Combining lime softening with alum coagulation for hard Ghrib dam water conventional treatment

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ABSTRACT

This work aims to control the efficiency of water softening as an inherent phenomenon in the coagulation process or, in other words, to evaluate the softening process as a secondary reaction which is producing simultaneously with the main reaction as the coagulation process. Ghrib Dam water is well-known for its high hardness ranging from 750 to 900 mg/L as $CaCO_3$. That is, this water is unpleasant to the domestic consumption. Conventional water treatment at the Ghrib Station is based on coagulation using aluminum sulfate $[Al_2 (SO_4)_3.18H_2O]$ (alum) as a single coagulant. Alum has a minimal effect on the total hardness and its human toxicity is not yet doubtful. This research introduces the concept of the replacement of alum by lime and sodium hydroxide (NaOH) in coagulation process at the Ghrib Station. Coagulation experiments on jar test using the three reagents (alum, lime, and NaOH) are performed and physicochemical analyses are conducted to evaluate the possibility of alum partial or total replacement for improving the treatment effectiveness in hardness reducing. The obtained results show that hardness is decreased at its half by combining simultaneously the three chemical products: alum = 15, lime = 100, NaOH = 100 mg/L. Additional survey is required to examine the complicated interaction in the Ca^{2+}/Mg^{2+}-DOM-Al ternary system to comprehensively define the contributions of the two mechanisms – lime softening and coagulation - to organic matter removal by coagulation.

1. Introduction

In a general manner, water is classified, according to its hardness, into four categories: soft (or freshwater), moderately hard, hard or very hard: 0-60, 60-120, 120-180, 180 mg/L as $CaCO_3$ and above, respectively (Ghernaout et al., 2017; Van der Bruggen et al., 2009; Rodier, 2009; Yildiz et al., 2003). Although hardness cannot have on water aesthetic or sensory effects, it is traditionally used to measure the power of reaction of water with soap, where hard water requires a considerable amount of

soap to produce foam (Anim-Mensah et al., 2008). On the other hand, total hardness (TH) causes scaling of hot water pipes, boilers and appliances (Rodier, 2009). A maximum acceptable concentration is not set due to the public tolerance towards hardness and can vary considerably depending on local conditions (Sivasankar and Ramachandramoorthy, 2011). Water with hardness content greater than 200 mg/L $CaCO_3$ is considered *"mediocre"* even if it is tolerated by consumers; waters with hardness greater than 500 mg/L $CaCO_3$ are unacceptable for the most domestic purposes (Rodier, 2009). Water hardness is due to dissolved polyvalent metal ions (Godskesen et al., 2012). In freshwater, the major ions responsible for hardness are calcium and magnesium ions; strontium, iron, barium and manganese ions also contribute (Ghizellaoui et al., 2005). Hardness can be measured by the reaction of

polyvalent metal ions in a water sample with a chelating agent such as ethylenediaminetetraacetic acid and is expressed as the equivalent concentration of calcium carbonate ($CaCO_3$) (Rodier, 2009; Gabrielli et al., 2006). Water hardness may be evaluated by determining separately the concentration of each element of the hardness and expressing their sum as an equivalent amount of $CaCO_3$ (Ordóñez et al., 2012; Fang et al., 2013; Nanda et al., 2010).

Although hardness is caused by cations, we often speak of carbonate hardness (temporary) and non-carbonate hardness (permanent) (Fang et al., 2013; Lee and Lee, 2000). The carbonate hardness concerns the amount of carbonates and bicarbonates that can be removed or precipitated by boiling. This type of hardness is responsible for the scaling of hot water pipes and boilers. Non-carbonate hardness is due to the association of cations responsible for the hardness with sulfates, chlorides and nitrates. It is also called *"permanent hardness"* because it cannot be removed by boiling. Alkalinity, an indicator of the water buffer capacity, is closely related to the hardness. Alkalinity is constituted mainly by molecular species or anions of weak acids, especially the hydroxide, carbonate and bicarbonate. Other species such as borates, phosphates, silicates, and organic acids, may also contribute to some extent. Although many species of solutes can contribute to alkalinity, alkalinity is expressed as an equivalent amount of $CaCO_3$ (Van der Bruggen et al., 2009; Nanda et al., 2010; Lee et al., 2013; Nason and Lawler, 2009).

In conventional water treatment industry, especially on a municipal level, frequently employed softening process is chemical precipitation (Yildiz et al., 2003; Ghernaout and Naceur, 2011; Ghernaout and Ghernaout, 2012; Ghernaout, 2013; 2017a; Brastad and He, 2013). In the chemical precipitation, lime (lime softening) and soda or caustic soda is added to hard water (Wesolowska et al., 2004; O'Donnell et al., 2016; Comstock et al., 2011). Increasing the pH of water by introducing chemical additives (alkalinity) transforms bicarbonates into carbonates. Ca^{2+} and Mg^{2+} are eliminated from water in the form of $CaCO_3$ and $Mg(OH)_2$. Because of the restricted solubility of $CaCO_3$ and $Mg(OH)_2$, during chemical precipitation process, zero hardness water cannot be achieved (Yildiz et al., 2003). On the other hand, classical softening plants performing soft water treatment comprise several unit operations like coagulation/flocculation (Ordóñez et al., 2012; Ghernaout et al., 2009, 2010, 2011; Cheng et al., 2017; Yan et al., 2009), sedimentation, re-carbonation and filtration (Wang et al., 2005). Sedimentation and filtration units are employed for separating particulate matters like $CaCO_3$ and $Mg(OH)_2$ (Yildiz et al., 2003; Comstock et al., 2011; Vahedi and Gorczyca, 2011). Lime softening is also recognized by the U.S. Environmental Protection Agency (EPA) as the best applied technique for arsenic, barium, beryllium, chromium, copper, fluoride, lead, mercury, cadmium, nickel, and radionuclides (Brastad and He, 2013; Kweon and Lawler, 2004). Nevertheless, the disadvantages of lime softening comprise the formation of a high-volume lime sludge stream (Brastad and He, 2013), and the required use of chemicals such as quick lime, coagulants (iron or aluminum based), soda ash, and acids for adjusting the pH (Ordóñez et al., 2012; Brastad and He, 2013).

On the other hand, Aluminum has been proven to have a causal action in dialysis encephalopathy and epidemiological studies propose a probable relationship between exposure to this toxic metal and a higher prevalence of Alzheimer's disease. This link is dependent on the duration of Al exposure and only becomes important if an individual has resided in an area with high Al in drinking water (> 0.1 mg/L) for many years (Ghernaout et al., 2011; Becaria et al., 2006; Walton, 2007; Flaten, 2001). Consequently, substituting Al chemicals or at least reducing their injection into water should constitute a main focus for water treatment specialists.

The objective of this work is to substitute aluminum sulfate (alum) as a coagulant in the Ghrib Station by other reagents, less toxic, that can play two roles simultaneously in both *coagulation* and *softening* processes. Coagulation is a process that aims to increase the tendency of the colloids in a suspension to attach to each other. Coagulation is also used to make the removal of some soluble substances by adsorption and precipitation. On the other hand, water softening is conceived for reducing calcium and magnesium concentrations.

Jar test experiments, using multiple doses and combinations of the three reagents alum, lime and NaOH, are made to the Ghrib Dam (GD) water during the two months of this study, March and April 2013. Physicochemical analyses are also performed to control the efficiency of water softening as an inherent phenomenon in the coagulation process, or in other words, to evaluate the softening process as a secondary reaction which is producing simultaneously with the main reaction as the coagulation process.

2. Materials and methods

Jar test experiments and physicochemical analyses are performed at the Central Laboratory of the Algerian Waters Company (Unity of Medea). Raw water samples are taken directly from GD. Physicochemical analyses and jar test experiments are performed immediately while respecting the conditions of transport and storage to the Laboratory. Jar test experiments are conducted in order to assess the softening effectiveness of lime and NaOH, with or without alum, during coagulation of GD water. The evaluation of the softening phenomena induced during coagulation process is done by calculating the reduction of the TH (%). TH (%) is, therefore, considered as an indicator of the coagulation/softening effectiveness:

$$TH(\%) = \frac{TH_i - TH_f}{TH_i} \times 100 \qquad (1)$$

where TH_i and TH_f mean initial and final hardness, respectively.

Other parameters indicating the treatment effectiveness are also controlled such as turbidity, calcium, magnesium, conductivity. Further, an increase in sodium content of the coagulated water is also considered as an indicator to confirm the coagulation/softening efficiency.

The doses of the reagents are designated by (alum, lime, NaOH) in mg/L; as an example: For (15-100-100), we read 15, 100, and 100 mg/L for alum, lime and NaOH, respectively. All reagents used are of analytical grade. Table 1 presents coagulation/softening reagents and their properties. The coagulation tests are done using G. Vittadini jar test which can carry six beakers. All physicochemical analyses are made according to the standard methods (Rodier, 2009).

Table 1: Coagulation/softening reagents and their properties

Reagent	Chemical formula	Molar mass (g/mol)	Solubility at 20°C (g/L)	Prepared concentration (g/L)	Concentration per mL of reagent (mg/mL)
Aluminum sulfate	$Al_2(SO_4)_3.18H_2O$	666	NA*	10	10
Lime	$Ca(OH)_2$	74	1.73	1.5	1.5
Sodium hydroxide	NaOH	40	1090	4	4

*NA: not available

Jar test procedure is as follows: strong mixing (180 rpm/min) during 3 min, weak mixing (40 rpm/min) during 17 min, and settling period of 30 to 60 min. Strong mixing is applied and reagents doses are injected in the water sample separately and consecutively, and the coagulation period (3 min) is allowed to get reagents dispersed and colloids destabilized. The flocculation period (17 min) is accorded to give the chance to the flocs to be formed.

3. Results and discussion

During this study, 9 samples (B1 to B9) are taken from GD water and analyzed. To assess the coagulation/softening efficiency, several jar tests experiments are performed using the three reagents (alum, $Ca(OH)_2$, NaOH). Table 2 presents all the initial and final results of GD water treated by coagulation/softening using the three chemical products. Several combinations are adopted: one compound alone, two compounds and three compounds at the same time.

3.1. Effect of alum, lime and NaOH taken separately

Coagulant injection (alum) at different doses, from 20 to 100 mg/L, is performed in E1 to E5 beakers, respectively. As seen in Table 2, a weak TH removal is oscillating between 9.7 to 12.6%.

Lime injection at different doses, from 10 to 50 mg/L, is realized in D1 to D5 beakers, respectively. As seen in Table 2, a weak TH removal is oscillating between 5.7 to 13.7%.

Sodium hydroxide (NaOH) injection at different doses, from 8 to 40 mg/L, is accomplished in F1 to F5 beakers, respectively. As seen in Table 2, a low TH removal is oscillating between 5.2 to 7.2%. For 8 mg/L NaOH, TH instead increased slightly from 765 to 785 mg/L.

Table 2: Initial and final results of GD water treated by coagulation using three reagents (alum, $Ca(OH)_2$, NaOH)

Beaker number	Alum (mg/L)	$Ca(OH)_2$ (mg/L)	NaOH (mg/L)	pH	Hardness (mg/L as $CaCO_3$)	Ca^{2+} (mg/L as $CaCO_3$)	Mg^{2+} (mg/L as $CaCO_3$)	Total alkalinity (mg/L as $CaCO_3$)	Na^+ (mg/L)	Cl^- (mg/L)	Conductivity (μS/cm at 25°C)	SO_4^{2-} (mg/L)	$KMnO_4$ oxidability (mg/L)	K^+ (mg/L)	Turbidity (NTU)
Initial B1				7.02	875	520	355	134	625	426	2290	1429	1.9	8.3	6.0
D1	0	10 [13.5]****	0	8.97	825 (5.7%)*	490 (5.8%)**	335 (5.6%)***	167	650	461	2140	1354	1.6 (15.8%)*****	8.5	9.2 (-53.3%)******
D2	0	20 [27]	0	8.12	825 (5.7%)	475 (8.6%)	350 (1.4%)	128	675	447	2130	1463	1.7 (10.5%)	8.4	24.7 (-311.7%)
D3	0	30 [40.5]	0	8.18	825 (5.7%)	485 (6.7%)	340 (4.2%)	120	675	447	2110	1470	1.3 (31.6%)	8.5	52.8 (-780.0%)
D4	0	40 [54]	0	8.12	785 (10.3%)	495 (4.8%)	290 (18.3%)	100	650	433	2080	1420	1.2 (36.8%)	8.5	76.4 (-1173.3%)
D5	0	50 [67.5]	0	8.13	755 (13.7%)	475 (8.6%)	280 (21.1%)	83	650	440	2080	1397	1.8 (5.3%)	8.4	88.3 (1371.7%)
E1	20 [8.8]****	0	0	6.81	790 (9.7%)	460 (11.5%)	330 (7.0%)	110	650	461	2330	1375	0.9 (52.6%)	8.3	0.6 (90.0%)
E2	40 [17.6]	0	0	6.79	765 (12.6%)	450 (13.5%)	315 (11.3%)	105	675	447	2320	1428	1.2 (36.8%)	8.4	0.4 (93.3%)
E3	60 [26.4]	0	0	6.70	775 (11.4%)	460 (11.5%)	315 (11.3%)	100	625	440	2340	1347	0.1 (94.7%)	8.6	0.4 (93.3%)
E4	80 [35.2]	0	0	6.59	780 (10.9%)	460 (11.5%)	320 (9.9%)	91	650	440	2330	1413	1.1 (42.1%)	8.4	0.4 (93.3%)
E5	100 [44]	0	0	6.48	765 (12.6%)	450 (13.5%)	315 (11.3%)	72	625	454	2310	1345	0.6 (68.4%)	8.3	0.6 (90.0%)
Initial B2				8.20	765	455	310	133	650	454	2390	1339	2.5	8.4	9.1
F1	0	0	8 [10]****	8.88	785 (-2.6%)	460 (-1.1%)	325 (-4.8%)	145	700	454	2370	1451	1.4 (44.0%)	8.3	4.6 (49.4%)
F2	0	0	16 [20]	8.99	725 (5.2%)	450 (1.1%)	275 (11.3%)	122	700	426	2320	1454	1.5 (40.0%)	8.3	3.4 (62.6%)
F3	0	0	24 [30]	8.83	720 (5.9%)	420 (7.7%)	300 (3.2%)	110	700	440	2340	1442	1.5 (40.0%)	8.3	4.7 (48.3%)
F4	0	0	32 [40]	8.77	715 (6.5%)	390 (14.3%)	325 (-4.8%)	100	700	454	2300	1452	1.6 (36.0%)	8.2	4.9 (46.1%)

F5	0	0	40 [50]	8.79	710 (7.2%)	350 (23.1%)	360 (-16.1%)	99	675	454	2310	1370	1.2 (52.0%)	8.2	4.1 (54.9%)
G1	60 [26.4]	6 [8.1]	0	6.72	750 (2.0%)	475 (-4.4%)	275 (11.3%)	118	600	398	2370	1312	1.5 (40.0%)	8.4	0.4 (95.6%)
G2	60 [26.4]	12 [16.2]	0	6.99	755 (1.3%)	485 (-6.6%)	270 (12.9%)	120	625	398	2340	1367	1.2 (52.0%)	8.4	0.9 (90.1%)
G3	60 [26.4]	18 [24.3]	0	7.31	765 (0.0%)	480 (-5.5%)	285 (-8.1%)	121	700	341	2320	1609	1.4 (44.0%)	8.4	0.7 (92.3%)
G4	60 [26.4]	20 [27]	0	7.65	775 (-1.3%)	450 (1.1%)	325 (-4.8%)	127	700	355	2330	1618	1.3 (48.0%)	8.3	0.7 (92.3%)
G5	60 [26.4]	30 [40.5]	0	7.95	805 (-5.2%)	430 (5.5%)	375 (-21.0%)	132	675	355	2320	1563	1.8 (28.0%)	8.3	0.6 (93.4%)
Initial B3				7.89	768	417	350	137	675	412	2340	1447	2.4	8.2	4.5
H1	0	15 [20.2]	8 [10]	8.30	735 (4.2%)	415 (0.5%)	320 (8.6%)	115	870	454	2200	1786	1.9 (-20.8%)	8.4	11.5 (-155.6%)
H2	0	15 [20.2]	16 [20]	7.85	775 (-1.0%)	410 (1.7%)	365 (-4.3%)	96	900	469	2190	1885	0.9 (62.5%)	8.4	12.9 (-186.7%)
H3	0	15 [20.2]	24 [30]	7.86	700 (8.8%)	400 (4.1%)	300 (14.3%)	92	870	412	2180	1833	1.5 (37.5%)	8.4	14.3 (-217.8%)
H4	0	15 [20.2]	32 [40]	7.81	700 (8.8%)	370 (11.3%)	330 (5.7%)	81	840	461	2160	1713	1.3 (45.8%)	8.4	15.4 (-242.2%)
H5	0	15 [20.2]	40 [50]	7.81	705 (8.1%)	375 (10.1%)	330 (5.7%)	74	840	469	2150	1714	1.2 (50.0%)	8.4	17.9 (-297.8%)
I1	60 [26.4]	0	8 [10]	7.10	800 (-4.2%)	490 (-17.5%)	310 (11.4%)	119	650	426	2330	1424	2.1 (12.5%)	8.2	0.4 (91.1%)
I2	60 [26.4]	0	16 [20]	7.40	740 (3.6%)	435 (-4.3%)	305 (12.9%)	130	650	426	2340	1356	1.9 (20.8%)	8.2	0.4 (91.1%)
I3	60 [26.4]	0	24 [30]	8.10	725 (5.5%)	440 (-5.5%)	285 (18.6%)	146	675	412	2370	1398	1.6 (33.3%)	8.2	0.3 (93.3%)
I4	60 [26.4]	0	30 [37.5]	8.40	760 (-1.0%)	445 (-6.7%)	315 (-10.0%)	149	650	412	2320	1376	1.5 (37.5%)	8.1	0.3 (93.3%)
I5	60 [26.4]	0	40 [50]	8.67	695 (9.5%)	455 (-9.1%)	240 (31.4%)	160	650	412	2340	1304	1.3 (45.8%)	8.1	0.3 (93.3%)
Initial B4				7.78	765	465	300	131	675	433	2240	1422	2.8	8.3	3.9
J1	10 [4.4]	15 [20.2]	20 [25]	8.22	775 (-1.3%)	440 (5.4%)	335 (-11.7%)	135	775	383	2320	1703	2.3 (21.7%)	8.6	2.8 (28.2%)
J2	20 [8.8]	15 [20.2]	20 [25]	7.97	775 (-1.3%)	415 (10.8%)	360 (-20.0%)	137	800	398	2330	1768	2.3 (21.7%)	8.7	2.1 (46.1%)
J3	30 [13.2]	15 [20.2]	20 [25]	8.25	775 (-1.3%)	430 (7.5%)	345 (-15.0%)	137	840	419	2340	1789	2.4 (14.3%)	8.8	2.8 (28.2%)
J4	40 [17.6]	15 [20.2]	20 [25]	8.12	735 (3.9%)	400 (14.0%)	335 (-11.7%)	148	800	405	2350	1676	2.0 (28.6%)	8.7	3.3 (15.4%)
J5	50 [22]	15 [20.2]	20 [25]	8.22	825 (-7.8%)	440 (5.4%)	385 (-28.3%)	149	800	369	2360	1809	1.9 (32.1%)	8.7	2.9 (25.6%)
K1	40 [17.6]	30 [40.5]	12 [15]	7.85	780 (-2.0%)	460 (1.1%)	320 (-6.7%)	143	780	398	2650	1691	2.2 (21.4%)	8.4	0.3 (92.3%)
K2	40 [17.6]	30 [40.5]	24 [30]	8.13	750 (-2.0%)	390 (16.1%)	360 (-20.0%)	110	810	405	2620	1747	2.1 (25.0%)	8.5	0.6 (84.6%)
K3	40 [17.6]	30 [40.5]	36 [45]	8.19	730 (-4.6%)	380 (18.3%)	350 (-16.7%)	100	810	412	2600	1728	1.5 (46.4%)	8.4	0.7 (82.0%)
K4	40 [17.6]	30 [40.5]	48 [60]	8.36	650 (15%)	350 (24.7%)	300 (0.0%)	95	810	376	2590	1705	1.4 (50.0%)	8.4	3.0 (23.1%)
K5	40 [17.6]	30 [40.5]	60 [75]	8.39	625 (18.3%)	290 (37.6%)	335 (-11.7%)	75	840	391	2570	1742	1.3 (53.6%)	8.4	3.3 (15.4%)
Initial B5				7.50	710	480	230	135	750	447	2620	1504	2.2	8.2	2.5
L1	40 [17.6]	38 [51.3]	12 [15]	7.56	750 (-5.6%)	405 (15.6%)	345 (-50.0%)	102	725	362	2600	1634	1.2 (45.4%)	7.9	0.4 (84.0%)
L2	40 [17.6]	38 [51.3]	24 [30]	7.64	750 (-5.6%)	375 (21.9%)	375 (-63.0%)	91	780	405	2570	1702	1.1 (50.0%)	7.9	0.5 (80.0%)
L3	40 [17.6]	38 [51.3]	4 [5]	7.74	720 (-1.4%)	340 (29.2%)	380 (-65.2%)	65	810	369	2540	1808	1.0 (54.5%)	7.8	0.4 (84.0%)
L4	40 [17.6]	38 [51.3]	48 [60]	7.86	680 (4.2%)	340 (29.2%)	340 (-47.8%)	61	840	412	2540	1779	0.9 (59.1%)	7.7	0.3 (88.0%)
L5	40 [17.6]	38 [51.3]	60 [75]	8.20	615 (13.4%)	336 (30.0%)	280 (-21.7%)	51	840	391	2510	1756	0.8 (63.6%)	7.7	0.5 (80.0%)
M1	40 [17.6]	45 [60.8]	12 [15]	8.40	750 (-5.6%)	450 (6.2%)	300 (-30.4%)	127	675	327	2670	1555	2.0 (9.1%)	8.1	4.2 (-68.0%)
M2	40 [17.6]	45 [60.8]	24 [30]	8.37	715 (-0.7%)	390 (18.8%)	325 (-41.3%)	120	700	355	2650	1541	1.7 (22.7%)	8.1	4.8 (-92.0%)
M3	40 [17.6]	45 [60.8]	36 [45]	8.35	730 (-2.8%)	360 (25.0%)	370 (-60.1%)	98	675	355	2620	1524	1.8 (18.2%)	8.1	4.8 (-92.0%)
M4	40 [17.6]	45 [60.8]	48 [60]	8.63	735 (-3.5%)	360 (25.0%)	375 (-63.1%)	90	725	334	2610	1675	1.9 (13.6%)	8.1	7.0 (-180.0%)
M5	40 [17.6]	45 [60.8]	60 [75]	8.33	700 (-1.4%)	345 (28.1%)	355 (-54.3%)	85	725	327	2600	1650	1.0 (54.5%)	8.1	7.6 (-204.0%)
Initial B6				7.44	775	505	270	141	780	462	2720	1603	3.5	8.3	0.3
N1	20 [8.8]	45 [60.8]	20 [25]	8.16	675 (12.9%)	390 (22.8%)	285 (-5.6%)	110	720	319	2650	1603	0.4 (88.6%)	8.4	5.8 (-1833.3%)
N2	20 [8.8]	45 [60.8]	40 [50]	8.13	725 (-6.5%)	325 (35.6%)	400 (-48.1%)	70	810	355	2620	1603	0.3 (91.4%)	8.3	5.1 (-1600.0%)
N3	20 [8.8]	45 [60.8]	60 [75]	8.53	600 (22.6%)	285 (43.6%)	315 (-16.7%)	48	870	355	2570	1828	0.6 (82.8%)	8.2	4.5 (-1400.0%)
N4	20 [8.8]	45 [60.8]	80 [100]	9.22	540 (30.3%)	250 (50.5%)	290 (-7.4%)	36	900	341	2560	1891	0.7 (80.0%)	8.1	0.4 (-33.3%)
N5	20 [8.8]	45 [60.8]	100 [125]	9.72	525 (32.3%)	225 (55.4%)	300 (-11.1%)	41	840	360	2560	1721	0.2 (94.3%)	8.0	0.4 (-33.3%)
O1	20 [8.8]	60 [81]	20 [25]	8.75	550 (29.0%)	122 (75.8%)	428 (-58.5%)	42	840	355	2560	1748	0.3 (91.4%)	7.9	0.6 (-100.0%)
O2	20 [8.8]	60 [81]	40 [50]	9.02	515 (33.5%)	118 (76.6%)	397 (-47.0%)	37	780	348	2560	1604	0.1 (97.1%)	8.0	2.0 (-566.7%)
O3	20 [8.8]	60 [81]	60 [75]	9.22	530 (31.6%)	285 (43.6%)	245 (9.2%)	33	810	319	2560	1726	0.2 (94.3%)	7.8	2.4 (-700.0%)
O4	20 [8.8]	60 [81]	80 [100]	9.73	530 (31.6%)	275 (45.5%)	255 (-5.6%)	42	810	319	2560	1717	0.2 (94.3%)	7.7	0.5 (-66.7%)
O5	20 [8.8]	60 [81]	100 [125]	10.11	510 (34.2%)	305 (39.6%)	205 (-24.1%)	46	900	341	2590	1854	0.2 (94.3%)	7.7	2.1 (-600.0%)
Initial B7				7.94	775	480	295	115	780	454	2700	1637	3.2	7.7	3.2
P1	20 [8.8]	90 [121.6]	20 [25]	8.13	645 (16.8%)	390 (18.8%)	255 (13.6%)	57	660	334	2520	1480	0.1 (96.9%)	7.6	8.2 (-156.2%)

P2	20 [8.8]	90 [121.6]	40 [50]	8.42	555 (28.4%)	325 (32.3%)	230 (22.0%)	40	750	341	2500	1589	0.1 (96.9%)	7.5	8.4 (-162.5%)
P3	20 [8.8]	90 [121.6]	60 [75]	9.23	560 (27.7%)	270 (43.8%)	290 (1.7%)	30	870	327	2490	1872	0.2 (93.7%)	7.5	0.2 (93.8%)
P4	20 [8.8]	90 [121.6]	80 [100]	9.59	550 (29.0%)	265 (44.8%)	285 (3.4%)	34	780	327	2480	1671	0.3 (90.6%)	7.4	0.2 (93.8%)
P5	20 [8.8]	90 [121.6]	100 [125]	9.74	480 (38.1%)	265 (44.8%)	215 (27.1%)	35	810	362	2480	1620	0.0 (100.0%)	7.4	0.3 (90.6%)
Q1	3 [1.3]	90 [121.6]	0	7.85	700 (9.7%)	370 (22.9%)	330 (-11.9%)	75	700	341	2610	1590	0.1 (96.9%)	8.6	25.6 (-700.0%)
Q2	5 [2.2]	90 [121.6]	0	7.93	700 (9.7%)	375 (21.9%)	325 (-10.2%)	78	700	412	2620	1491	0.2 (93.7%)	8.6	26.3 (-721.9%)
Q3	8 [3.5]	90 [121.6]	0	7.98	725 (6.5%)	390 (18.8%)	335 (-13.6%)	75	650	362	2620	1481	0.3 (90.6%)	8.6	27.7 (-765.6%)
Q4	20 [8.8]	90 [121.6]	0	7.92	680 (12.3%)	345 (28.1%)	335 (-13.6%)	80	750	369	2630	1632	0.3 (90.6%)	8.6	22.9 (-615.6%)
Initial B8				6.57	775	438	337	90	700	440	2720	1531	4.9	8.7	3.8
R1	15 [6.6]	120 [162]	25 [31.2]	8.27	610 (21.3%)	438 (0.0%)	285 (15.4%)	37	780	327	2440	1627	0.2 (95.9%)	8.0	15.5 (-307.9%)
R2	15 [6.6]	120 [162]	50 [62.5]	9.06	565 (27.1%)	325 (25.8%)	275 (18.4%)	34	870	362	2440	1826	0.1 (97.9%)	8.0	2.2 (42.1%)
R3	15 [6.6]	120 [162]	75 [93.7]	9.53	540 (30.3%)	290 (33.8%)	285 (15.4%)	29	900	383	2430	1841	0.1 (97.9%)	7.9	0.5 (86.6%)
R4	15 [6.6]	120 [162]	100 [125]	9.69	500 (35.5%)	255 (41.8%)	240 (28.8%)	31	990	391	2430	1980	0.0 (100.0%)	7.9	0.3 (92.1%)
R5	15 [6.6]	120 [162]	125 [156.2]	9.74	500 (35.5%)	260 (40.6%)	260 (22.8%)	29	725	398	2440	1419	0.0 (100.0%)	7.8	0.4 (89.5%)
S1	15 [6.6]	75 [101.3]	100 [125]	10.45	550 (29.0%)	250 (42.9%)	300 (11.0%)	88	870	440	2300	1654	0.2 (95.9%)	8.1	0.4 (89.5%)
S2	15 [6.6]	90 [121.6]	100 [125]	10.64	555 (28.4%)	250 (42.9%)	305 (9.5%)	80	840	391	2290	1671	0.3 (93.9%)	7.9	0.2 (94.7%)
S3	15 [6.6]	105 [141.9]	100 [125]	10.35	560 (27.7%)	270 (38.3%)	290 (13.9%)	73	780	391	2260	1558	0.3 (93.9%)	7.9	0.3 (92.1%)
S4	15 [6.6]	120 [162]	100 [125]	10.58	575 (25.8%)	325 (25.8%)	250 (25.8%)	65	780	391	2260	1580	0.4 (91.8%)	7.9	0.3 (92.1%)
S5	15 [6.6]	135 [182.4]	100 [125]	10.58	580 (25.2%)	370 (15.5%)	210 (37.7%)	61	810	391	2290	1653	0.5 (89.8%)	7.9	0.3 (92.1%)
Initial B9				8.35	800	452	348	152	540	447	2410	1135	5.4	9.3	2.6
T1	15 [6.6]	120 [162]	25 [31.2]	9.51	500 (37.5%)	280 (38.0%)	220 (36.8%)	40	760	383	2210	1501	1.1 (79.6%)	8.1	0.4 (84.6%)
T2	15 [6.6]	120 [162]	50 [62.5]	9.95	480 (40.0%)	275 (39.1%)	205 (41.1%)	41	720	355	2230	1436	0.9 (83.3%)	8.1	0.4 (84.6%)
T3	15 [6.6]	120 [162]	75 [93.7]	9.60	400 (50.0%)	250 (44.7%)	150 (56.9%)	34	680	376	2180	1255	0.8 (85.2%)	8.1	0.5 (80.8%)
T4	15 [6.6]	120 [162]	100 [125]	10.15	455 (43.1%)	250 (44.7%)	205 (41.1%)	41	680	391	2210	1280	0.7 (87.0%)	8.1	0.6 (76.9%)
T5	15 [6.6]	120 [162]	125 [156.2]	9.86	450 (43.8%)	250 (44.7%)	200 (42.5%)	41	720	408	2220	1341	0.7 (87.0%)	8.1	3.0 (-15.4%)
U1	0	110 [148.6]	130 [162.5]	7.98	625 (21.9%)	345 (23.7%)	280 (19.5%)	48	510	398	2215	1330	0.7 (87.0%)	8.3	11.4 (-338.5%)
U2	5 [2.2]	110 [148.6]	130 [162.5]	8.02	625 (21.9%)	310 (31.4%)	315 (9.5%)	42	660	391	2210	1399	2.1 (61.1%)	8.5	11.2 (-330.8%)
U3	10 [4.4]	110 [148.6]	130 [162.5]	8.10	625 (21.9%)	325 (28.1%)	300 (13.8%)	53	660	390	2220	1389	2.3 (57.4%)	8.3	10.8 (-315.4%)
U4	15 [6.6]	110 [148.6]	130 [162.5]	7.98	625 (21.9%)	300 (33.6%)	325 (6.6%)	54	690	383	2220	1461	2.4 (55.6%)	8.7	12.1 (-365.4%)
U5	20 [8.8]	110 [148.6]	130 [162.5]	7.86	625 (21.9%)	290 (35.8%)	335 (3.7%)	50	660	390	2210	1391	2.1 (61.1%)	8.4	10.6 (-307.7%)

3.2. Effect of lime and NaOH at fixed alum dose

3.2.1. Effect of lime and NaOH at fixed alum dose at 40 mg/L

Another set of jar test experiments are performed fixing alum dose at 40 mg/L, which is found achieving a good efficiency 12.6% (see E2 in Table 2). Different combinations are examined: J4, K1 to 5, L1 to 5, and M1 to M5 (Table 2). A good TH removal is obtained (18.3%) for K5 (40-30-60) beaker with sodium and sulfate increase, 24.4% and 7.5%, respectively.

3.2.2. Effect of lime and NaOH at fixed alum dose at 20 mg/L

A similar set of jar test experiments as those performed at the previous Section are realized for alum dose 20 mg/L. As seen in Table 2, different combinations are performed: N1 to 5, O1 to 5, P1 to 5, Q4, and U5. As shown in Table 2, an optimal result (38.1% of TH removal) is achieved for P5 (20-90-100).

3.2.3. Effect of lime and NaOH at fixed alum dose at 15 mg/L

A similar set of jar test experiments as those performed at the two previous Sections are realized for alum dose 15 mg/L. As seen in Table 2, different combinations are performed: R1 to 5, S1 to 5, T1 to 5, and U4. As shown in Table 2, an optimal result (50.0% of TH removal) is achieved for T3 (15-120-75).

3.3. Results discussion

3.3.1. Treatment effectiveness

The conditions for efficient softening with good precipitation of $CaCO_3$ and $Mg(OH)_2$ are combined if:
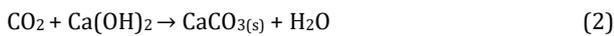
- The pH is close to 10.
- The concentration of lime, $Ca(OH)_2$, and that of NaOH are equal to or equivalent to the concentrations of carbonate hardness and non-carbonated hardness, respectively. In other words, for an optimal dose of lime, between 100 and 120 mg/L, it is equal to that of alkalinity. Similarly for

NaOH at an optimal dose of between 100 and 130 mg/L, it is equal to the non-carbonated hardness. It should be remembered that the concentrations must be expressed in mg $CaCO_3$/L to speak of the equivalence between lime and alkalinity on the one hand, and soda and non-carbonated hardness on the other hand.
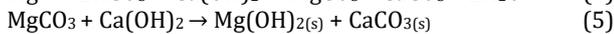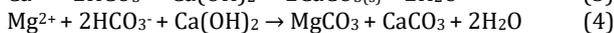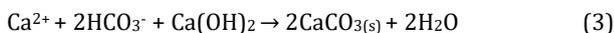
From the point of view of our objective, namely the substitution or reduction of the use of alum, as a toxic chemical product, and because of the excessive hardness of the treated water at the Ghrib station, it is necessary to reduce this hardness. Therefore, we adopt the doses of the reagents: alum, lime, and soda at T3 (15-120-75) mg/L. For this purpose, we consider that these injections are optimal, for them the yield obtained was considered maximum.

The reduction of the most studied parameters is quite obvious with the exception of sodium (Table 2), which has increased very clearly. This elevation is due to the replacement of Ca^{2+} and Mg^{2+} cations by Na^+ cation as is well-known for softening (Arrigo et al., 2007). The chemical reactions describing these phenomena are grouped as follows (Ordóñez et al., 2012; Wang et al., 2005; Abdessemed and Nezzal, 2008):
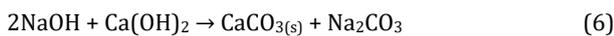
1. Carbonic acid neutralization:

$$CO_2 + Ca(OH)_2 \rightarrow CaCO_{3(s)} + H_2O \tag{2}$$

2. Carbonate hardness precipitation:

$$Ca^{2+} + 2HCO_3^- + Ca(OH)_2 \rightarrow 2CaCO_{3(s)} + 2H_2O \tag{3}$$
$$Mg^{2+} + 2HCO_3^- + Ca(OH)_2 \rightarrow MgCO_3 + CaCO_3 + 2H_2O \tag{4}$$
$$MgCO_3 + Ca(OH)_2 \rightarrow Mg(OH)_{2(s)} + CaCO_{3(s)} \tag{5}$$

3. Precipitation of non-carbonate hardness due to calcium:

$$2NaOH + Ca(OH)_2 \rightarrow CaCO_{3(s)} + Na_2CO_3 \tag{6}$$
$$Ca^{2+} + Na_2CO_3 \rightarrow CaCO_{3(s)} + 2Na^+ \tag{7}$$
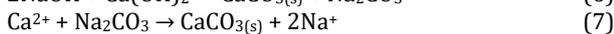
4. Precipitation of the non-carbonate hardness due to magnesium:

$$Mg^{2+} + Ca(OH)_2 \rightarrow Mg(OH)_{2(s)} + Ca^{2+} \tag{8}$$
$$Ca^{2+} + Na_2CO_3 \rightarrow CaCO_{3(s)} + 2Na^+ \tag{9}$$
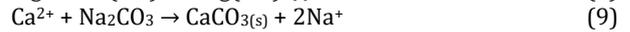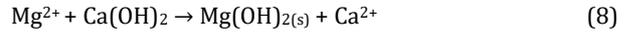
According to Reaction (9), 2 moles of Na^+ are released while 1 mole of Ca^{2+} precipitates. Hence, the remarkable increase of sodium.

Total alkalinity (TAC) decreased in the T3 case from 152 to 34 mg $CaCO_3$; this decrease is of a carbonate hardness type:

$$TH - alkalinity = non\text{-}carbonate\ hardness \tag{10}$$

or

$$TH = carbonate\ hardness + non\text{-}carbonate\ hardness \tag{11}$$

The non-carbonate hardness is related to the ions of sulfates, chlorides, and nitrates. So, the remaining as hardness is mainly non-carbonate hardness. Based on the dissolution of the remaining inorganic salts as hardness, the nature of these compound salts can be inferred in a balanced and imaginable manner. These salts are: $CaCl_2$, $CaSO_4$, $MgCl_2$, $MgSO_4$, etc.

3.3.2. Exploitation of the jar test results

In order to exploit the results obtained in a profitable way, the coagulation/softening using lime of GD water can become easily exploitable if we manage to master the methods of applying this treatment (order of reagents injection, intensity and duration of the reagent mixtures). Indeed, it is desirable to use all the information collected from this study in a summarized form and accessible to use when needed.

Table 3 summarizes some selected cases of treatment reduced to one or two reagents. Table 3 shows the importance of adding alum in a minimal quantity, which remains to be better well optimized, in the treatment with lime or soda alone, in order to facilitate the softening process and to contribute to the coagulation of the resulting fine seeds of the softening reaction.

Table 3: Summary of selected cases of treatment reduced to one or two reagents

Used reagents	Reagents combination	Alum (mg/L)	Lime (mg/L)	NaOH (mg/L)	TH removal percent (%)
Alum-Lime	Q4 (20-90-0)	20	90	0	12.3
Alum-NaOH	I5 (60-0-40)	60	0	40	9.5
Lime-NaOH	U1 (0-110-130)	0	110	130	21.9
Alum	E2 (40-0-0)	40	0	0	12.6
Lime	D5 (0-50-0)	0	50	0	13.7
NaOH	F5 (0-0-40)	0	0	40	7.2

It may seem illogical to use alum at such a high pH (~10), but it is to obtain magnesium aluminate (and not aluminum hydroxide) to better precipitate magnesium. Coagulation process itself may be necessary either to treat troubled waters or to promote the precipitation of dissolved organic matter (DOM) (Kweon and Lawler 2004; Ghernaout et al., 2014, 2015a; Vahedi and Gorczyca, 2014).

3.3.3. Ranking of the first 20 yields obtained with their corresponding doses

A comparison is made between the best TH removals yields obtained from 30 to 50% in this study. The best yields are shown schematically in the graph of Fig. 1.
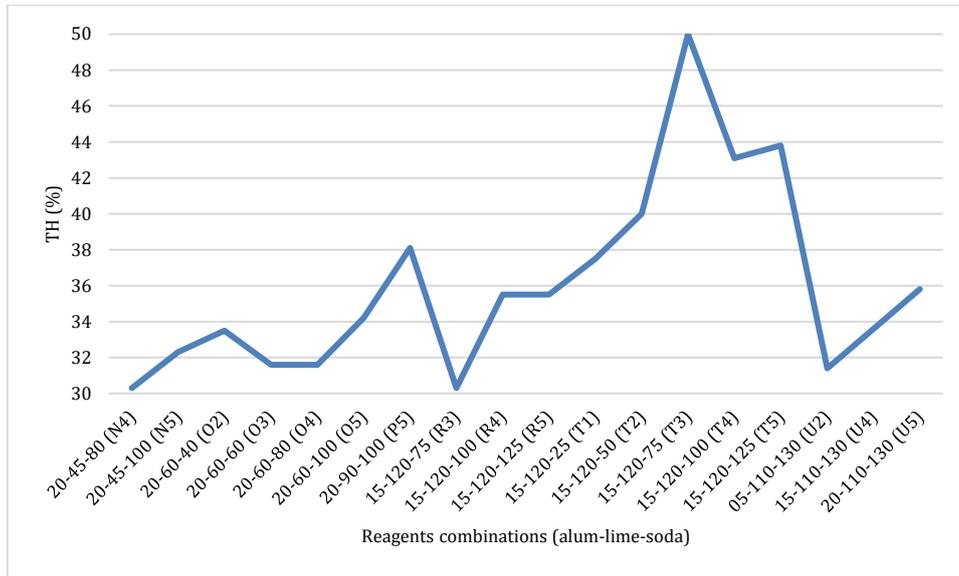
Fig. 1: Ranking of the first 20 yields obtained in descending order of efficiency

3.3.4. Advantages of coagulation with lime and soda

The main advantages of using lime and soda in coagulation process are summarized as follows (Ghernaout et al., 2009; 2015b; 2017b; Vahedi and Gorczyca, 2014; Ghernaout and Ghernaout, 2010; 2012; Ghernaout and Boucherit, 2015; Boucherit et al., 2015):

1. Precipitation of $CaCO_3$ typically reduces color, total organic carbon (TOC), and disinfection byproducts (DBPs) precursors by 10 to 30 percent.
2. Precipitation of magnesium hydroxide generally removes 30 to 60% of TOC and DBPs precursors, and 50 to 80 percent of color.
3. A good disinfection occurs when pH is of the order of 11 or more (in the case where lime is in excess).
4. Precipitation of phosphates occurs when pH is of the order of 11.
5. Precipitation of heavy metals can take place: Fe, Pb, Zn, Co, Ni, and Hg.

Cheng et al. (2009) examined brackish water with a silica concentration of 30 mg/L as SiO_2 for silica removal using in-line coagulation/ultrafiltration (UF) processes to avoid scaling problems in a reverse osmosis (RO) membrane treatment plant. Other than pH and coagulant dosage, mixing intensity and velocity gradient are also a critical factor with respect to silica removal. Different velocity gradient (G), pH and alum dose were tested and contour plots were used to determine the optimum silica removal. The optimum silica removal of 65% was achieved with G = 2000 s^{-1}, pH = 7.1 and alum dosage = 30 mg/L as Al_2O_3, corresponding to a maximum of 87% recovery for the RO membrane plant. Silica was removed by co-precipitation of aluminum hydroxide, supported by the results of turbidity and silica measurement. Streaming potential by electrokinetic analyzer was used to analyze the UF membrane before and after filtration, and a slight reduction of negative charge was observed at medium pH, resulting from the adsorption of positively charged coagulated particles on the negatively charged UF membrane surface (Cheng et al., 2009).

Esmaeilirad et al. (2015) examined the combination and sequence of softening and electrocoagulation (EC), basically as a version of chemical coagulation, methods to treat hydraulic fracturing flowback and produced water from shale oil and gas operations. They evaluated the influence of chemical softening on EC. In the softening process, pH was elevated to 9.5 and 10.2 before and after EC, respectively. Softening, when applied before EC, was more performant for eliminating turbidity with samples from wells older than one month (99% versus 88%). Nevertheless, neither technique was successful in treating samples collected from early flowback (1-day and 2-day samples), likely due to the high concentration of organic matter (OM). For TOC, hardness, Ba, Sr, and B removal, application of softening before EC appeared to be the most efficient approach, likely due to the formation of solids before the coagulation process (Esmaeilirad et al., 2015).

O'Donnell et al. (2016) studied the performance of coagulation/filtration and lime-soda ash softening treatment methods to eliminate strontium (Sr) from surface and ground waters. Their coagulation/filtration jar test results on natural waters proved that conventional treatment with Al and Fe coagulants were able to achieve only 12% and 5.9% Sr removal, while lime softening removed as high as 78% from natural strontium-containing ground water. Controlled batch experiments on synthetic water illustrated that Sr removal during the lime-soda ash softening was affected by pH, Ca concentration and dissolved inorganic carbon concentration. In all softening jar tests, the final Sr concentration was directly related to the initial Sr concentration and the removal of Sr was directly

associated with Ca removal. Precipitated solids showed well-formed crystals or agglomerates of mixed solids, two polymorphs of $CaCO_3$ (vaterite and calcite), and strontianite, depending on initial water quality conditions. X-ray diffraction analysis suggested that Sr was likely incorporated in the $CaCO_3$ crystal lattice and was likely responsible for removal during lime softening.

3.3.5. OM removal under softening conditions

At natural pH, i.e., without adjusting it, $KMnO_4$ oxidability removal is considered. For E3 (60-0-0), N2 (20-45-40), N5 (20-45-100), O1 (20-60-20) to O5 (20-60-100), P1 (20-90-20) to P5 (20-90-100), Q1 (3-90-0) to Q4 (20-90-0), R1 (15-120-25) to R5 (15-120-125), and S1 (15-75-100) to S5 (15-135-100), the $KMnO_4$ oxidability removal is higher than 90%. Alum alone (E3), alum with lime (Q1 and Q4), and alum with lime and NaOH are efficient for OM removal.

Similar results are obtained by Yan et al. (2008) who studied the treatment of organic-polluted high hardness surface water using enhanced softening via precipitation employing different coagulants, such as $FeCl_3$, $AlCl_3$ and polyaluminum chloride (PACl), in bench scale experiments. Higher elimination of natural organic matter (NOM) (UV_{254} removal attained ~50%) was obtained under enhanced softening with coagulant injection conditions as compared with conventional coagulation at natural pH or by softening alone without coagulation. They observed that PACl could improve the generation of Mg $(OH)_2$ precipitate to eliminate NOM efficiently at relative lower pH range (pH <10). At this condition, the pH for enhanced softening can be reduced importantly into the practical pH range for treatment plant operation. The efficiency of enhanced softening is influenced importantly not only by pH, coagulant type and dose, but also by raw water quality such as NOM characteristics, magnesium content and hardness. NOM with more hydrophobic content (Ghernaout, 2014) and higher molecular weight may be removed more easily in some situations. For water with higher content of Mg^{2+}, the inflexion pH for favorable UV_{254} removal is lower. They found that it is of practical value for organic-polluted water with high content of magnesium to be treated by enhanced softening with PACl (Yan et al., 2008).

Liao and Randtke (1986) studied the elimination of organic contaminants using lime softening. Co-precipitation seemed to be the important elimination mechanism for organic substances in low concentrations, and only those substances able to adsorb onto $CaCO_3$ were able to be eliminated using lime softening. All of the removable anionic compounds were able to coordinate with Ca^{2+} to form relatively insoluble complexes. Fatty acids, which form relatively soluble complexes with Ca^{2+}, and hydrophobic chemicals (Ghernaout, 2014) unable to coordinate with Ca^{2+} were not eliminated. They concluded that softening is generally performant in eliminating polymeric electrolytes possessing acidic oxygen-containing residues, such as carboxyl, phenol, enol, phosphoryl, phosphonyl, sulfuryl and sulfonyl groups. Simple monomeric molecules are not expected to be efficiently removed unless they polymerize or possess phosphorus-containing functional groups such as phosphoryl and phosphonyl.

Coro and Laha (2001) demonstrated that important color removal is obtained employing the existing lime softening process with activated silica coagulant by augmenting the softening pH to 11 or more. Increased pH also generated a > 30% decrease in TOC. They suggested, in the case of higher raw water color (≥ 80 units), the addition of ferric chloride as coagulant at dosages of 80 mg/L is recommended.

Zhou et al. (2017) quantified the competition between coagulant Al^{3+} and hardness cations to bind onto DOM using absorbance spectroscopy acquired at different Al^{3+} concentrations in the absence and presence of Ca^{2+} or Mg^{2+}. They established that, in the presence of either Mg^{2+} or Ca^{2+}, an augmenting depression of the binding of Al^{3+}-DOM could be detected in the differential spectra of DOM with the increasing of Mg^{2+} or Ca^{2+} at a level of 10, 100 and 1000 mM, with the observation being more significant at higher pH from 6.5 to 8.5. The results of zeta potentials of DOM illustrated that the competition of hardness cations results in the negative DOM being less efficiently neutralized by Al^{3+}. They proved that the elimination of DOM using coagulation would significantly deteriorate with the presence of hardness cations, which would compete with coagulant Al^{3+} to neutralize the unsaturated sites in DOM (Zhou et al., 2017).

4. Conclusion

This work concerned the study of coagulation/softening (softening is induced during coagulation) of GD water using lime as coagulant and its use in combination with NaOH to reduce TH at an acceptable level by consumers. Experiments and physicochemical analyses performed at the Central Laboratory of the Algerian Waters Company (Unity of Medea) enable us to reach a number of conclusions:

1. The partial substitution of alum with lime and NaOH in coagulation process of GD water is quite possible. Indeed, the best reduction of TH of 50% is obtained for this combination: alum = 15, lime = 100, NaOH = 100 mg/L. Poor yields, between 0 and 5%, are discarded; this can be explained by the poor choice of reagents' doses. Average yields, ranging from 5 to 15%, can be related to the lack of one of the reactants since one or two reagents alone are injected. Yields located in the range of 15-30% are considered as low successful results for which the operating conditions are not favorable. Results from 31 to 40% are considered as good tests since the softening is relatively well

implemented. The most effective yields are ranging from 43-50%.

2. The full substitution of alum by lime and NaOH, i.e., as coagulation/softening products taken alone without alum, is not applicable as an induced softening method. However, lime and NaOH as proper softening agents would be added in a separate softening process following coagulation process in a well-controlled manner in order to exploit all benefits of the softening method itself.

3. The substitution of alum is only partially applicable as alum is used in reduced dosages in order to promote the precipitation of $CaCO_3$ and magnesium hydroxide by which softening process is supposed to be realized.

4. As a perspective, it is useful to conceive a complementary process stage devoted to softening at the GD water treatment plant for more pleasant water consumption.

5. Hardness cations are ubiquitous and abundant in source water, while the effect of hardness on the performance of coagulation for DOM removal in water treatment remains unclear due to the limitation of methods that can characterize the subtle interactions between DOM, coagulant and hardness cations. This study presented a new focus for discussing the mechanism and performance of coagulation in high hardness and alkalinity/pH water; however, additional survey is required to examine the complicated interaction in the Ca^{2+}/Mg^{2+}-DOM-Al ternary system to comprehensively define the contributions of the two mechanisms – lime softening and coagulation - to DOM removal by coagulation.

Acknowledgment

List of abbreviations

DBPs	Disinfection by-products
DOM	Dissolved organic matter
EC	Electrocoagulation
G	Velocity gradient (s^{-1})
GD	Ghrib Dam
NOM	Natural organic matter
OM	Organic matter
PACl	Polyaluminum chloride
RO	Reverse osmosis
TAC	Total alkalinity
TH	Total hardness
TH$_f$	Final total hardness (mg/L as $CaCO_3$)
TH$_i$	Initial total hardness (mg/L as $CaCO_3$)
TOC	Total organic carbon
UF	Ultrafiltration
UV_{254}	Ultraviolet absorbance at 254 nm.

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