

Article

Sustainable Use of Reservoir Sediment through Partial Application in Building Material

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Abstract: Sediment, often considered a by-product of various activities within river basin management to be disposed of, or a pollutant to be controlled, is increasingly being acknowledged as a resource in need of management. The paper deals with the possibility of reusing sediment from two Slovak reservoirs (Klusov and Ruzin) as an alternative raw material in concrete production. Concrete specimens were prepared by a combination of original reservoir sediment, reservoir sediment mechanically activated by dry milling, reservoir sediment mechanically activated by dry milling together with biomass incinerator fly-ash as a binder. To improve the strength properties of specimens, sodium hydroxide (NaOH) was used as a sediment activator. Mixtures containing 40% of binder replacement by the above-mentioned combinations of original and treated sediments were tested for flexural and compressive strengths after 28, 90 and 365 days of curing. The results showed that the mixtures prepared from sediments milled without and with addition of fly ash as cement replacement satisfied the strength requirements for the compressive strength class C16/20 according to the European standard except the composites prepared with NaOH as the sediment activator. Addition of NaOH into composites in the concentration of 5 M as an activator of sediment indicated the negative impact on compressive and flexural strengths and thus NaOH was not an effective pozzolanic activator for sediments. This study reveals that the sediment may be considered as 40% cement substitution in building materials.

Keywords: cement; sediment; concrete; replacement; activator

1. Introduction

Declining of the storage capacity and the lifetime of water reservoirs, significantly reduced by the high rates of sedimentation, are at present important sediment-related problems. In natural and agricultural basins, sediment is derived from weathering and erosion of minerals, organic material and soils in upstream areas and from erosion of river banks and other in-stream sources [1]. The sedimentation of reservoirs over multiple years causes large capacity losses of the reservoirs. Many studies that have been undertaken to estimate the sediment load of the world's rivers vary widely in terms of the assumptions made in the studies and reported effects of accelerated erosion due to human activities thereby reflecting the difficulty in obtaining reliable values for sediment concentration and discharge in many countries. Wohl [2] presented that human enterprises which cause soil erosion have increased the transport of sediment by the world's rivers approximately 2.3 billion tons a year, but the storage in reservoirs has more than compensated for this increase, reducing sediment flux to the oceans by 1.4 billion tons a year. Over 100 billion tons of sediment are now stored in reservoirs built mostly within the past fifty years. According to Walling [3] the annual sediment load of all the world's rivers together varies between 24 and 30 billion tons, but it depends on the river and the discharge. The reservoir sedimentation occurs worldwide at a rate of about 0.8 percent per year, but the sedimentation

rate in many regions is higher [3]. The study by White [4] showed that worldwide approximately 40,000 large reservoirs suffer from sedimentation and it was estimated that between 0.5% and 1% of the total storage capacity is lost annually. The same value of 1% was reported in [5]. The average age of reservoirs is now about 30 years and since many reservoirs have been designed with a dead storage for sedimentation of about 50 years, serious sedimentation problems are going to develop with about 40 percent of the storage capacity in reservoirs affected within the next 20 years. Most of the existing reservoirs in the world could be completely silted up in 200–300 years from now [3]. Inflow of sediments into many reservoirs and better care of long term reservoir sustainability emphasize the importance of sediments dredging. Periodic sediment removal is important in order to maintain the effectiveness of these structures [6] and the management of dredged sediments is a significant issue in the world. Several assessments demonstrated that the total amount of sediment dredged in Europe reaches 100–200 million cubic meters per year [7].

It is not only the quantity of sediment which affects downstream areas but also its quality [8] that complicates sediment management. Sediment is on the borderline of soil, waste and water, which has in general resulted in a non-coherent legislation from these sectors [9]. According to the Directive on Hazardous Waste [10] and the European Waste Catalogue [11], contaminated dredged material is categorized as a waste, generating its negative image. The options for such sediment are limited to processing (treatment) or confined disposal. Placing dredged material in a landfill facility is one of the most expensive propositions and is only practiced when no other solutions are available [12,13]. Sediments mainly got local attention of water managers confronted with manmade sediment-traps, where associated contamination poses an environmental or human risk [14]. However, the implementation of the Water Framework Directive (WFD) [15] shifts the scope from the local sediment management (amongst them also dredged material) to the river basin scale sediment management [16]. The WFD does not specifically address sediment nor dredged material, although sediments are a natural and essential part of the aquatic environment and their management has to play an important role in water legislation [15–17]. However, the favorite dredged material management options are natural options and beneficial re-use is a way to encourage the use of dredged material as a potential resource and not as a waste [18]. Management options for the dredged material depending on its physical and chemical characteristics [19–21] are sediment relocation and the use of dredged material as substrate for soil amendments to enhance agriculture [22] or retaining sediment within the natural sediment system to support sediment-based habitats [1,23]. With the shortage in some regions of the world of natural resources in many engineering fields, the beneficial reuse of sediments as a construction material [19,24] can optimize the management of the natural resources [25]. Clay and silt as the most common materials acquired from the maintenance dredging in rivers, reservoirs and ports, can find more engineering uses [18].

Many investigations have revealed the use of dredged sediments as a minor or major component in construction industry. The production of lightweight aggregates (LWA) made from dredged sediments has been evaluated in many studies [26–30] and the results have shown their suitability for lightweight aggregate production due to their perpetual availability, homogeneity and mineralogical and chemical composition [26]. The practical use of dredged sediments into brick production as a suitable alternative to their current disposal paths has also been demonstrated [31–35]. In the study provided by Samara et al. [31] sediments were introduced into the brick-manufacturing process as a replacement of quartz sand with a substitution ratio of 15%. This substitution resulted in an increased compressive strength (63%) and decreased porosity (10%). Results presented in [34] reported that 50% replacement of natural brick-making clay by sediments reached the compressive strength meeting the American Society for Testing and Materials (ASTM) requirements for the severe grade building brick and reuse of sediments could be an option for clays. In other studies [36,37] replacement of sand by sediments in construction with a substitution ratio up to 19% was assessed. The mixtures of contaminated sediment and mineral modifiers melted together to produce cement were studied in the USA [38]. The feasibility of using the dredged sediment as partial replacement of cement in mortars is

presented by Kazi Aoual-Benslafa et al. [39]. The authors studied the compressive strength and weight losses mortars with addition of sediment at varying percentages (5%, 10%, 15% and 20% by mass of cement). The results showed that sediments can most suitably be substituted for the 5% of the cement used [39].

The objective of this paper is to assess the applicability of reservoir sediments as a replacement of cement in concrete production. The use of 40% cement replacement by original dredged sediments and sediments treated by mechanical and chemical activation is the novelty in this scientific field.

2. Materials and Methods

2.1. Study Area

A laboratory-scale study was conducted with the sediments dredged from the Klusov and Ruzin reservoirs situated in Eastern Slovakia. The Klusov small water reservoir is situated in the agricultural watershed in Bardejov district and it falls in the Topla partial river basin. Its total capacity is about 72,000 m³ and it covers the area of 0.022 km². The Ruzin water reservoir is located in the north-eastern part of the Slovak Ore Mountains in the Kosice region. It is situated on the inlet of the Hornad river and its main tributaries namely Hnilec, Opatka and Bela. This territory was historically known for intensive mining and ore processing activities. The Ruzin reservoir has a capacity of 59 million cubic meters with the surface area of 3.9 km².

The siltation measurements of these reservoirs have shown that 260,000 m³ of sediment settle in the Ruzin reservoir (0.5% of its total capacity) [40] and 1300 m³ in the Klusov reservoir (1.8% of its total capacity) [41], every year. A significant part of the sediment comes from soil erosion in the watershed and through bank abrasion. This highlights the need for sediment management and one option for such management might be to consider using the sediment in building materials.

2.2. Material Characterization

From the Klusov reservoir, the composite sediment sample was taken from the area near the dam (SK, GPS 49.25113°N, 21.23377°E) and at the inflow to the Ruzin reservoir (SR, GPS 48.82191°N, 21.08018°E). In laboratory conditions, the composite sediment samples were air dried at laboratory temperature, mechanically homogenized and quartered.

Particle size distribution (PSD) of sediment samples was realized using a Mastersizer 2000 (Malvern Instruments Ltd, Malvern, Worcestershire, UK) particle size analyser with wet sample dispersion. Measurement parameters were set to 2800 rpm for pump speed and ultrasound turned on for 5 min due to disaggregation of sediment particles. Crystalline minerals in the reservoir sediments were studied by the X-ray powder diffraction (XRD) method, under Cu-K α radiation, with a Bruker D2 Phaser instrument (Bruker AXS GmbH, Karlsruhe, Germany). The step size of 0.04° (2 θ) and step time of 3 s for about 2 h were the running parameters. X-ray fluorescence (XRF) spectrometry, through SPECTRO iQ II (Spectro Analytical Instruments GmbH, Kleve, Germany), was used to determine the chemical composition of the sediments. Fourier transform infrared spectroscopy (FTIR) was used to confirm the carbonate and calcium silicate formation of hardened mixtures using a Spectrometer Alpha-T (Bruker Optik GmbH, Ettlingen, Germany) with the Attenuated Total Reflection (ATR) technique. Measurements were carried out in transmittance mode, in the range 400–4000 cm⁻¹, with resolution of 4 cm⁻¹.

2.3. Preparation of the Mixtures

After the initial physico-chemical characterization, concrete specimens were prepared with 40% Portland cement replacement by a combination of original reservoir sediment. This proportion of 40% was selected from the literature [42] and was deemed suitable as supplementary cementitious materials containing less CaO can be blended into cements only up to 30–40% total weight without significant loss in binding properties. The 40% proportion of replacement is generally being considered as the

most effective way of reducing the environmental impact and CO₂ emissions from the cement and concrete production [42]. Also, the problem with sediment disposal could be solved. In the first stage of the study, the sediments from the Klusov small water reservoir (SK) were used as a partial cement replacement. From an economic and technical point of view and due to the fine-grained texture of these sediments similar to cement, they were used in concrete without any modifications. The second part of the research focused on the reuse of sediments from the Ruzin water reservoir (SR) due to their perpetual quantity and subsequently environmental and water management problems. Due to their coarse-grained texture, sediments from Ruzin were mechanically activated by dry milling. The aim was to optimize the composition of the blended concrete specimens to improve the strength of the concrete mixtures prepared with these sediments as filler. In addition to milling, the sediment from Ruzin was activated by two other ways. For the low proportion of CaO in sediment sample, the fly-ash (FA) from local biomass incinerator plant was added into the dry milling process. Also, reservoir sediments were chemically activated with the addition of sodium hydroxide as an activator into milling process. This activator was chosen based on the previous studies [43–46], which concluded that the inclusion of NaOH as chemical activator into geopolymer matrix increased the compressive strength of the hardened geopolymer concrete. Alkali activators transform the amorphous structure into a skeletal structure that exhibits cementitious properties [47]. Polymeric gel with variable composition is formed in the media of high alkalinity. The gel is produced when the solution of high alkalinity reacts with the starting materials. The main behaviour of the products formed via these conditions is that it exhibits excellent mechanical properties at the early period of hydration. Granizo et al. [48] stated that mixing of metakaolin with certain quantities of sodium hydroxide possesses interesting mechanical properties. Palomo et al. [49] suggested that the activation of metakaolin (Si + Al) with a medium to high alkaline solutions is the alkali activation model [45]. For low-calcium precursors, rich in SiO₂ and Al₂O₃ (such as the studied sediment), the alkali activation is suitable [50,51].

The detailed mixing proportions are presented in Table 1. Portland cement (CEM I 42.5 N) and two different fractions of natural river aggregate (0/4 mm and 4/8 mm) were used as other raw materials for preparing the concrete mixtures. Natural aggregate was evaluated according to the standard STN EN 12620 [52]. The concrete specimen composition was designed according to STN EN 206-1 [53] for strength class C 25/30. Water-to-cement ratio of 0.55 was used and no plasticizer was added.

Table 1. Mixing proportions of blended concrete specimens.

Mixture Class C25/30 **	Binder (kg/m ³)		Filler-Natural Aggregate (kg/m ³)		Water (kg/m ³)
	CEM I	SK/SR	Fraction 0/4 mm	Fraction 4/8 mm	
Control SM0	350	-	1123	717	193
SKM1	210	140	1123	717	193
SRM1	210	140 (milled)	1123	717	193
SRM2	210	140 (milled with NaOH)	1123	717	193
SRM3	210	140 (milled with FA)	1123	717	193
SRM4	210	140 (milled with FA and NaOH)	1123	717	193

SK/SR—sediments from Klusov/Ruzin reservoir; SKM/SRM—mixtures prepared with sediments from Klusov/Ruzin reservoir; Control SM0—control mixture; FA—fly ash; ** see text below for exact mixture details.

Control mixture (SM0) was prepared by mixing cement (CEM I 42.5 N), natural river aggregate with particle size sorts 0/4 and 4/8 mm and water at solid/liquid ratio of 0.55. The mixture SKM1 was prepared using 40:60 proportions of fine-grained sediment from the Klusov reservoir and Portland cement. The mixture SRM1 was prepared using 40% of coarse-grained sediment from Ruzin milled for 3 min in laboratory vibrating mill as a cement replacement. The mixture SRM2 was prepared using 40% of sediment from Ruzin milled for 3 min in laboratory vibrating mill together with granulated NaOH in a ratio of 2/1 (sediment/NaOH) to achieve 5 M NaOH solution. The mixture SRM3 was prepared using 40% of sediment from Ruzin milled for 3 min in laboratory vibrating mill together with biomass incinerator fly-ash in a ratio of 1/1 (sediment/fly ash). The mixture SRM4 contained 40% of sediment from Ruzin milled for 3 min in laboratory vibrating mill together with biomass

incinerator fly-ash and granulated NaOH in a ratio of 1/1/1 (sediment/fly ash/NaOH). Double set of mixtures were prepared. For testing flexural strengths of the hardened concrete mixtures, three prisms (40 mm × 40 mm × 160 mm) were cast per mixture and studied age. The tests carried out after 28, 90 and 365 days of curing according to the STN EN 12390-5 [54]. Load was applied gradually until the specimens failed. After test of flexural strength, the prism halves were tested in compression according to the STN EN 12390-3 [55]. ELE ADR 2000 (ELE International Ltd, Leighton Buzzard, Bedfordshire, UK) testing machine was used for all studying composites.

3. Results and Discussion

3.1. Properties of Dredged Sediments

The particle size distribution of original sediments dredged from the Klusov and Ruzin reservoir and mechanically activated (milled) sediment from Ruzin, characterized by grading curves, is presented in Figure 1.

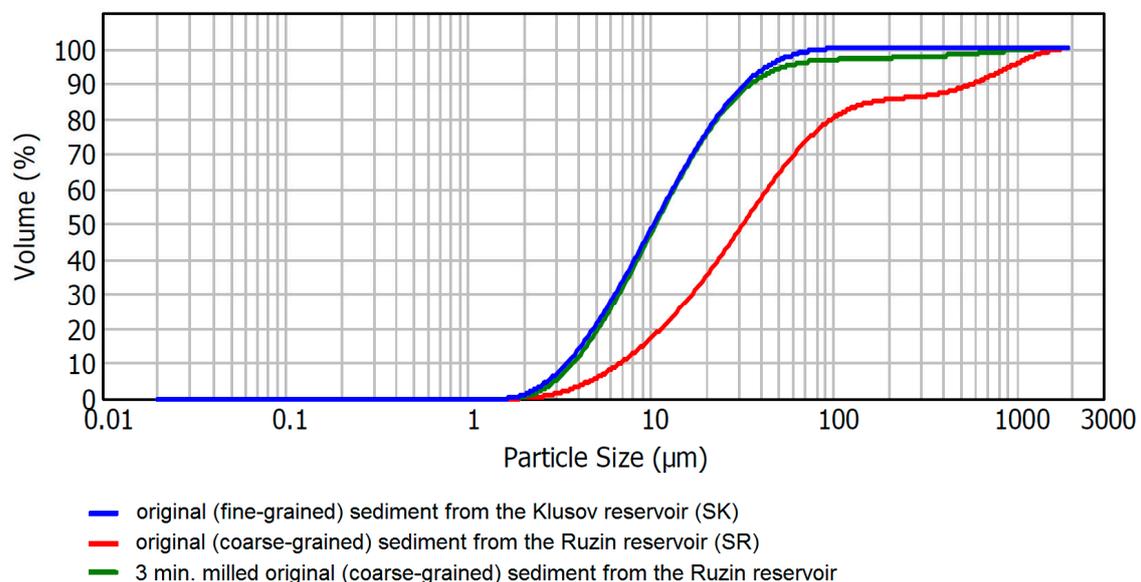


Figure 1. Particle size distribution measured for original and mechanically activated sediments.

These results illustrate that the clay and silt-sized material, with a proportion of the particles below 50 µm exceeding 95%, dominated in fine-grained sediment sample dredged from the Klusov reservoir from the area near the dam. However, the original sediment sample dredged from the Ruzin reservoir contained only about 56% of particles smaller than 50 µm. Fine-grained sediment texture similar to cement grain size was achieved by milling the sediment for 3 min. After milling, the proportion of these particles increased to 94%.

The XRD measurements were realized for original sediments dredged from both reservoirs (Figures 2 and 3) and for fly ash (Figure 4). The X-ray diffractogram of the sediments demonstrates that quartz peaks are dominant and remarkably visible in both samples, followed by muscovite and nontronite peaks, eventually albite in Ruzin sediments, what confirmed the presence of silicates. Biomass fly ash has a diverse mineralogical composition and it is rich in carbonate (calcite), hydroxide (portlandite) and sulphate (arcanite) minerals.

To assess the suitability of the composition of the raw material for composites manufacture, the chemical composition of the sediments and fly ash was determined by XRF spectrometry. Chemical composition data of the dredged sediments and fly ash expressed as percentage of major oxides is shown in Table 2 and is compared with chemical composition of the Portland cement.

The data indicate that the main components in the sediments were SiO₂ and Al₂O₃. Also, the presence of Fe₂O₃ was recorded in noticeable concentration. The presence of these oxides is essential for the formation of water-insoluble solid structures in Portland cement manufacture. The silica ratio, parameter useful in describing clinker characteristics and typically between 2.0 and 3.0, is 2.9 for dredged material from Klusov and 2.5 from Ruzin. The SiO₂ content in sediments is associated with quartz particles, while Si, Al and Fe oxides are associated with muscovite, nontronite and clinochlore structure present in the sediments, as was confirmed with the XRD measurements. In the fly ash and Portland cement, the main component was CaO associated with calcite and portlandite minerals. The results showed similar chemical and mineralogical composition of sediments in the studied reservoirs.

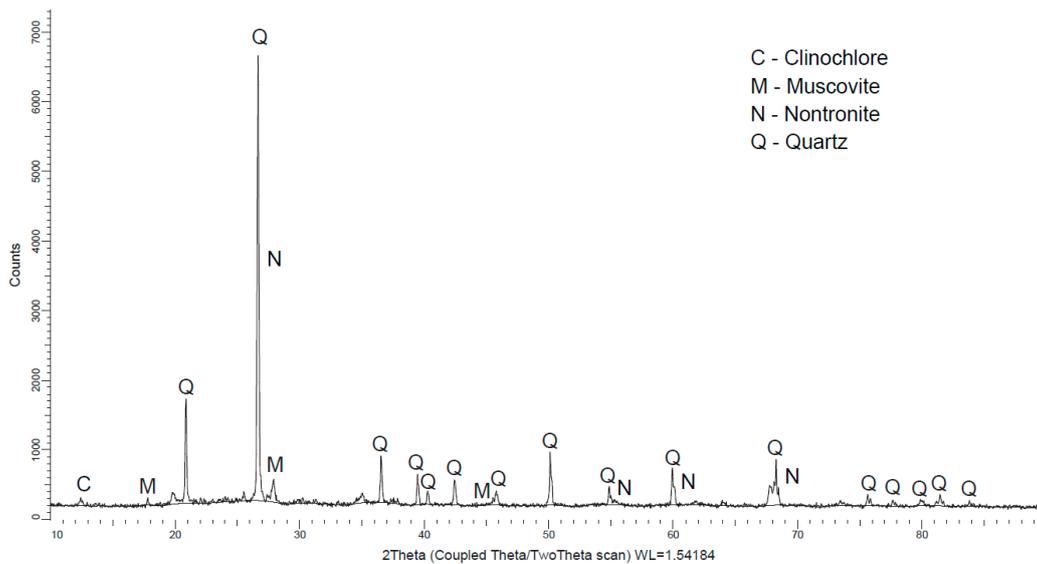


Figure 2. Diffraction patterns of the sediment dredged from the Klusov reservoir (SK).

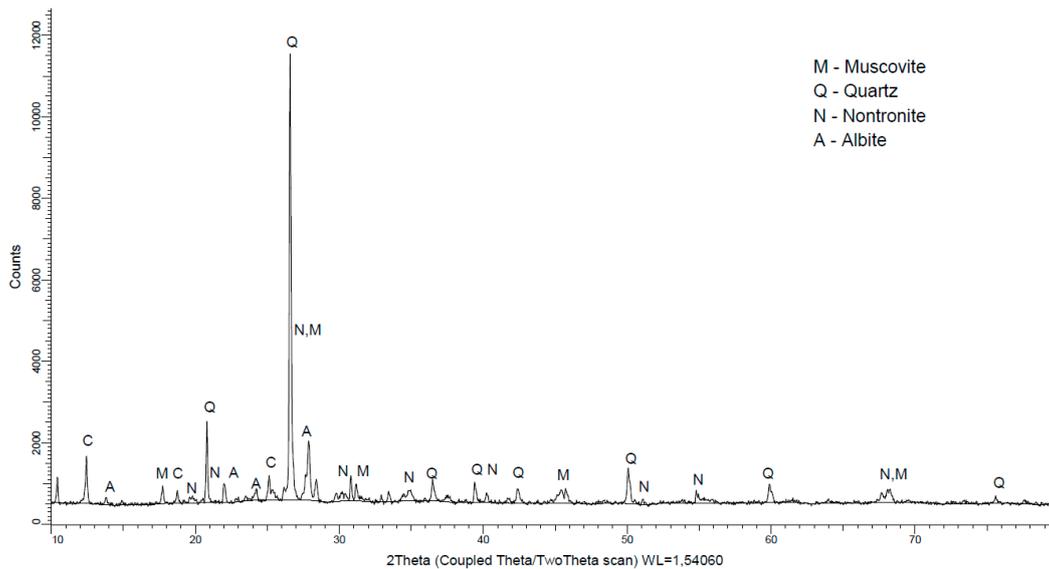


Figure 3. Diffraction patterns of the sediment dredged from the Ruzin reservoir (SR).

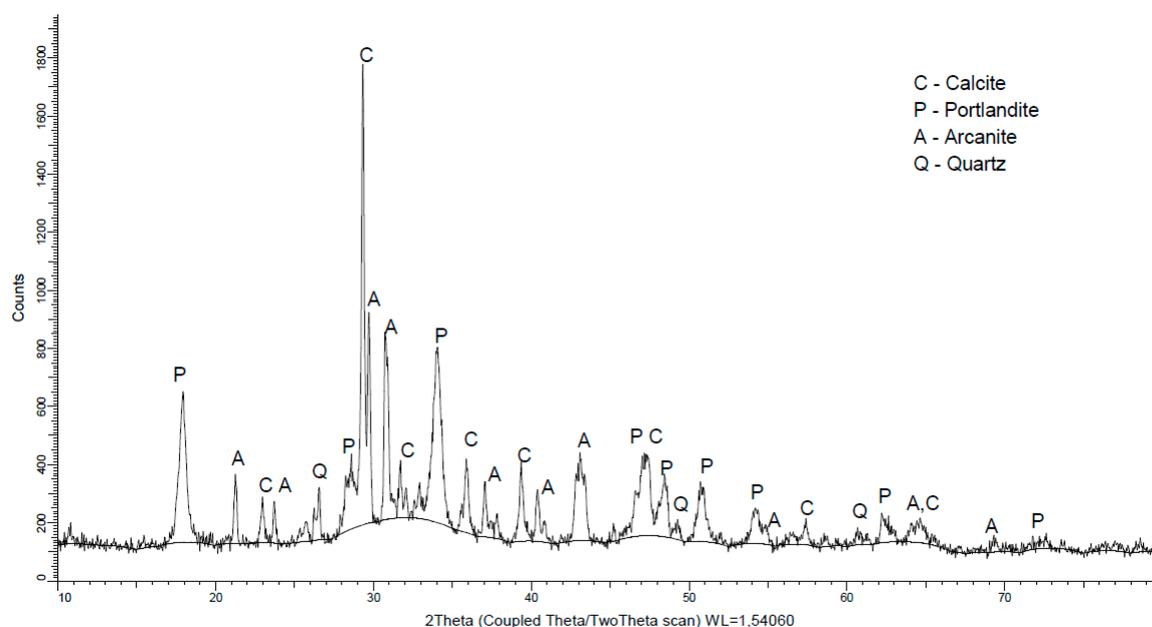


Figure 4. Diffraction patterns of the biomass incinerator fly-ash (FA).

Table 2. Chemical composition of raw materials.

Total weight (%)	Sediment Klusov	Sediment Ruzin	Biomass Fly Ash	Portland Cement
SiO ₂	68.56	63.09	4.71	19.5
Al ₂ O ₃	18.1	16.76	1.22	4.7
Fe ₂ O ₃	5.15	7.95	0.86	3.2
CaO	1.10	2.59	43.04	64.2
MgO	2.40	5.49	5.58	1.3
K ₂ O	2.95	2.37	11.49	0.78
SO ₃	0.11	0.06	6.58	3.2
Cl ⁻	0.034	0.016	0.92	0.047
LOI	5.1	5.8	1.55	0.9

3.2. Sediment as a Binder

In order to find out the applicability of the original and the treated reservoir sediments as a partial replacement of cement in concrete, prepared composites were subjected to compressive and flexural strengths after 28, 90 and 365 days of curing. The results are illustrated in Figures 5 and 6. All data are presented as the mean \pm standard deviation of triplicate results for the flexural strength and as the mean of the six determinations made on a double set of three prisms for compressive strengths. Determinations that varied by more than $\pm 10\%$ from the mean were discarded.

Composites, prepared with 40% Portland cement replacement by sediments from the Klusov (SKM1) and Ruzin (SRM1) reservoirs with the similar PSD, chemical and mineralogical composition, showed analogous development of compressive strengths. By comparing these samples, it can be concluded that the compressive strength after 28 days of hardening was about 20 MPa in average for both mixtures, and after 365 days, the compressive strength of SRM1 was lower by only about 16%, compared with the SKM1 sample. The mixture SRM1 exhibited slower rise in strength with increasing curing time. After 28 days of hardening these composites (SKM1 and SRM1) achieved half the strength values of the control concrete mixture. After 365 days, the strengths of both composites slightly increased to 55/64% (SRM1/SKM1) compared to the control mixture.

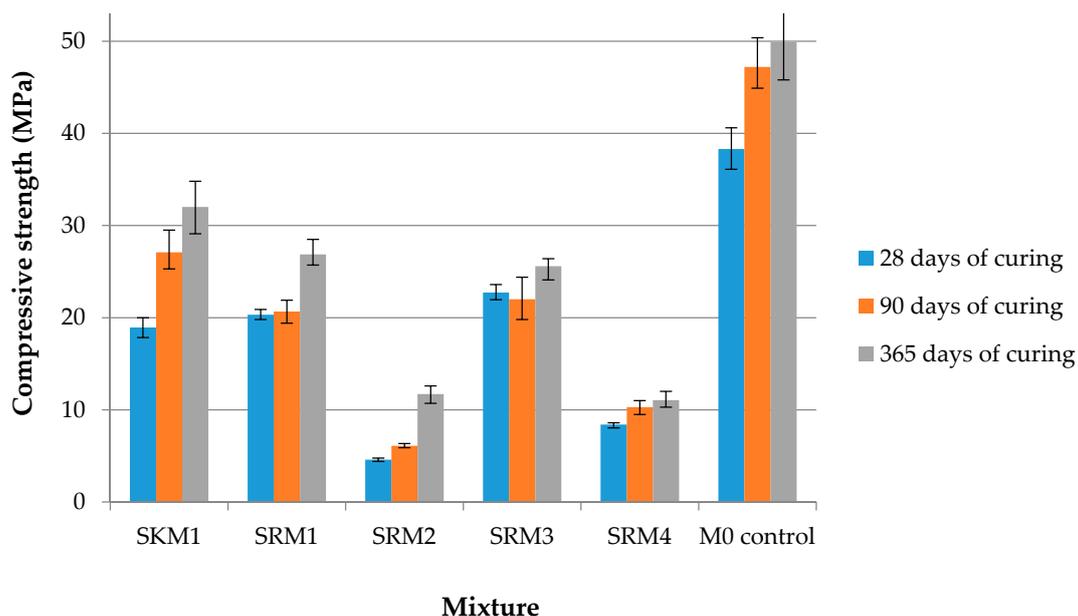


Figure 5. The compressive strength development of prepared composites.

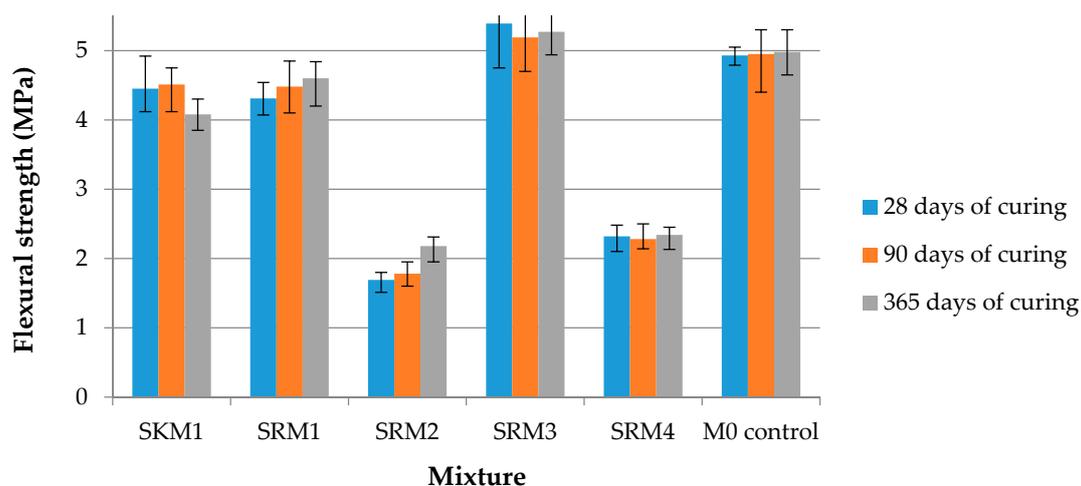


Figure 6. The flexural strength development of prepared composites.

Another composite (SRM3), prepared with the addition of biomass fly ash into milling process together with sediment (due to the low content of CaO in the sediment), achieved 12% higher 28-days compressive strength, compared with SRM1 (without FA). Compressive strength after 365 days reached a value almost nearly identical to the SRM1 (26 MPa). The expected increase in compressive strength due to the addition of fly ash has not been demonstrated. The reason for this could be due to the low concentration of components in fly ash such as SiO_2 and Al_2O_3 necessary for C-S-H/C-A-H gel formation responsible for the strength.

The lower compressive strength values of composites SKM1, SRM1 and SRM3 at 50–60% of the control mixture might have been caused by the higher value of LOI of sediments (5–6%), which led to the lesser strength development.

The last group consisted of composites prepared from Ruzin sediment activated by milling together in the presence of a NaOH activator (SRM2) and combination of fly ash and NaOH (SRM4). These composites achieved the lowest values of the compressive strengths among all prepared mixtures.

Higher initial strengths were achieved in the sample SKM4 by the addition of fly ash. After 365 days, the values of compressive strengths reached about 11 MPa in both the samples. The lower values of compressive strength of SRM2 and SRM4 mixtures compared to others indicate that the conversion degree to C-S-H gels in composite system is probably too low due to a higher OH^- concentration in the system, that causes the shift of equilibrium towards the left side in the hydration process of C_3S and C_2S [45,47]. C-S-H gels can be formed from precipitated $\text{Ca}(\text{OH})_2$ at pH around 12.4 [56]. However, the addition of NaOH caused a higher pH of the fresh mixture (around 13.8) and retarded the process of mixture hydration [57,58]. According to Martinez-Ramirez and Palomo [47] as the alkaline concentration of the hydrating solution increases, the degree of hydration of anhydrous silicates reduces, and for this reason, the compressive strength also gets reduced. The comparison of the compressive strength development for composites with and without NaOH addition (SRM2 vs. SRM1 and SRM4 vs. SRM3) confirm this fact.

The structure development during hardening reflecting compressive strength increase can be demonstrated also through FTIR spectra of prepared composites compared with the control concrete mixture (presented in Figure 7).

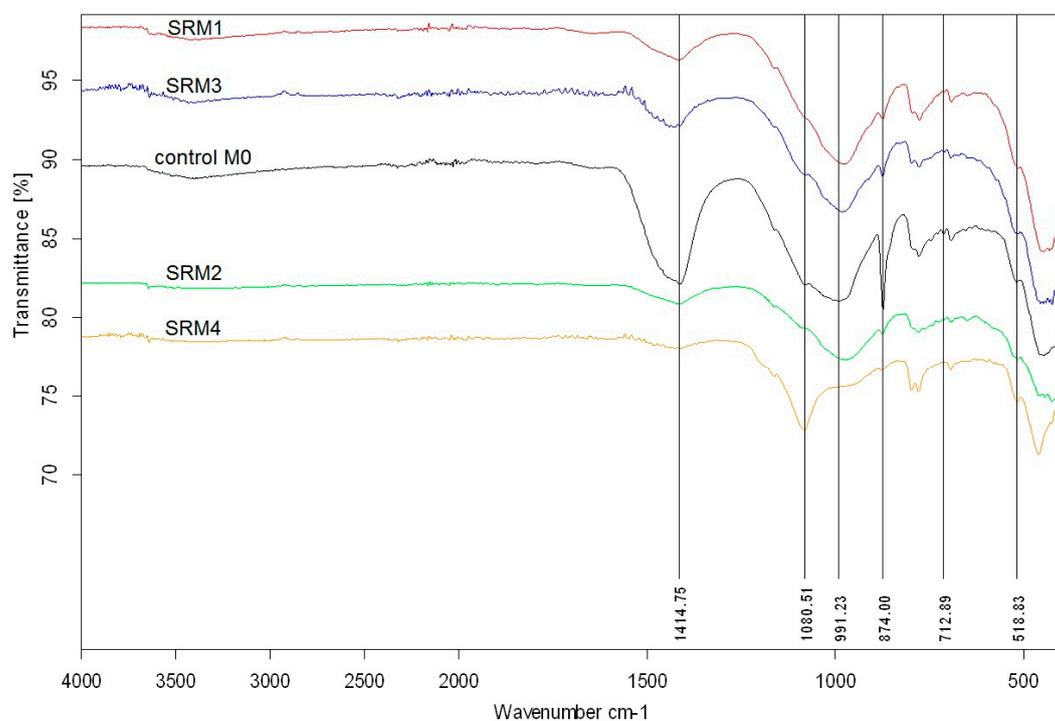


Figure 7. Fourier transform infrared spectroscopy (FTIR) spectra of prepared composites.

In the FTIR curves of the SRM1–SRM4 composites and control composite M0, the absorption bands of calcium carbonate, that are described by the stretching vibrations $\nu_3\text{-CO}_3^{-2}$ (1415 cm^{-1}), $\nu_2\text{-CO}_3^{-2}$ (874 cm^{-1}) and $\nu_4\text{-CO}_3^{-2}$ (712 cm^{-1}), were observed. The presence of CaCO_3 is attributed to the atmospheric CO_2 absorbed during the air hydration sampling [59]. However, the peaks at about 1415 cm^{-1} and 712 cm^{-1} in the FTIR curves of the SRM2 and SRM4 composites (prepared with NaOH) reflect a lower level of carbonation, causing the low compressive strength of these mixtures. Also, the bands at 1080 , 991 and 518 cm^{-1} corresponding to the calcium silicate phases due to stretching Si-O bond were observed.

A similar trend in the strengths' development was recorded in the flexural strengths. The lowest flexural strengths at 2 MPa reached the samples SRM2 and SRM4, with the NaOH as chemical activator. Composites SKM1 and SRM1 (without sediment modification) had strengths at a constant level of

about 4.5 MPa. The highest flexural strength was recorded in SRM3 sample (with biomass fly ash and without NaOH activator). These strengths (5.2–5.4 MPa) were at levels of the control mixture (about 5 MPa).

4. Conclusions

In this paper, we investigated a possible approach for reusing sediments dredged from two Slovak reservoirs, Klusov and Ruzin, as a 40% Portland cement replacement in building materials. Based on our results the following conclusions can be drawn: (1) Composites prepared from sediments without any chemical modifications achieved half strength values of the control concrete mixture; (2) Mechanical activation of sediments by their milling together with fly ash, for the purpose of introducing CaO content in the mixture, did not have significant impact on compressive strength; (3) Mixtures prepared with sediments milled without, and with addition of fly ash as cement replacement, satisfied the strength requirements for the compressive strength class C16/20 according to the European standard EN 206-1; and (4) Addition of NaOH into composites in the concentration of 5 M as an activator of sediment had a negative impact on compressive and flexural strengths and thus NaOH did not act as an effective pozzolanic activator for sediments.

Based on the results of the compressive and flexural strengths, the use of reservoir sediment as a replacement of Portland cement in concrete with a substitution ratio of 40% seems to be suitable without any further chemical modifications.

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Author Contributions: Natalia Junakova and Jozef Junak conceived and designed the experiments; Jozef Junak performed the experiments; Natalia Junakova and Jozef Junak analyzed the data; Natalia Junakova wrote and edited the paper. Both authors read and approved the final manuscript.

Conflicts of Interest: The authors declare no conflict of interest.

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