MICROSTRUCTURAL CHARACTERIZATION OF FINE RECYCLED AGGREGATES BY SEM-MLA

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Abstract

Cement paste from previous construction projects is responsible for increasing the porosity of recycled aggregates and lowering their mechanical properties. The characterization of these products is fundamental for guiding mineral processing, though there are few established procedures. The scope of this study involved the technological characterization of fine aggregates produced from construction and demolition waste with the intention of assessing the associations and liberation of the main phases. The experiment involved heavy liquid separations, chemical and mineralogical analyses and scanning electron microscopy mineral liberation analysis (SEM-MLA) image analysis. The results confirmed that recycled sand is composed of a high content of low-porosity particles (high density) with low attached cement paste content. The main phases are tectosilicates, carbonates, phyllosilicate, mafic minerals and cement paste. The method proved to be a valuable tool for assessing the cement paste content and phase liberation; the attained results are consistent with mineral separations.

Keywords: recycled fine aggregates, recycling, construction and demolition waste, image analysis

1 INTRODUCTION

Construction and demolition wastes represent a significant amount of total municipal solid waste (in many cases, more than 50%). Their composition is basically of mineral origin (around 90% of the weight) and liable to be recycled through mineral processing. Previous studies showed that about 50% of the weight of the product generated in the comminution of recycled aggregates corresponds to the fine fraction (particles below 4.8 mm) [1]. For a long time, the fine fraction has been disregarded or used as road base pavement, however recent studies have shown that the fine fraction properties are not that different from the coarse fractions and can be improved with the appropriate processing [2].

The main difficulty in the production of high quality recycled aggregates is related to the presence of cement paste from previous constructions still adhered to the aggregates. This is responsible for increasing the porosity of recycled aggregates and diminishing their mechanical properties, such as durability, elasticity and resistance to compression. Therefore, the characterization and quantification of this phase is essential for assessing the quality of the recycled aggregate through microstructural characterization, which is the focus of this study.

The characterization of the phase associations is fundamental for the processing of most raw materials in order to have a better understanding of the behaviour of the mineral of interest in mining operations and

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mineral processing as well as to measure the efficiency of separations processes and quality control on the final products [3, 4].

The phase associations can currently be carried out by automated image analysis that has surpassed semi-quantitative optical microscopy or SEM semi-automatic methods [5–9]. Alternatively, mineral separations also allow phase associations and liberations to be evaluated. The methods take into account the differences in the physical properties of phases, such as density and magnetic susceptibility [3, 5, 10–12].

Regarding recycled aggregates produced from construction and demolition waste, there are few studies on microstructural characterization and those there are focus essentially on the transition zone between aggregates and cement paste [13–16] or on the microfractures produced in aggregates after successive crushing stages [17].

Thus, the goal of this work is to establish a procedure for characterizing the association between the cement paste and the recycled aggregates aiming at determining the phase's liberation as a tool for evaluating the efficiency of the recycling process of a fine aggregate produced by tertiary crushing of construction and demolition waste.

2 MATERIALS AND METHODS

The study was carried out on recycled sand produced by tertiary crushing of construction and demolition waste obtained from "Urbem Tecnologia Ambiental", a recycling plant located in Sao Paulo metropolitan area. The recycled sand was produced according to the process described by Ulsen et al. [18].

The experimental procedure consisted of:

1. Sampling the recycled sand produced by tertiary crushing (<3 mm);
2. Determining the grain size distribution by wet sieving on screens with nominal apertures of 2.0, 1.2, 0.60, 0.30 and 0.15 mm, successively;
3. Evaluating the weight distribution in heavy liquid sequential separation products at 2.2, 2.5, 2.6 and 2.64 g/cm^3 densities (bromoform admixtures with ethyl alcohol);
4. Determining chemical composition of each product by X-ray fluorescence in fused beads (XRF; Axios Pro, PANalytical) and loss on ignition (LOI) at 1050°C;
5. Determining the mineral composition by X-ray powder diffraction (XRD) of each product with an X'Pert MPD diffractometer, PANalytical, with CuKα radiation (λ = 1.54178 Å);
6. Assessing the main phases of each product using scanning electron microscopy with automated image analysis (SEM-MLA) on polished sections with Quanta 600F, FEI together with a Quantax 4030 EDS system, Bruker and a MLA – mineral liberation analysis system from FEI.

3 RESULTS

3.1 Chemical and mineral modal compositions

The chemical composition and distribution of the main compounds in heavy liquid separation products are summarized in Table 1. The mineral modal compositions by XRD and representatives images from stereomicroscope are shown in Figures 1 and 2, respectively.

The chemical and mineralogical analyses demonstrated that the composition of each product is strictly related to the density of the separation. The mineralogical characterization by XRD indicates that the main phases are (a) tectosilicates (mainly quartz, albite, orthoclase and anorthite) from the granite, (b) carbonates (calcite and dolomite), (c) phyllosilicates (muscovite, biotite and kaolinite). Gibbsite and chrysotile were identified by XRD as possibly present but not certified due to their low grades. Calcite is attributed to both the previous aggregate and the cement paste.

3.2 Heavy liquid separations

The weight distribution demonstrated that just 5.8% of the weight (average for the total −3.0 + 0.15 mm) is below 2.2 g/cm^3; this product was composed basically of cement paste particles. The product concentrated in density between 2.2 and 2.5 represented 20% of the weight on average and was composed mainly of ceramics and mineral particles with a high content of attached cement paste. The high content of CaO+LOI in these products (39.5% and 24.5% from lighter to denser products) confirmed the presence of cimentitious materials.

At the same time, 70% of the weight was concentrated in product above 2.5, of which 21% was in product 2.5–2.6, 29% at 2.6–2.64 and 25% above 2.64. The product 2.5–2.6 was primarily characterized by high feldspar content as assayed by XRD and chemical analysis (increasing in the alkalis grades). In product 2.6–2.64, the increase of quartz proportion was notable. In contrast, the 2.64 product sink presented a totally different composition since it showed higher concentration of mafic minerals (micas, amphibole, pyroxene), heavy minerals and free particles of aggregate minerals (quartz and feldspar), as demonstrated by the increase in the SiO_2, Na_2O, K_2O grades. The increase in CaO+LOI content in this product did not represent an increase in cement paste content but the presence of another CaO phase with high LOI, like carbonates (in accordance with XRD) related to the previous aggregates.

The elemental distribution demonstrated that the product $d < 2.2$ recovered 17.5% of the total CaO+LOI content despite representing just 5.8% of the weight; for products below 2.5 g/cm^3 (25.4% wt), this distribution surpassed 50%. The removal of porous particles concentrated in float products at 2.5 enabled the achievement of a product with 76.4% weight, about 80% of the total content of silica, alumina, iron oxide, Na_2O+K_2O and just 46% of the total CaO+LOI. Due to the presence of carbonates in product $d > 2.64$, the distribution of cement paste may be overestimated; these results were revaluated based on the MLA analysis.

3.3 Quantitative image analysis by SEM-MLA

The grouped database established for the MLA assessment comprised the following phases: quartz, feldspar, cement paste, carbonates, mafic minerals (pyroxene + amphibole) and other phases. The phase distribution was assayed for each product and each size fraction as shown in Table 2; the total is summarized in Figures 3 and 4.

The main phases were quartz, feldspar and cement paste. Micas represented from 2% to 6% of weight and were basically related to heavy products (>2.64) such as the carbonates and other mafic minerals (pyroxene and amphibole). Minor minerals identified in heavy products were apatite, hematite, titanite and zircon.

On fractions above 1.2 mm, the content of cement paste in product $d < 2.2$ ranged between 30 and 40%, while the content of quartz + feldspar was about 50–60%. For the finest fractions, a progressive enrichment in cement paste content in the lighter product was noticed (from 51% to 81%), indicating a greater liberation of the cement paste from the aggregate phases towards the fine fractions. The content of quartz also increased for the finest fractions (from 35% to 64%) while the content of feldspar decreased (from 36% to 16%), demonstrating its preferential concentration at coarser fractions along with the mafic minerals (origin related to the coarse natural aggregates – granitic and/or gneissic rocks).

In accordance with the chemical and mineralogical results, the quantitative image analyses stated that density separation produced a heavy product ($d > 2.5$) with a low cement paste content and more than 90% of mineral aggregate phases (quartz, feldspar, mafic minerals and others). For products with densities of up to 2.64, there was a regular tendency for an increase in quartz content and a decrease in feldspar and cement paste; although in the product $d > 2.64$, there was a notable decrease in quartz and an increase in other phases, such as carbonates and mafic minerals; cement paste almost disappeared in this product.

The carbonates presence of 9% on average (from 7.8% to 11%) in the heaviest product explains the increase in CaO+LOI content assayed by XRF and XRD (calcitic aggregates).

Compositional images from SEM-MLA for selected products are shown in Figure 5 and clearly demonstrate the distinct composition of the products.

The quantification of the carbonates content is particularly important in the presence of calcareous aggregates, which are attributed to cement paste using the current methods for assessing cement paste content in recycled aggregates (HCl acid leaching [19] and CaO+LOI content [20]). Still, the partition of Ca in cement paste, carbonates and mafic minerals (Table 3) in the studied sample demonstrates that the higher the product density, the lower the proportion of Ca associated with the cement paste (from 95% to 14% on average) and the higher the association of carbonates (from 4.3% to 55%) and mafic minerals (25% on product $d > 2.64$). So both methods – acid leaching and CaO+LOI content – cannot be used to quantify the amount of cement paste when carbonates aggregates are identified or the cement paste will be overestimated.

The occurrence of phases with no cement paste attached (here called "mineral particles") into free or locked particles is shown in Figure 6. There was a progressive increase in the proportion mineral particles (free of cement paste) in denser products. While the products below 2.5 g/cm^3 represented up to 30% of free mineral particles, this proportion increased to around 90–95% in products sink at 2.6 g/cm^3.

4 CONCLUSIONS

The heavy liquid analysis demonstrates the potential density separation of the recycled fine aggregates produced by the total comminution of construction and demolition waste; the results encourage further mineral processing separation to obtain a fine recycled aggregate with less cement paste content.

The mineralogical characterization by XRD indicates that the main phases are tectosilicates (mainly quartz and feldspar), carbonates and phyllosilicates.

The weight distribution in heavy liquid separation products reveals that the amount of high porous phases with low strength ($d < 2.2$) is 5.8% on average. On the other hand the proportion of low porous phases ($d > 2.5$) is very high, exceeding 70% of the weight . The composition of products attained by heavy liquid separation is strictly related to the density of the separation; cement paste tends to concentrate in light products while quartz, feldspar, carbonates and mafic minerals are associated with denser products. The presence of carbonates in product $d > 2.64$ demonstrates that the determination of cement paste content by CaO+LOI overestimates this phase. The average partition of Ca revealed that just 77% is associated with the cement paste.

The elemental distribution demonstrates that product $d < 2.2$ recovers 17.5% of the total CaO+LOI content even though it represents only 5.8% of the weight; for products below 2.5 g/cm^3 (25.4% wt), this distribution surpasses 50%. The removal of porous particles concentrated in float products at 2.5 therefore enables the accomplishment of a product with 76% of the total weight, about 80% of the total content of silica, alumina, iron oxide, Na_2O+K_2O and just 46% of the total CaO+LOI. According to the quantitative image analyses, product $d > 2.5$ presents a low content of cement paste and a total content of mineral phases (quartz, feldspar, mafic minerals and others) >90%.

The occurrence of phases with no cement paste attached ("mineral particles") progressively increases in denser products. The products below 2.5 g/cm^3 present up to 30% of free cement paste particles while this proportion increases to around 90–95% in products sink at 2.6 g/cm^3.

The results of heavy liquid separations and image analysis are comparable; SEM-MLA image analysis can be used on recycled fine aggregates for the characterization of phase content and their main associations to establish the calcium distribution and to assess the liberation of the mineral phases from the cement paste.

5 REFERENCES

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TABLE 1: Chemical composition and distribution of the main compounds on heavy liquid separation products.

Total calc. –3.0 + 0.15 mm	Fraction (wt%)	Grades (%)									Distribution (%)				
		SiO_2	Al_2O_3	Fe_2O_3	Na_2O	K_2O	MgO	CaO	PF	CaO+PF	SiO_2	Al_2O_3	Fe_2O_3	CaO+LOI	Na_2O+K_2O
d < 2.2	5.8	44.2	7.36	2.89	2.32	0.42	1.22	23.3	16.2	39.5	3.7	5.2	5.0	17.5	2.5
2.2 < *d* < 2.5	19.6	59.2	7.08	2.59	1.61	0.58	2.26	14.1	10.4	24.5	17.0	17.1	15.3	36.9	14.7
2.5 < *d* < 2.6	20.7	70.4	9.90	2.49	0.63	1.34	4.45	4.51	3.75	8.26	21.3	25.2	15.4	13.1	31.6
2.6 < *d* > 2.64	28.7	85.1	5.19	2.22	1.30	1.28	0.23	1.82	1.40	3.22	35.7	18.3	19.1	7.08	19.5
d > 2.64	25.2	60.3	11.0	5.97	2.37	2.41	2.14	7.87	5.27	13.1	22.2	34.2	45.2	25.4	31.7
Total	100	68.4	8.14	3.34	1.19	1.39	2.41	7.55	5.48	13.0	100	100	100	100	100

TABLE 2: Composition of the sequential heavy liquid separation products (wt%).

Product	Fraction 3.0–2.0 mm					Fraction 2.0–1.2 mm					Fraction 1.2–0.6 mm					Fraction 0.6–0.3 mm					Fraction 0.3–0.15 mm				
	qz	fd	cp	carb	oth	qz	fd	cp	carb	oth	qz	fd	cp	carb	oth	qz	fd	cp	carb	oth	qz	fd	cp	carb	oth
d < 2.2	57	5.7	30	2.7	5.4	46	6.1	39	3.3	5.1	36	5.5	51	2.5	4.9	16	5.6	71	1.8	5.0	9.4	3.9	81	2.3	3.7
2.2 < *d* < 2.5	52	21	16	4.8	5.5	57	17	18	2.9	5.4	47	19	22	2.5	8.9	48	17	27	2.3	6.4	25	14	49	3.6	7.9
2.5 < *d* < 2.6	27	43	8.1	4.3	17	39	43	4.5	1.0	12	54	35	4.7	0.8	4.9	49	37	7.7	1.0	5.3	59	26	9.6	1.1	4.0
2.6 < *d* < 2.64	43	42	1.9	3.1	9.7	62	31	1.6	0.2	6	79	17	1.2	0.3	2.2	88	7.6	1.8	0.2	2.2	87	8.0	2.2	0.5	2.4
d > 2.64	23	44	1.2	7.8	24	32	34	1.7	11	22	43	25	1.4	8.7	22	45	17	1.8	9.7	26	51	11	2.1	8.1	27
Total	35	36	7.8	5.6	16	47	29	8.7	4.3	12	56	22	9.1	3.1	9.1	64	16	10	2.4	7.7	60	14	15	2.5	8.1

Note: qz – quartz, fd – feldspar, cp – cement paste, carb – carbonates, oth – other phases represented by ceramics + micas + pyroxene + amphibole + others.

TABLE 3: Partition of Ca among phases.

Products	3.0–2.0 mm			2.0–1.2 mm			1.2–0.6 mm			0.60–0.30 mm			0.30–0.15 mm			Total 3.0–0.15 mm		
	cp	carb	ma	cp	carb	ma	cp	carb	ma	cp	carb	ma	cp	carb	ma	cp	carb	ma
d < 2.2	94	5.7	–	96	3.6	–	97	3.1	–	94	5	–	95	4.5	–	95	4.3	–
2.2 < *d* < 2.5	86	14	–	90	8.9	–	92	6.9	–	90	7.1	–	92	7.4	–	90	8.6	–
2.5 < *d* < 2.6	73	24	–	82	12	4.8	89	10	–	85	6.9	7.4	76	12	12	82	13	–
2.6 < *d* < 2.64	36	46	12	65	8.3	23	73	13	12	98	1.7	–	98	1.9	–	76	13	–
d > 2.64	12	51	31	16	61	18	11	59	24	14	53	27	19	49	25	14	55	25
Total	60	27	11	71	22	5.8	75	17	5.6	81	12	5.6	93	5.2	1.7	77	16	5.7

Note: cp – cement paste, carb – carbonates, ma – mafic minerals: pyroxene+amphibole.

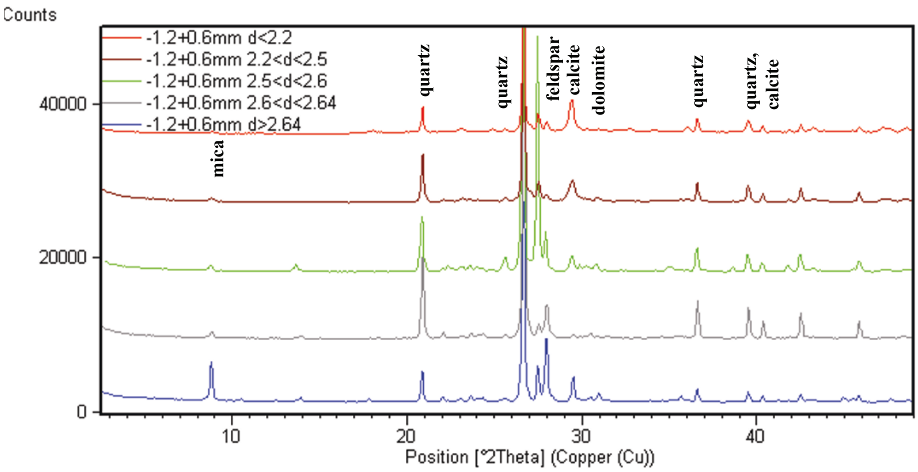
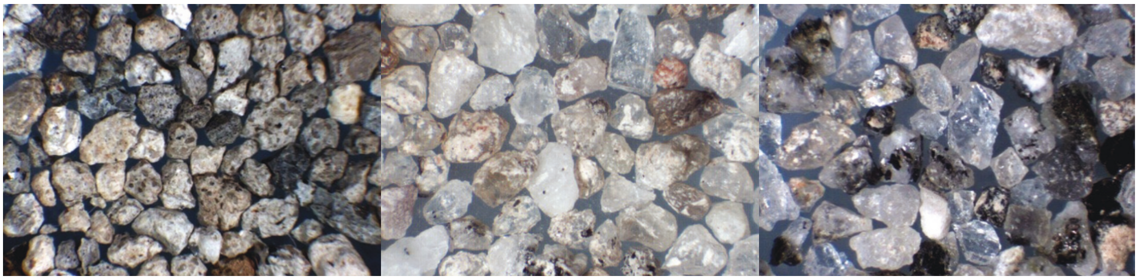


Figure 1: Selected characteristic XRD patterns from heavy liquid separation products.



(a) <2.2 g/cm^3 (b) 2.6<d<2.64 g/cm^3 (c) d>2.64 g/cm^3

Figure 2: Stereoscope images of heavy liquid separation products (fraction −1.2 + 0.60 mm).

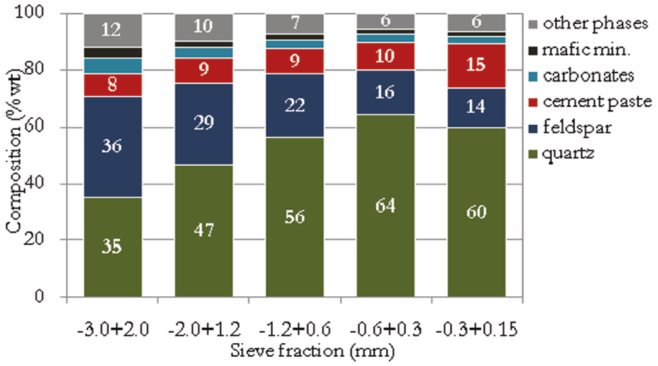


Figure 3: Phase distribution among different size fractions.

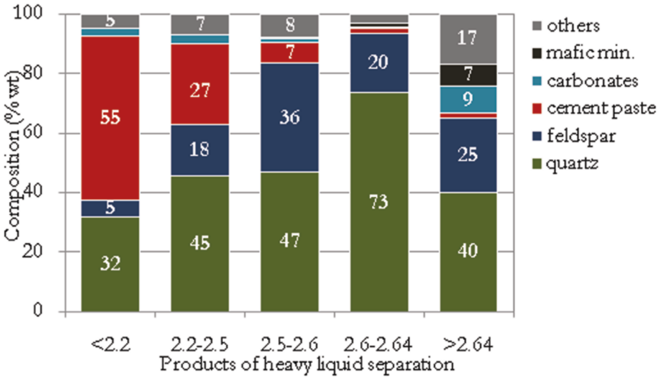
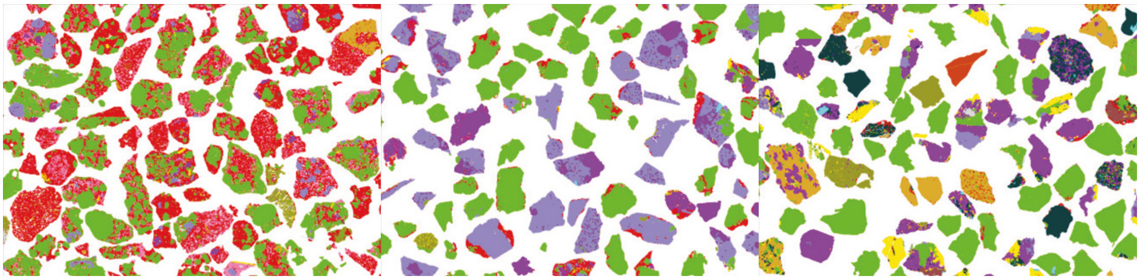


Figure 4: Phase distribution among heavy liquid separation products.



(a) <2.2 g/cm^3 (b) 2.5<d<2.6 g/cm^3 (c) d>2.64 g/cm^3

Main phases: red – cement paste, green – quartz, violet – feldspar, navy blue – mafic minerals, yellow – mica, orange – calcite, terracotta – dolomite

Figure 5: Comparison of products from heavy liquid separation.

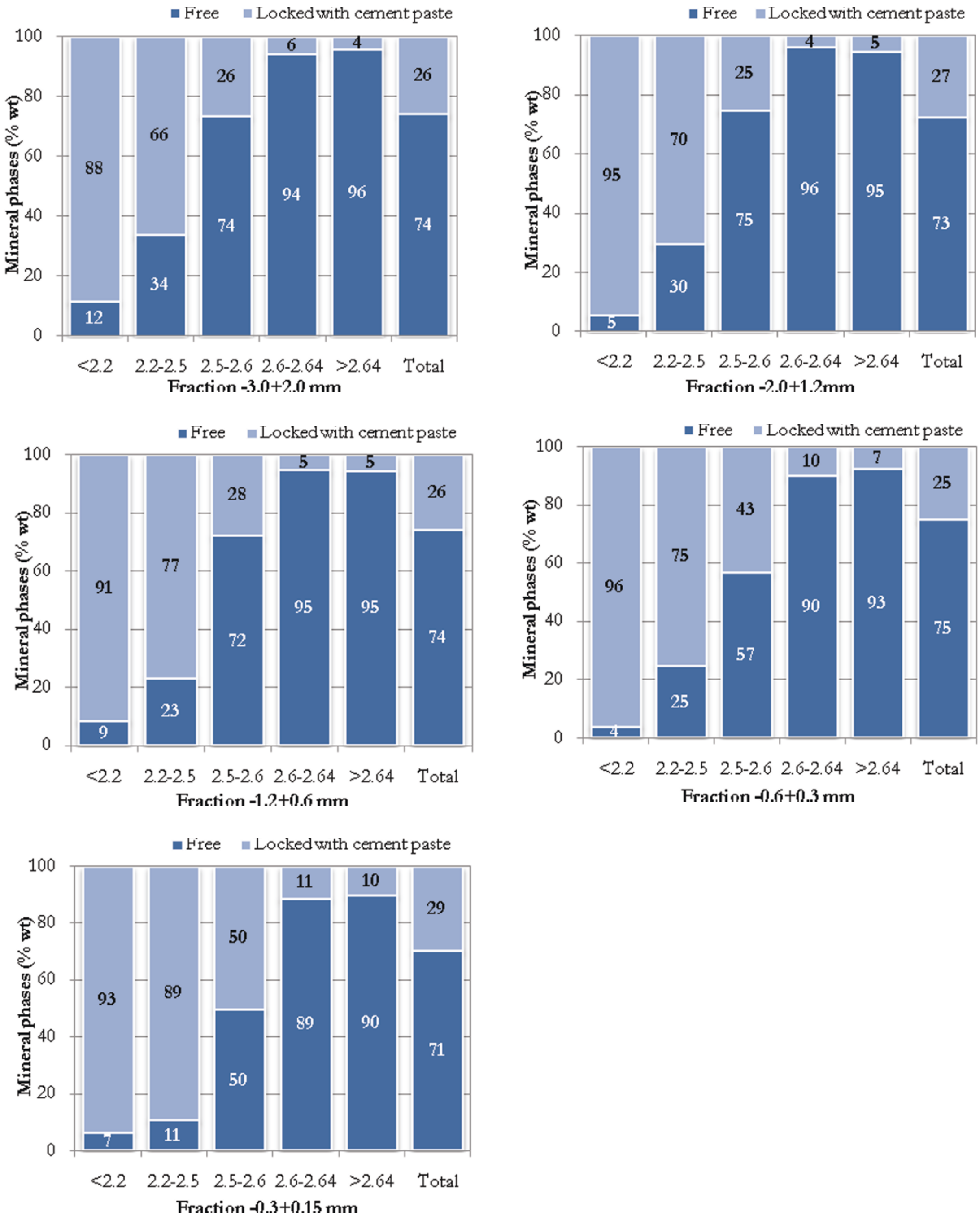


Figure 6: Association of mineral phases for decreasing size fractions.