Desalination and Water Treatment
www.deswater.com

doi:10.5004/dwt.2017.20229

68 (2017) 361–368
March

Performance evaluation of solidification of Cr^{3+} or Ni^{2+} in industrial wastewater with Sorrel's cement

Chengyou Wu[a,*], Huifang Zhang[b], Hongfa Yu[a,c]

[a]*School of civil engineering, Qinghai University, Xining, 810016, China, Tel. +8614797006546, email:wuchengyou86@163.com*
[b]*Qinghai Institute of Salt Lakes, Chinese Academy of Sciences, Xining 810008, China, email: zhf233007@126.com*
[c]*Department of Civil Engineering, Nanjing University of Aeronautics and Astronautics, Nanjing 210016, China, email: yuhongfa@nuaa.edu.cn*

Received 25 August 2015; Accepted 15 August 2016

ABSTRACT

Sorrel's cement has been investigated as a means of solidifying Cr^{3+} or Ni^{2+} in wastewater. The setting time, compressive strength, water resistance, and leaching toxicity of solidified products of Sorrel's cement with Cr^{3+} or Ni^{2+} have been studied comprehensively to assess the applicability of Sorrel's cement in industrial wastewater treatment. Analysis of solidified products of Sorrel's cement with Cr^{3+} or Ni^{2+} was conducted by XRD, TG, FTIR, and SEM. Results demonstrate that the solidification of Cr^{3+} or Ni^{2+} inhibits the hydration process of Sorrel's cement. Results also indicate the inhibition of $5Mg(OH)_2{\cdot}MgCl_2{\cdot}5H_2O$ (5 phase) formation, which is the main strength phase in Sorrel's cement, subsequently leading to the reduction of compressive strength of Sorrel's cement from 136 MPa to 67 MPa; the reduced compressive strength still far exceeds the standard value of 7 MPa. Additionally, the water resistance of cement solidified products also meets the requirements specified in the related standard. The leaching toxicity of Cr^{3+} or Ni^{2+} from solidified products of Sorrel's cement was also evaluated. The highest observed leaching concentration from solidified products reached 0.110 mg/L and 0.311 mg/L for Cr^{3+} and Ni^{2+}, respectively, indicating that Sorrel's cement solidified products with Cr^{3+} or Ni^{2+} exhibits an acceptably low leaching toxicity.

Keywords: Sorrel's cement; Solidification; Cr^{3+}; Ni^{2+}; Leaching toxicity; Compressive strength; Water resistance

1. Introduction

Solidification technique is defined as a disposing-waste process that waste is solidified and stabilized by adding curing agent, in which harmful substances in waste usually transform into insoluble, nontoxic and immobile substances [1]. In the 1950s, solidification technique was applied firstly in USA for disposing radioactive waste, and afterward, has been gradually developing as a pretreatment process for heavy metals-containing waste or wastewater since the 1970s [2]. Solidification of heavy metal ions in waste has been investigated by low-cost curing agents such as slag [3], lime, beringite, red mud [4], cement [5–10], ash [11,12], glass [13], and other specific reagents [14–18]. The immobilization mechanisms involved in such solidification systems include metal ion sorption, phase mixing and substitution, as well as precipitation of metal hydroxides, carbonates, sulfides and phosphates [4,6,17,18].

In practical applications, cementitious materials are widely used for stabilizing heavy metals in waste. In the USA, an independent panel of scientists and engineers commissioned by the Nuclear Regulatory Commission (NRC) concluded that cementitious solidification technologies ought to be given serious consideration for wastes. Portland cement is the most commonly adopted medium for the solidification technique [6]. However, magnesia cement materials have exhibited superior

---

*Corresponding author.

Presented at 2015 Qingdao International Conference on Desalination and Water Reuse, June 29 – July 2, 2015 Qingdao, China

characteristics for the substitution of wood and steel as compared to the Portland cement, largely due to their remarkable advantages including fast hardening, high strength, strong cohesiveness, as well as good abrasion resistance, fireproofing, and sound insulation properties [19,20]. Magnesia cement has high early strength and does not require moisture curing, which is more suitable than ordinary Portland cement for quickly solidifying heavy metal ions in wastewater prior to landfill to lower the leaching rate. Furthermore, the lower alkalinity of magnesia cement (magnesium oxychloride cement with pH of 9–10) compared to the higher ones of ordinary cement (pH of 12–13) [21] avoids sparingly soluble metal salts calcium chromate ($CaCrO_4$, $K_{sp} = 7.1 \times 10^{-4}$) [22,23] generation and reduces the leaching probability of amphoteric metal ions [24]. Some previous studies have reported the significance of waste solidification using magnesia cement materials. Buj et al. have studied the effects of Pb^{2+}, Ni^{2+}, Zn^{2+}, Cu^{2+} and Cr^{3+} on the leaching toxicity and mechanical properties of magnesium phosphate cements. It was pointed out that magnesium phosphate cement showed a better solidification behavior for Pb^{2+} and Cr^{3+} under acidic or neutral conditions and the compressive strength decreased with the increase of heavy metal content [7,8]. Compared to magnesium phosphate cement, Sorrel's cement was investigated to solidify Cr^{3+} or Ni^{2+} in the present study because of its lower production cost. The setting time, compressive strength, water resistance, and leaching toxicity of solidified products of Sorrel's cement with Cr^{3+} or Ni^{2+} were measured and evaluated comprehensively.

2. Experimental section

2.1. Materials

According to the calcination temperature of magnesium minerals, magnesium oxide can be divided into light-burn magnesium oxide (calcination temperature: 700~800°C) and dead-burn magnesium oxide (calcination temperature: more than 1000°C). The preparation of Sorrel's cement requires highly active magnesium oxide, which can be obtained by calcining raw magnesite at 700~800°C [20]. Therefore, the light-burn raw magnesite powder used in this study was calcined at 750°C and chemical compositions are listed in Table 1. It can be found that the content of total MgO was 85.5%, and active MgO (A-MgO) was detected to be only 58.7% according to hydration method [25]. Magnesium chloride ($MgCl_2 \cdot 6H_2O \geq 98\%$) used in this study is provided by Qinghai Good-Friend Magnesium Industry Co., Ltd, China. Analytically pure $Cr(NO_3)_3$ and $Ni(NO_3)_2$ were purchased from Shanghai Boer Chemical Reagent Co. Ltd., China.

Table 1
Chemical compositions of light-burn raw magnesite powder

Chemical composition	Mass fraction (%)
MgO	85.5
SiO_2	6.1
CaO	3.1
Fe_2O_3	1.0
CO_2	4.0
SO_3	0.2

2.2. Sample preparation

Magnesium chloride and $Cr(NO_3)_3$ or $Ni(NO_3)_2$ were simultaneously dissolved together into water with various concentrations of Cr^{3+} or Ni^{2+} (0, 10 g/L, 20 g/L, 30 g/L, 40 g/L). The Cr^{3+} solutions were of approximate pH = 5–6, while the Ni^{2+} solutions were of approximate pH = 3–4. Due to the different hydrolysis constants of Cr^{3+} and Ni^{2+}, the magnesium chloride solutions with Cr^{3+} or Ni^{2+} show different acidity. However, the Cr^{3+} solutions or the Ni^{2+} solutions have relatively constant pH values instead of gradually various ones with various concentrations of Cr^{3+} or Ni^{2+} because magnesium chloride with high concentration in the solutions restraints the hydrolysis of heavy metal ion. The mixture solution was then added to light-burn raw magnesite powder to produce a cement paste. The molar ratio of A-MgO, $MgCl_2$ and H_2O in the cement samples was fixed at 7:1:15. After being stirred for 30 min, the mixture slurry was cast into PVC molds of 20 mm × 20 mm × 20 mm; the samples were removed from the molds after the cement was set. The cement specimens were then cured at (20 ± 2)°C and a relative humidity of (35 ± 5)%. In the following discussion, cement samples solidified with Cr^{3+} (or Ni^{2+}) are referred to as Cn or Nn, respectively, where "n" denotes the concentration of Cr^{3+} or Ni^{2+} in the magnesium chloride solution of the sample. For example, C40 denotes the solidified cement sample was prepared with a Cr^{3+} concentration of 40 g/L. The control samples consisted of cement samples without the addition of any heavy metals.

2.3. Setting time analysis

The setting time of cement specimens was determined by a Vicat apparatus at ambient temperature with standard method: tests method for water requirement of normal consistency, setting time and soundness of Portland cement (GB/T 1346–2001). The initial setting time was defined as when a Vicat needle of 1 mm in diameter would penetrate the cement specimen to a point of 5 ± 1 mm from the bottom of the mould. The final setting time was defined as when a 5 mm cap ring would leave no visible mark when placed on the sample.

2.4. Compressive strength analysis

Unconfined compressive strength was tested by using a concrete compressive machine with a maximum load of 100 kN. The cubes cured in air were tested for their 1st day, 3rd day, 7th day, 28th day compressive strength at a loading rate of 5 mm/min. The compressive strength of cement solidified samples dipped in water for different times was tested with the same method meanwhile.

2.5. XRD, SEM, TG and FTIR analysis

Cement samples were analyzed in powdered form, with a particle size (D90) < 74 μm. The crystal phase composition of the cement samples was determined by X-ray diffraction (XRD) with Cu radiation, 30 kV of acceleration voltage and 2θ range of 5°–75°. The microstructures of the cement samples cured in air for 28 d were characterized by scanning electron microscopy (SEM, JSM-5610LV) on a fractured surface with gold coating. The thermogravity (TG) curves were recorded with a Q600 differential scanning calorimetry and the samples were heated from 40° to 1000°C at 5°/min. Fourier Transform Infrared Spectroscopy (FTIR) analysis was performed on a NEXUS Fourier transform infrared spectroscopy.

2.6. Toxicity characteristics leaching procedure

The leachability of Cr^{3+} or Ni^{2+} from solidified cement products was investigated according to the standard leaching method presented in the toxicity characteristics leaching procedure (TCLP, EPASW-86), developed in the USA [26]. The solidified cemented products cured in air for various amounts of time were crushed and passed through 40-mesh (0.45 mm) sieve before dipping in water with a liquid–solid ratio of 5:1 and a stirring rate of 110 rpm for 24 h. In addition, the powder of the solidified cement products cured in air for 28 d was dipped into hydrochloric acid solution of pH from –1 to 5 and an identical liquid–solid ratio for 24 h. The leaching concentrations of Cr^{3+} or Ni^{2+} from solidified cement products were analyzed by ICP-AES with a detection limit of 10 μg/L.

3. Results and discussion

3.1. Setting time of solidified cement products

As an important index of cement hydration speed, setting time is the vital step for fabrication a dense and chemically stable materials in Sorrel's cement [27]. The setting times of Sorrel's cement solidified samples are depicted in Table 2.

Results clearly indicate that the incorporation of Cr^{3+} or Ni^{2+} can extend both the initial and final setting times of Sorrel's cement. For instance, the final setting time of C40 was 7.5 h, comparing with 4.1 h of control sample. Results further indicate that the incorporation of Cr^{3+} or Ni^{2+} can inhibit cement hydration process. Sorrel's cement hydration reactions can be written as the formulas below [28,29]:

Table 2
Setting time of Sorrel's cement solidified products without (Control) or with Cr^{3+} or Ni^{2+}

Samples	Initial setting time (h)	Final setting time (h)
Control	2.3	4.1
C10	2.6	5.5
C20	3.6	6.2
C30	3.9	7.1
C40	4.8	7.5
N10	2.4	4.5
N20	2.4	4.9
N30	2.8	5.8
N40	3.2	6.5

$$MgO + H_2O \rightarrow Mg(OH)_2 \quad (1)$$

$$Mg(OH)_2\,(s) \rightarrow Mg^{2+} + OH^- \quad (2)$$

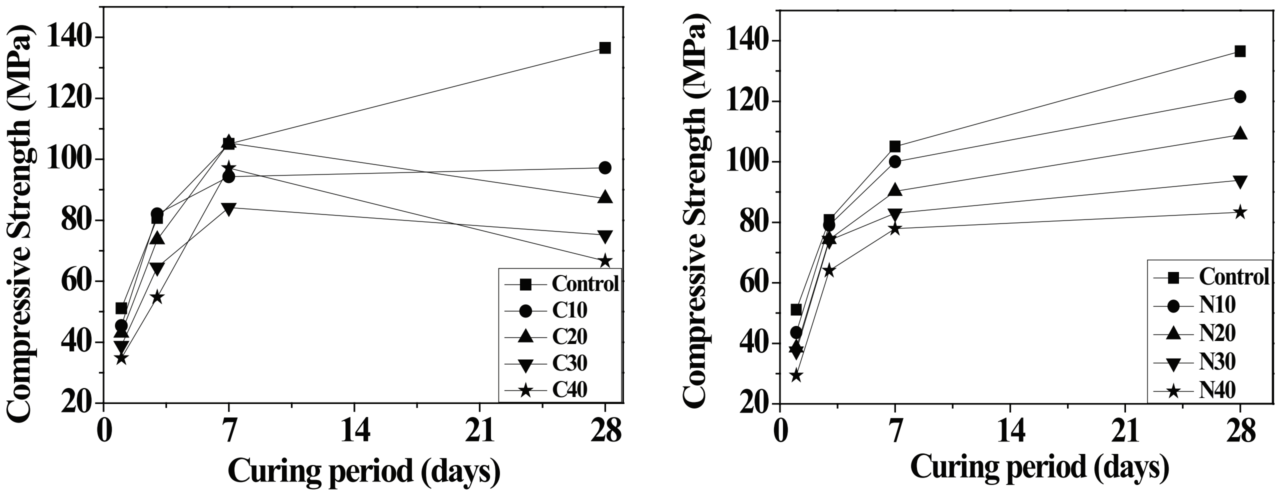
$$6Mg^{2+} + 10OH^- + 2Cl^- + 8H_2O \rightarrow 5Mg(OH)_2{\cdot}MgCl_2{\cdot}8H_2O(s) \quad (3)$$

When the Mg^{2+}, OH^- and Cl^- in the cement system reach saturation, $5Mg(OH)_2{\cdot}MgCl_2{\cdot}8H_2O$ (5 phase) can be produced and crystallize, leading to the coagulation of the cement sample. Cr^{3+} or Ni^{2+} can interact with the OH^- resulting from the reaction described by Formula (2) acutely to produce insoluble hydroxides in the cement system, and consequently inhibit the reaction described by Formula (3), which may result in the extension of the setting time of solidified cement samples. Furthermore, the setting time for Cr samples is longer than that of Ni samples with identical heavy metal concentration. A possible reason for this phenomenon is that the consumption amount of OH^- by trivalent Cr^{3+} is greater than that by bivalent Ni^{2+} over the same period of time. For instance, the theoretical amount of OH^- interacted with Cr^{3+} in the C40 cement system is predicted approximately at 2.3 mol/L, while the theoretical amount of OH^- interacted with Ni^{2+} in the N40 cement system is predicted to reach approximately 1.4 mol/L when the heavy metal ions have reacted completely.

3.2. Compressive strength of solidified cement products

The compressive strength is one of the most important indexes for evaluating the stability of cement solidified products. Fig. 1 shows the compressive strength development of solidified cement products with Cr^{3+} or Ni^{2+}. Results indicate that the compressive strength of solidified cement products decreases as the dosage of Cr^{3+} or Ni^{2+} in cement samples increases. The 28th-day compressive strength of solidified cement products declines according to the following orders: Control (136 MPa) > C10 (97 MPa) > C20 (87 MPa) > C30 (75 MPa) > C40 (67 MPa) and Control (136 MPa) > N10 (122 MPa) > N20 (109 MPa) > N30 (94 MPa) > N40 (83 MPa). Although the incorporation of Cr^{3+} or Ni^{2+} has adverse effects on the compressive strength of solidified cement products, the 28th-day compressive strength (67 MPa and 83 MPa for C40 and N40, respectively) far exceed China's 7 MPa characteristic requirements for solidified cement body cured for 28 d [30].

The compressive strength of Sorrel's cement is closely related to its phase composition [31]. Fig. 2 depicts the crystal phase composition of the control, C40, N40 samples cured in air for 28 d. The results indicate that crystalline substances in Sorrel's cement samples with (C40, N40) or without heavy metal (Control) are $5Mg(OH)_2{\cdot}MgCl_2{\cdot}8H_2O$ (5 phase) and MgO. Fig. 3 depicts the deformation ν (O–H)



(a) (b)

Fig. 1. The compressive strength development of cement solidified products: (a) with Cr^{3+}; (b) with Ni^{2+}.

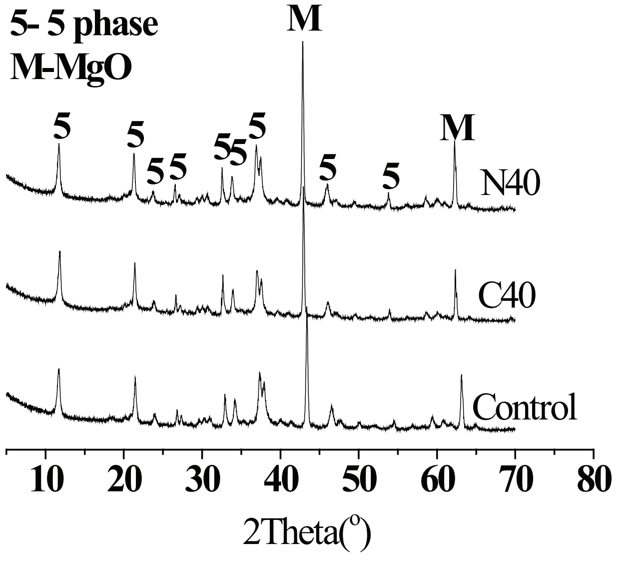


Fig. 2. XRD spectra of cement solidified products cured in air for 28 d.

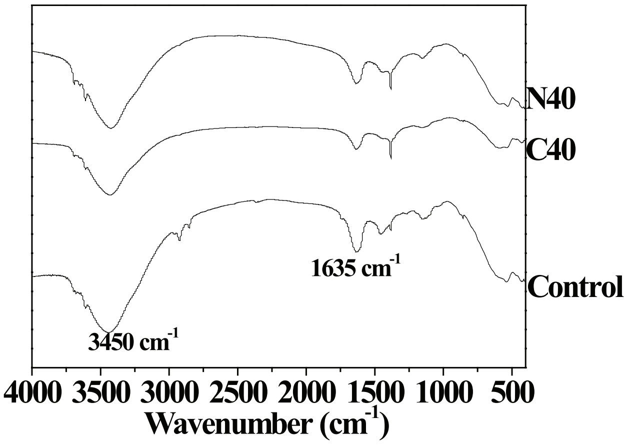


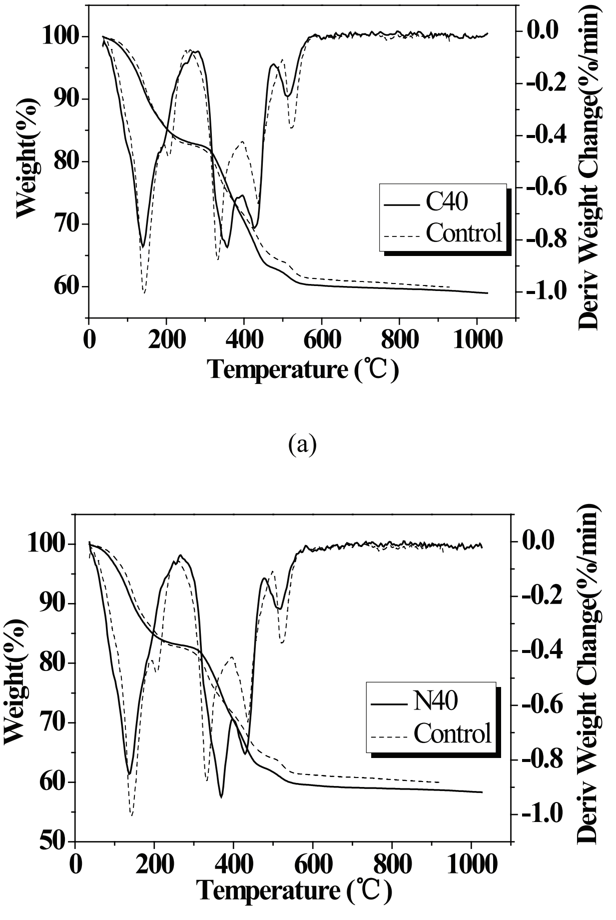
Fig. 3. FTIR of cement solidified products cured in air for 28 d.

and δ (O–H) intensity centers at 3450 cm^{-1} and 1635 cm^{-1}, respectively, which are associated with the presence of hydroxyl groups in the solidified cement system.

Fig. 4 shows TG/DTG analysis of solidified cement products, verifying the first water loss temperature of phase 5 at 137~142°C, as previously reported by Xia's study [32]. The weightlessness rate of the 5 phase in Sorrel's cement with Cr^{3+} or Ni^{2+} is less than that of the control sample, which indicates that the yield of the 5 phase in solidified cement samples with Cr^{3+} or Ni^{2+} is less than that in control sample. Therefore, it can be concluded that the addition of Cr^{3+} or Ni^{2+} can inhibit the formation of 5 phase in Sorrel's cement, thus resulting in a the decrease in the compressive strength of Sorrel's cement samples.

Furthermore, the compressive strength of Cr samples decreased from 7 days to 28 d, while that of Ni samples increased. Fig. 5 shows the microstructure of solidified Sorrel's cement samples both without (Control) and with Cr^{3+} (a) or Ni^{2+} (b) cured in air for 28 d. 5 phase presents the typical needle-like crystal structure in control sample. However, adding Cr^{3+} can alter the crystallize pattern of 5 phase. It is remarkable that 5 phase is not needle-like but gelatinous in C40 samples. Usually, the solid phase volume of magnesia cement expands during the hydration process. In the early reaction, the hydration product (5 phase) mainly plays a gelling and filling effect; in the later hydration period, it extrudes the capillary pores and results in the crystallization pressure stress in the cement system. Compared with

Ni samples (with needle-like 5 phase), gelatinous 5 phase in (Fig. 5a) has lower tensile strength and is insufficient to overcome the crystallization pressure stress, leading to the visible decrease of the Cr sample strength.



(a)

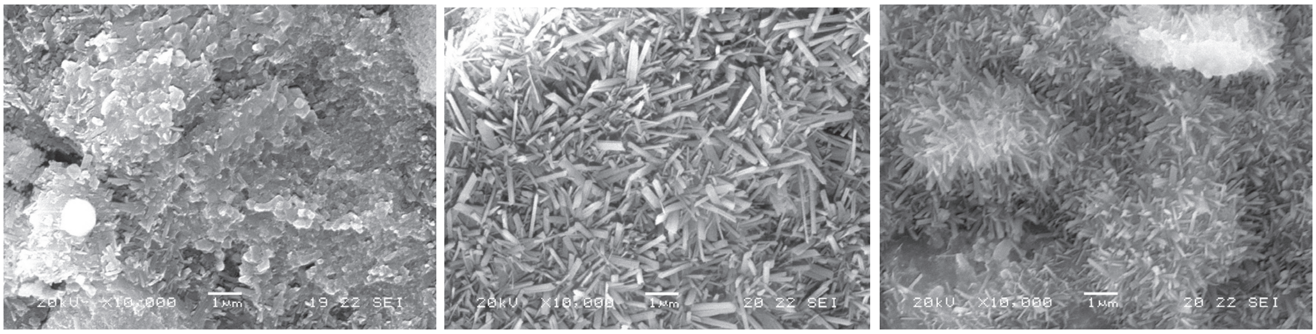
(b)

Fig. 4. TG curves of cement solidified products cured in air for 28 d. (a) for C40 and control test and (b) for N40 and control test.

3.3. Water resistance of solidified cement products

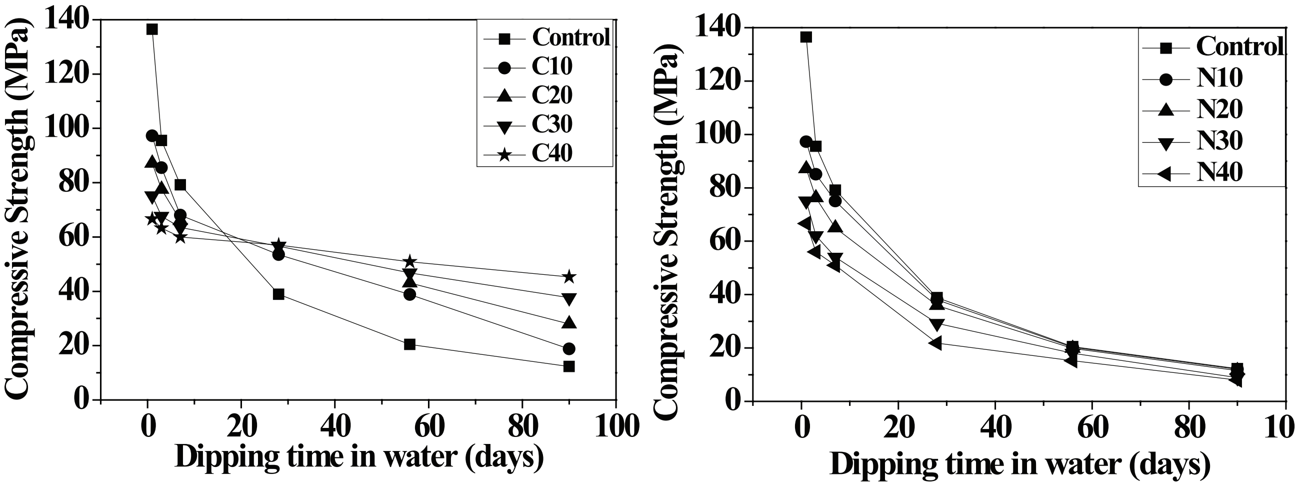
Solidified cement products with heavy metals are usually disposed of in landfills [33]; therefore, solidified products must possess favorable mechanical properties. The US Environment Protection Agency (abbreviated as USEPA) recommends a minimum compressive strength of 0.35 MPa for solidified products after being dipped in water for 28 d [26]. The compressive strengths of solidified cement products with Cr^{3+} or Ni^{2+} after dipping in water for various amounts of time are depicted in Fig. 6. The compressive strength of solidified cement products decreases obviously after dipping in water with and without heavy metals, which is accordant with the results previously obtained in Chau's study [34]. The compressive strength of solidified products of Sorrel's cement with Cr^{3+} after dipped in water for 28 d is greater than that of control samples, and the reduction rate decreases as the dosage of Cr^{3+} increases. For example, the compressive strength of C40 sample decreases to 45 MPa from 66 MPa by 32% after being dipped in water for 90 d, while that of Control samples decreases by 91% from 136 MPa to 12 MPa after the identical treatment. However, the compressive strength of solidified cement products with Cr^{3+} or Ni^{2+} far exceeds 0.35 MPa. The water resistance of Sorrel's cement is poor due to the dissolubility of the needle-like crystal present during the 5 phase [35]. In Cr samples, 5 phase is observed to be gelatinous phase which is more stable than a needle-like 5 phase [36].

The crystal phase composition of solidified Sorrel's cement samples without (Control) and with Cr^{3+} (C40) or Ni^{2+} (N40) after being dipped in water for 90 d has also been depicted in Fig. 7. Results indicate that 5 phase disappears and transforms to $Mg(OH)_2$ in both control sample and N40 sample after being dipped in water, whereas some evidence of 5 phase remains in C40 sample. The crystal (r_x) radii are equal to 0.65Å, 0.64Å and 0.70Å for Mg^{2+}, Cr^{3+} and Ni^{2+}, respectively [37]. Therefore, as compared to Ni^{2+}, Cr^{3+} more easily participates in the formation process of 5 phase and even to form gelatinous material like 5 phase through phase mixing and substitution interaction. Future area of study include the influence mechanism of metal ions on 5 phase morphology and the existence state of metal ions in the cement system.



(a) (b) (Control)

Fig. 5. SEM images of Sorrel's cement solidified samples without (Control) or with Cr^{3+} (a) or Ni^{2+} (b) cured in air for 28 d.



(a) (b)

Fig. 6. The compression strength of Sorrel's cement solidified products without (Control) or with Cr^{3+} (a) or Ni^{2+} (b) dipped in water for different times.

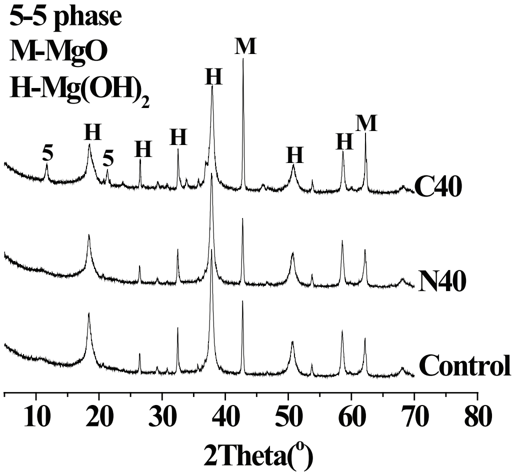


Fig. 7. XRD spectra of cement solidified products without (Control) or with Cr^{3+} (C40) or Ni^{2+} (N40) after dipping in water for 90 days.

3.4. Leachability of heavy metals from solidified Sorrel's cement products

Leaching toxicity of solidified cement products with Cr^{3+} or Ni^{2+} should be low enough that samples could be buried safely in underground to avoid pollution of groundwater. The leaching concentrations of Cr^{3+} and Ni^{2+} from solidified Sorrel's cement samples cured for various lengths of time are listed in Table 3. All the data represent lower values than the highest concentration, as described by the related standards in China and USEPA (Table 4).

Table 3
Leached concentration of heavy metal from cement solidified samples cured for different time (mg/L)

	1 day	3 days	7 days	28 days
C10	BDL*	BDL	BDL	0.015
C20	BDL	BDL	BDL	0.048
C30	BDL	BDL	0.012	0.079
C40	BDL	BDL	0.025	0.110
N10	0.285	BDL	BDL	BDL
N20	0.398	0.101	BDL	BDL
N30	0.422	0.363	0.255	0.180
N40	0.543	0.480	0.390	0.311

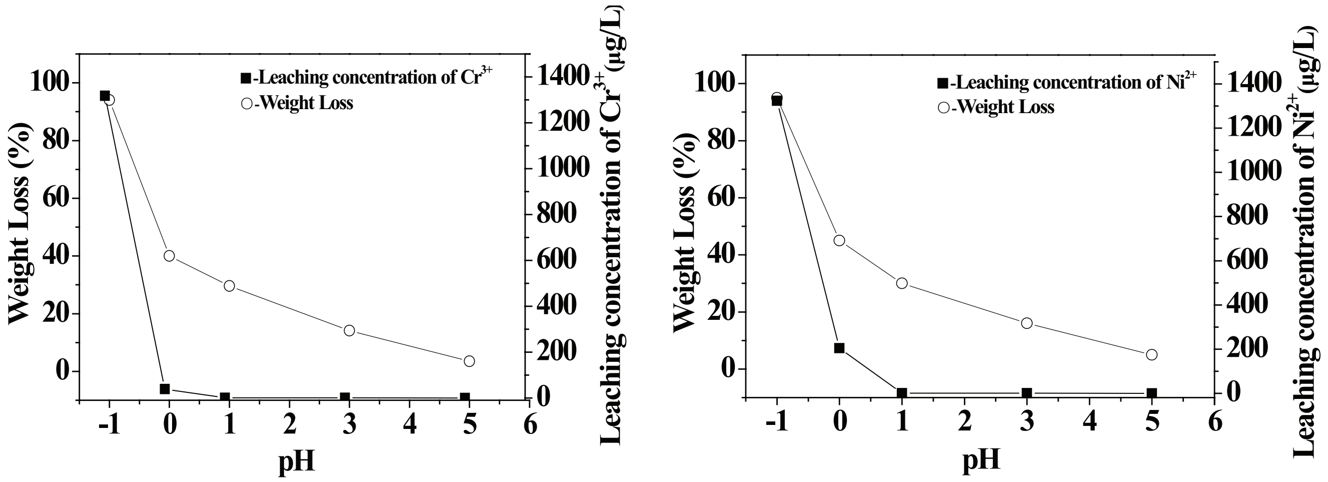
* BDL: Below the detection limit.

Table 4
Identification standard for hazardous waste – The highest concentration allowed in leachate (mg/L)

Standard	Cr (mg/L)	Ni (mg/L)
GB5085.1-1996	10	10
USEPA TCLP	5	5

Additionally, the leached concentration of Cr^{3+} is far less than that of Ni^{2+}. This may be due to more stable gelatinous 5 phase and chromium compound like 5 phase (mentioned above) existed in the cement products with Cr^{3+} have lower solubility than the components of Ni^{2+}. Meanwhile, the leaching concentration of Ni^{2+} decreased as the curing time increases. The reason is speculated that firstly Ni^{2+} adsorbed on the surface of the needle like 5 phase is quickly leached out, and thereafter Ni^{2+} which is solidified in 5 phase by phase mixing and precipitation interaction is leached out slowly with the dissolution of 5 phase in the cement system.

Fig. 8 displays the leaching concentration of Cr^{3+} or Ni^{2+} from solidified cement samples dipped in hydrochloric acid



(C40) (N40)

Fig. 8. Leached concentration of Cr^{3+} (C40) or Ni^{2+} (N40) and weight loss from Sorrel's cement samples dipped in hydrochloric acid solution of different pH.

solution of various pH values. Cr^{3+} or Ni^{2+} can leach out rapidly only when pH value of the hydrochloric acid solution is less than 1. When pH value of hydrochloric acid solution exceeds 1, the leached concentration of Cr^{3+} or Ni^{2+} is nearly constant with the acidity of solution increases, despite the weight loss of solidified Sorrel's cement samples increases continually. Experimental results indicate that leaching toxicity of Cr^{3+} or Ni^{2+} solidified in Sorrel's cement is low enough to be safely buried underground according to the related standards because Sorrel's cement is an alkali system.

4. Conclusions

In this work, Sorrel's cement has been investigated to solidify Cr^{3+} and Ni^{2+}. The setting time, compressive strength, water resistance, and leaching toxicity have been comprehensively investigated to assess the applicability of Sorrel's cement in industrial wastewater treatment. The primary conclusions are described as follows:

1. The solidification of Cr^{3+} or Ni^{2+} can inhibit the hydration process of Sorrel's cement, and setting time of Sorrel's cement paste extends as the dosage of Cr^{3+} or Ni^{2+} increases.
2. The compressive strength of solidified Sorrel's cement products decreases from 136 MPa to 83 MPa (in the case of Ni^{2+}) and 67 MPa (in the case of Cr^{3+}) because Cr^{3+} or Ni^{2+} inhibits the formation of $5Mg(OH)_2 \cdot MgCl_2 \cdot 8H_2O$ (5 phase) in Sorrel's cement system.
3. Water resistance of Sorrel's cement can be greatly improved by adding Cr^{3+} because adding Cr^{3+} alters the morphology of 5 phase and leads to lower solubility of 5 phase. And compressive strength of Sorrel's cement solidified products with Cr^{3+} (53 MPa) or Ni^{2+} (8 MPa) after dipping in water for 28 d is far higher than that in the related standard (0.35 MPa for 28 d).
4. The highest leaching concentration is only 0.110 mg/L and 0.311 mg/L for Cr^{3+} and Ni^{2+}, respectively from the solidified products, indicating that solidified Sorrel's cement products with Cr^{3+} or Ni^{2+} demonstrates an appropriately low leaching toxicity.

Acknowledgements

This work was supported by the National Natural Science Foundations of China (Grant No. U1407104, No.51662035 and No. 21401209) and Natural Science Foundations of Qinghai Province (Grant No. 2015-ZJ-937Q and No. 2015-ZJ-947Q).

References

[1] D. Grasso. Hazardous Waste Site remediation, Source Controls Lewis, 1993, 20–30.
[2] E.F. Barth. An overview of the history, present status and future direction of solidification/stabilization technologies for hazardous waste treatment, J. Hazard. Mater., 24 (1990) 103–109.
[3] H.N. Cho, J.H. Shim, J.Y. Park. Performance evaluation of solidification/stabilization of dredged sediment using alkali-activated slag, Desal. Water Treat., 57(22) (2016) 10159–10168.
[4] E. Lombi, R.E. Hamon, S.P. McGrath, M.J. McLaughlin. Lability of Cd, Cu, and Zn in polluted soils treated with lime, beringite, and red mud and identification of a non-labile colloidal fraction of metals using isotopic techniques, Environ. Sci. Technol., 37 (2003) 979–984.
[5] F.Q. Deng, G.G. Qian, MSWI fly ash based novel solidification/stabilization matrices for heavy metals, J. Wuhan Univ. Technol. Mater., 23 (2008) 955–960.
[6] M.L.D. Gougar, B.E. Scheetz, D.M. Roy, Ettringite and C-S-H Portland cement phases for waste ion immobilization: A review, Waste Manage., 16 (1996) 295–303.

[7] I. Buj, J. Torras, D. Cassellas, M. Rovira, J. de Pablo. Effect of heavy metals and water content on the strength of magnesium phosphate cements, J. Hazard. Mater., 170 (2009) 345–350.

[8] I. Buj, J. Torras, M. Rovira, J. de Pablo. Leaching behaviour of magnesium phosphate cements containing high quantities of heavy metals, J. Hazard. Mater., 175 (2010) 789–794.

[9] F. Jin, F. Wang, A. Al-Tabbaa, Three-year performance of in-situ solidified/stabilised soil using novel MgO-bearing binders, Chemosphere, 144 (2016) 681–687.

[10] J.L. Ma, Y.C. Zhao, J.M. Wang, Effect of magnesium oxychloride cement on stabilization/solidification of sewage sludge, Construct. Build. Mater., 24 (2010) 79–83.

[11] Y.B. Wang, L. Huang, R. Lau, Conversion of municipal solid waste incineration bottom ash to sorbent material for pollutants removal from water, J. Taiwan Inst. Chem. Eng., 60 (2016) 275–286.

[12] J. Yang, S.G. Zhang, D.A. Pan, B. Liu, C.L. Wu, A.A. Volinsky, Treatment method of hazardous pickling sludge by reusing as glass-ceramics nucleation agent, Rare Metals, 3 (2016) 269–274.

[13] J. Yliniemi, J. Pesonen, M. Tiainen, M. Illikainen, Alkali activation of recovered fuel-biofuel fly ash from fluidized-bed combustion: stabilization/solidification of heavy metals, Waste Manage., 43 (2015) 273–282.

[14] B.R. Zhang, W.X. Zhou, H.P. Zhao, Z.P. Tian, F.T. Li, Y.N. Wu, Stabilization/solidification of lead in MSWI fly ash with mercapto functionalized dendrimer chelator, Waste Manage., 2016, DOI: 10.1016/j.wasman. 2016. 02. 001.

[15] J.W. Beak, S.R. Mallampati, H.S. Park, Novel synthesis and applications of Thiomer solidification for heavy metals immobilization in hazardous ASR/ISW thermal residue, Waste Manage., 49 (2016) 181–187.

[16] F.H. Wang, B. Zhao, F. Zhang, J. Gao, H.T. Hao, S.T. Zhang, A novel heavy metal chelating agent sixthio guanidine acid for in situ remediation of solis contaminated with multielements: its synthesis, solidification, biodegradability, and leachability, J. Soils Sedim., 2 (2016) 371–381.

[17] Y.J. Du, M.L. Wei, K.R. Reddy, H.L. Wu, Effect of carbonation on leachability, strength and microstructural characteristics of KMP binder stabilized Zn and Pb contaminated soils, Chemosphere, 144 (2016) 1033–1042.

[18] F.H. Wang, F. Zhang, Y.J. Chen, J. Gao, B. Zhao, A comparative study on the heavy metal solidification/stabilization performance of four chemical solidifying agents in municipal solid waste incineration fly ash, J. Hazard. Mater., 300 (2012) 451–458.

[19] H.P. Lu, P.L. Wang, N.X. Jiang, Design of additives for water-resistance magnesium oxychloride cement using pattern recognition, Mater. Lett., 20 (1994) 217–223.

[20] C.K. Chau, Z.J. Li, Accelerated reactivity assessment of light burn magnesium oxide, J. Amer. Ceram. Soc., 91 (2008) 1640–1645.

[21] Z.J. Li, C.K. Chau, Influence of molar on properties of magnesium oxychloride cement, Cement Concrete Res., 37 (2007) 866–870.

[22] G. Kakali, S. Tsivilis, A. Tsialtas, Hydration of ordinary Portland cement made from raw mix containing transition element oxides, Cement Concrete Res., 28(3) (1998) 335–340.

[23] D.R. Lide, Handbook of Chemistry and Physics, 78th ed., 1997–1998.

[24] C. Belebchouche, K. Moussaceb, A. Tahakourt, A. Ait-Mokhtar, Parameters controlling the release of hazardous waste (Ni^{2+}, Pb^{2+} and Cr^{3+}) solidified/stabilized by cement-CEM I, Mater. Structure, 48(7) (2015) 2323–2328.

[25] J.M. Dong, H.F. Yu, L.M. Zhang, Study on experimental condition of hydration methods of determining active magnesium oxide content, J. Salt Lake Res., 18 (2010) 38–41.

[26] USEPA, Method 1311: Toxicity Characteristic Leaching Procedure, in: Response, Office of Solid Waste and Emergency (Ed.), Test Methods for Evaluating Solid Waste, SW-846, U.S. Environmental Protection Agency, Washington, DC, 1992.

[27] D. Hall, R. Stevens, B. EI-Jazairi. The effect of retarders on the microstructure and mechanical properties of magnesia-phosphate cement mortar, Cement Concrete Res., 31 (2001) 455–465.

[28] D.H. Deng, C.M. Zhang, The formation mechanism of the hydrate phase in magnesium oxychloride cement, Cement Concrete Res., 29 (1999) 1365–1371.

[29] H.F Yu, Magnesium magnesium oxychloride cement and it's application, Build. Mater. Industry China, (1993) 180–190.

[30] GB 14569.1–2001. Characteristic requirements for solidified waste of low and intermediate level radioactive waste—Cement solidified waste in China.

[31] C.K. Chau, Z.J. Li. Microstructures of magnesium oxychloride cement, Mater. Structures, 41 (2008) 853–862.

[32] S.P. Xia, P.F. Xing, S.Y. Gao, Studies on the basic compounds of magnesia cement: the thermal behaviour of magnesium oxychlorides, Thermochim. Acta, 183 (1991) 349–363.

[33] W.F. Cole, T. Demediuk, X-ray, thermal and dehydration studies on magnesium oxychloride, Austral. J. Chemistry, 8 (1955) 234–235.

[34] C.K. Chau, J. Chau, Z.J. Li, Influences of fly ash on magnesium oxychloride motar, Cement Concrete Compos., 31 (2009) 250–254.

[35] H.Q. Li, G.Z. Li, Y.Z. Yu, The influence of compound additive on magnesium oxychloride cement/urban refuse floor tile, Construct. Build. Mater., 22 (2008) 521–525.

[36] Q.Q. Liu, Durability, failure mechanism and life prediction method of glass fiber reinforced magnesium oxychloride cement siding [D]. Nanjing University of Aeronautics and Astronautics, 2010.

[37] E.R. Nightingale. Phenomenological theory of ion solvation-effective radii of hydrated ions, J. Phys. Chem., 9 (1959) 1381–1387.