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Volume reduction of the evaporator concentrate for incorporation in cement

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ABSTRACT

This study evaluates different routes of drying and cementation of Evaporator Concentrate (EC) from nuclear reactors. The Pressurized Water Reactor is one of the most used in nuclear power plants, and it uses boric acid as a coolant to control the nuclear fission. When the boric acid solution loses its characteristics, it is directed to an evaporation system, generating the EC. The cementation is used as a technique to immobilize this kind of waste, due to the low cost and ease of use of the cement, in addition to its good stability and mechanical strength. However, the EC is a liquid solution, which leads to a high volume of wastes. The inclusion of a drying step might be advantageous in providing a reduction of the EC volume to be solidified, consequently reducing the number of packages generated. The tests were performed at the Cementation Laboratory (LABCIM) at CDTN. The equipment chosen for the drying step was the lab-oven, because it is easy to use and allows a full drying at relatively low temperatures. The cementation was carried out with different amounts of dry EC incorporated and with various water/cement matrix (w/c) relations. As established by the norm CNEN NN 6.09, the compressive strength of homogeneous cementitious products, with 28 days of curing, must be greater than or equal to 10MPa for a safe disposal of the radioactive wastes. Formulations with w/c between 0.25 and 0.27 and incorporation of dry EC between 10% and 20% showed the best results so far.

Keywords: **evaporator concentrate, volume reduction, cementation.**

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1. INTRODUCTION

Despite the stigma around nuclear energy and radioactivity, this technology brings numerous benefits to our society, not only providing electricity, but also being applied in medicine, agriculture, industry, geochronology, etc. [1]. Moreover, Nuclear Power Plants (NPPs) don't release greenhouse gases into the atmosphere, therefore not contributing to global warming; they also occupy relatively small areas and use a low-cost material – Uranium – as fuel [2].

In 2016 there were 441 nuclear reactors in operation, and 64% of them were Pressurized Water Reactors (PWR) [3]. The Brazilian NPP Central Nuclear Álvaro Alberto also uses PWRs in its facilities. This kind of reactor uses a coolant system to control the nuclear fission and avoid uncontrolled chain reactions - this coolant solution contains, among other components, boric acid.

When the coolant loses its characteristics it is sent to an evaporation system along other liquid wastes, where their volume is reduced and the Evaporator Concentrate Waste is generated. The cement solidification is one of the main methods used in the treatment of this kind of waste due to the cement's high stability, resistance and low cost [4].

However, the cementation of the EC solution generates a high volume of cemented products. For this reason, drying the EC before the cementation step could be advantageous since it would reduce the number of packings generated and would also allow a greater incorporation of the waste on cement. The standard CNEN NN 6.09 determines that the cemented products must have a compressive strength greater than or equal to 10 MPa after 28 days of curing for a safe disposal of the cemented waste [5].

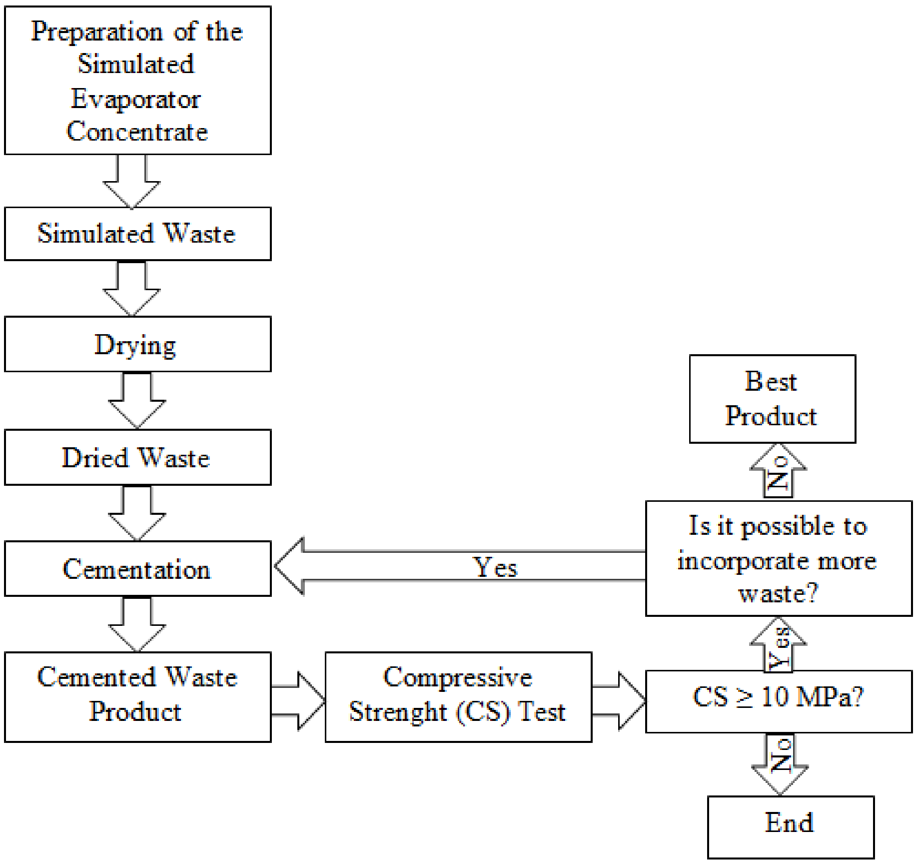
One of the compounds responsible for the hardening of the cement is called etringite, a calcium sulfoaluminate [6]. This is one of the major compounds formed during the cement hydration and is intimately related to its strength – therefore, the lack of etringite is associated to low Compressive Strength's results.

To fulfill these requirements, experiments were carried out to define the best route of drying and cementation of the EC waste, in order to maximize the amount of waste incorporated on cement while maintaining a high compressive strength value.

2. MATERIALS AND METHODS

The experiments were performed according to the flowchart presented below in Figure 1, adapted from Faria [2].

Figure 1: *Flowchart of the methodology followed in the experiments.*



2.1. Simulated evaporator concentrate

To carry out the experiments, a simulated waste was prepared in order to reproduce the real evaporator concentrate waste produced in PWR reactors. The chemical composition of the simulated EC, as determined by ELETRONUCLEAR, is showed in Table 1.

Table 1: Chemical composition of the simulated EC.

Reagents	Quantity (g)
$MgCl_2$	0.082
SiO_2	0.089
NaCl	0.374
Al_2O_3	0.800
$Ca(NO_3)_2$	1.591
NaOH	1.570
H_3BO_3	369.0

These reagents were added to a beaker containing 2.75L of water at 80°C to avoid boron precipitation, and this solution was stirred until their complete solubilization.

2.2. Drying of the simulated waste

As showed in Figure 2, the method chosen to dry the EC solution was evaporation using a lab oven, as this equipment is easy to use and to control. The liquid waste was dried for approximately 2 weeks at 110°C. After the wastes were completely dried, their humidity was measured.

Figure 2: *Sample of EC solution drying in the lab oven.*

2.3. Cementation and compressive strength test

After drying the simulated EC, the next step was the cementation. The w/c ratio, defined in Equation 1, is an important parameter and must be controlled to obtain a good cemented product.

$$\frac{w}{c} = \frac{\text{water (g)}}{\text{cement (g)}} \tag{1}$$

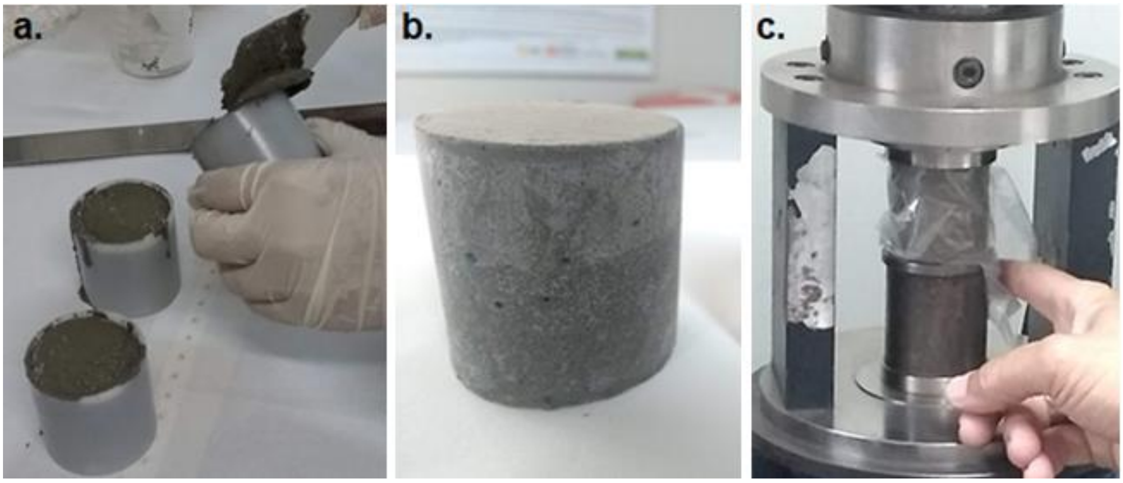
The experiments were performed with varied amounts of dried waste and a w/c ratio ranging from 0.25 to 0.33. The process consisted of mixing the determined amount of dried EC with a neutralizer (NaOH) and water for 30s to increase the pH to approximately 9. After that, the cementitious matrix – CP-V Portland cement, active silica and Calcium Hydroxide – was added to the mixture and stirred for 1 minute. At this point, fluidizer was added to some of the mix if needed, and the paste was homogenized for 5min. The formulation of the experiments with the best results are shown in Table 2.

Table 2: Formulations used to cement the dried evaporator concentrate.

Experiment	a/c	Dried EC Incorporated (%)	Fluidizer
001	0,25	10	Yes
002	0,27	10	Yes
003	0,33	10	No
004	0,33	13	No
005	0,27	15	Yes
006	0,26	17	No
007	0,26	20	No

The paste was then put into cylindrical molds of 5cm x 5cm and the samples were reserved for 7, 14 or 28 days. After the curing time, the specimens were demolded, sanded and submitted to the compressive strength (CS) test (Figure 3).

Figure 3: *Cemented EC being put in cylindrical molds (a), demolded sample (b) and a sample during the compressive strength test (c).*



To deeply evaluate the presence of ettringite, samples were analyzed after the CS test with a Scanning Electron Microscope (SEM). The ettringite is easily recognized by its needle-like structure.

3. RESULTS AND DISCUSSION

After the simulated EC was dried, the product is a white powder, resembling kitchen salt (Figure 4). The dried waste had an average humidity of 26.39%.

Figure 4: *Simulated EC dried in a lab oven.*

The CS results (Table 3) show that none of the experiments achieved a compressive strength value above the limit established by CNEN NN 6.09. However, the experiments 007-1, 007-2 and 007-3 showed good potential when compared to results obtained previously [2].

Table 3: Compressive strength results of the cemented products.

Experiment	Curing Time (days)	CS (MPa)
001-1	7	1.35
002-1	7	0.93
003-1	7	0.48
003-2	28	1.18
004-1	7	0.30
004-2	28	1.65
005-1	7	1.49
005-2	28	2.20
006-1	7	1.29
006-2	28	1.53
007-1	7	4.79
007-2	14	4.91
007-3	28	5.76

Analyzing Figure 5, it is possible to notice that good CS results were achieved with higher incorporation of EC. It is also possible to conclude that, in general, there isn’t a great improvement in CS value when comparing curing of 7 and 28 days for the same experiment. This might happen because the cement used provides high early strength, not improving much with more curing days.

Figure 5: *Relation between the percentage of EC incorporated, the curing time and the compressive strength for each experiment.*

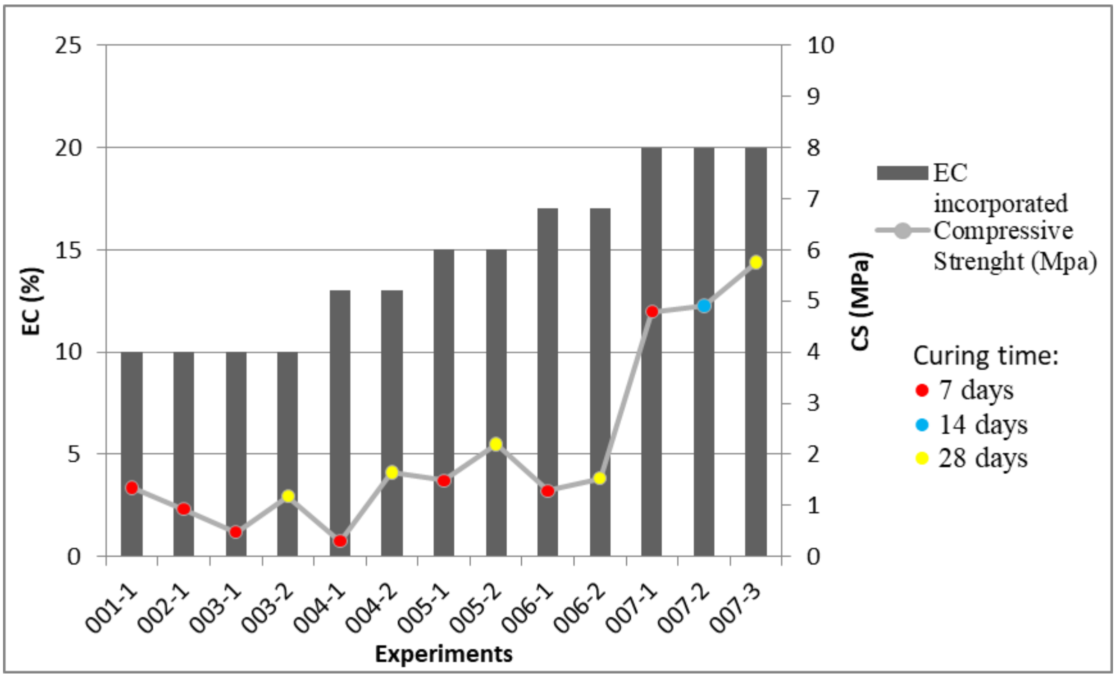
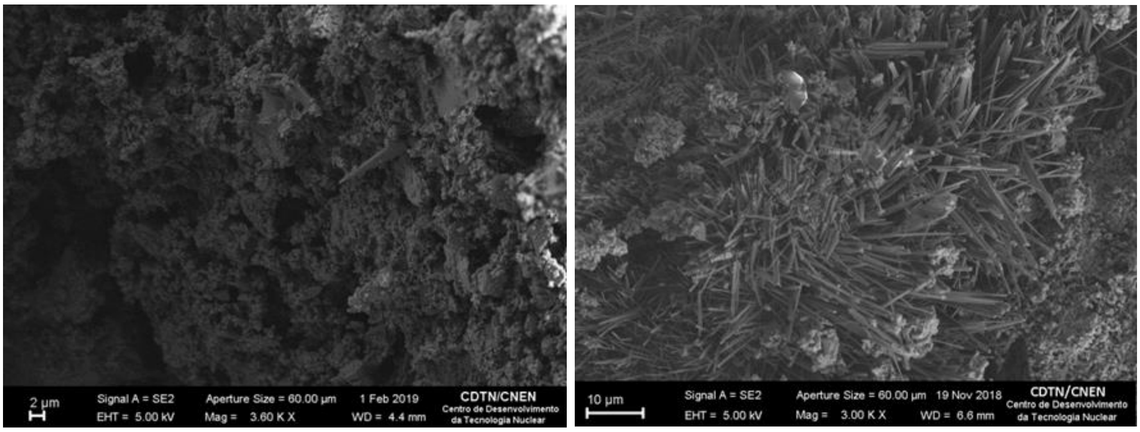


Figure 6 shows the results for the SEM analysis of the samples with higher and lower CS values. It is possible to notice that the sample with the best CS results (007-3) have a lot of ettringite, while the one with lower compressive strength didn't have any trace of it. Since ettringite is produced during the hardening of the cement, this result shows that this step must be better controlled in future studies.

Figure 6: *SEM results of each experiment 004-1 (left) and experiment 007-3 (right).*



4. CONCLUSION

The highest compressive strength achieved with these experiments was 5.76 MPa with 20% of dried EC incorporated, not complying with the minimum of 10 MPa required by CNEN NN 6.09. Previous works were able to obtain a compressive strength of 31.69 MPa with 5% of dried EC incorporated, while the maximum amount of liquid EC incorporated in cement was 3% [2]. This shows that the drying step is advantageous and that it is possible to incorporate a greater amount of EC in cement. Further studies are being held to adjust the formulation and the cementation route, in order to achieve the required minimum compressive strength value.

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