is determined by the decrease in density and the formation of a larger area of contact zones between BTw and the cement matrix.

Figure 8. Dependence of concrete compressive strength on BTw amount
In Figure 9, microstructure of concrete sample with 4% BTw after 28 days of hydration is shown. During hydration, C-S-H microcrystals are formed, which, due to the optimal spatial distribution and adhesion, form the basis of cement stone strength. Microstructure analysis shows that with 4% BTw a denser structure of cement stone with the hydrates of portlandite and CSH is formed.

![Figure 9. Image of microstructure](image)

Water absorption results of concrete are presented in Figure 10. After 96 hours of soaking in water, very similar water absorption values are obtained. Water absorption by batch ranges within the margin of error, i.e., from 4.4% to 4.5%. The kinetics of water absorption is slightly different, but basically BTw does not increase the water absorption on concrete.

![Figure 10. Dependence of concrete water absorption on BTw amount](image)

The open and closed porosities of the concrete samples as well as the K_f coefficient are given in Table 8 and the forecasted frost resistance based on K_f coefficient is given in Figure 11.
Table 8. Values of closed, open porosities and \( K_f \) coefficient.

| Mark | Open porosity \( P_o, \% \) | Closed porosity \( P_c, \% \) | \( K_f \), values |
|------|-----------------|-----------------|-----------------|
| \( K_0 \) | 10.1            | 3.9             | 4.2             |
| \( K_2 \) | 10.5            | 3.6             | 3.9             |
| \( K_4 \) | 10.4            | 4.2             | 4.5             |
| \( K_6 \) | 10.0            | 6.0             | 6.7             |

The results in Table 8 show that \( K_6 \) concrete samples have the lowest open and the highest closed porosities. The \( K_f \) coefficient of this batch, calculated according to formula (1), is the highest and its value is about 60\% higher than that of the control samples. \( K_4 \) samples with 4\% BTw have slightly higher \( K_f \) value compared to the control sample. Figure 11 shows that after adding 4\% BTw to the mixture, the frost resistance of the samples increases by 3\% compared to the control sample and increases by 26\% when 6\% BTw is added.

4. Conclusions

Bituminous roofing production waste has been investigated to be valorized in concrete mixtures. The main constituents of BTw are bitumen, calcite and mineral bedding. According to the X-ray analysis, BTw is made up of quartz, calcite, feldspar (orthoclase), dolomite and hematite minerals.

Assessing the results of the study of the physical-mechanical properties of cement concrete, 4\% of BTw replacing gravel by weight is a suitable and optimal amount for the preparation of cement concrete mixtures. The compressive strength and water absorption remain similar to the control samples, it reduces the density and increases the frost resistance of the concrete, resulting in a dense microstructure with visible C-S-H hydrates and portlandite.

References

[1] S. Guo, J. Hu, Q. Dai, “A critical review on the performance of Portland cement concrete with recycled organic components,” J. Clean. Prod., vol. 188, pp. 92–112, 2018.

[2] S. Shirzad, M. A. Aguirre, L. Bonilla, M. A. Elseifi, S. Cooper, L. N. Mohammad, “Mechanistic-empirical pavement performance of asphalt mixtures with recycled asphalt shingles,” Constr. Build. Mat. vol. 160, pp. 687–697, 2018.

[3] J. R. Willis, P. Turner, “Characterization of asphalt binder extracted from reclaimed asphalt shingles,” National center for asphalt technology. 2016.

[4] T. Townsend, P. E. J. Powell, E. I. C. Xu, “Environmental issues associated with asphalt shingle recycling”. Prepared by: Innovative waste consulting services, LLC Gainesville, Florida. 2007.
[5] B. Sengoz, A. Topal, “Use of asphalt roofing shingle waste in HMA,” Constr. Build. Mat. vol. 19, pp. 337–346, 2005.

[6] US EPA. Resource conservation. from roofs to roads; 2012. <http://www.epa.gov/wastes/conserve/imr/cdm/pubs/roof_br.pdf> [accessed July 2013]

[7] B. Nam, H. Maherinia, A. H. Behzadan, “Mechanical characterization of asphalt tear-off roofing shingles in hot mix asphalt,” Constr. Build. Mat. vol. 50, pp. 308–316, 2014.

[8] R. Yang, S. Kang, H. Ozer, I. L. Al-Qadi, “Environmental and economic analyses of recycled asphalt concrete mixtures based on material production and potential performance,” Res. Cons and Rec, vol.104, pp. 141–151, 2015.

[9] A. A. Cascione, R. C. Williams, J. Yu, “Performance testing of asphalt pavements with recycled asphalt shingles from multiple field trials,” Constr. Build. Mat., vol 101, pp. 628–642, 2015.

[10] J. W. Arnold, B. Behnia, M. E. McGovern, B. Hill, W. G. Buttlar, H. Reis,”Quantitative evaluation of low-temperature performance of sustainable asphalt pavements containing recycled asphalt shingles (RAS),” Constr. Build. Mat., vol. 58, pp. 1–8, 2014.

[11] Vermont agency of natural resources, recycled shingles in road applications; September 1999.

[12] D. Hansan, K. Y. Foo, T. A. Lynn. Evaluation of roofing shingles in HMA. National center for asphalt technology; 1997.

[13] Polk county waste resource management division, Jones Edmunds and inovative waste consulting service. Beneficial use of asphalt shingles from construction and demolition debris in hot mix asphalt plants; Florida Department of Environmental Protection; 2010.

[14] X. Shu, B. Huang, “Recycling of waste tire rubber in asphalt and Portlandcement concrete: an overview,” Constr. Build. Mat. vol. 67, pp. 217–224, 2013.

[15] R. Si, S. Guo, Q. Dai, “Durability performance of rubberized mortar andconcrete with NaOH-Solution treated rubber particles,” Constr. Build. Mat., vol. 153, pp. 496–505, 2017.

[16] J. An, B.H. Nam, H. Youn, “Investigation on the effect of recycled asphalt shingle (RAS) in Portland cement mortar,” Sustainability, vol. 8(384), pp. 1–16, 2016.

[17] R. Mačiulaitis, “Frost resistance and durability of façade bricks. Frostwiderstand und Dauerhaftigkeit keramischer Fassadenerzeugnisse. Fasadinės keramikos atsparumas šalčiui ir ilgaamžiškumai”. Vilnius: Technika, 132 p. 1996.

[18] M. Daukšys, E. Ivanauskas, S. Juočiūnas, D. Pupeikis, L. Šeduikytė, “The assessment of prediction methodology of concrete freezing and thawing resistance,” Mat. Sc. (Medžiagotyra), vol. 18(4), pp. 403-409, 2012.

[19] A. E. Sheikin, L. M. Dobshic, “Cement concrete and high frost resistance,” Stroyizdat, Len. Dep., N., 1989, p. 128 (in Russian).

[20] A. Mobili, C. Giosuè, V. Corinaldesi, F. Tittarelli “Bricks and concrete wastes as coarse and fine aggregates in sustainable mortars” Adv. Mater. Sci. Eng. vol. 2018, pp. 1-11, 2018.

[21] F. Tittarelli, C. Giosuè, A. Mobili “Recycled glass as aggregate for architectural mortars” Int. J. Concr. Struct. Mater. vol. 12(1), p. 57, 2018.