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THE CHEMOMECHANICAL EFFECT AND THE MECHANOCHEMICAL EFFECT ON HIGH-PERFORMANCE CONCRETE SUBJECTED TO STRESS CORROSION

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ABSTRACT

This paper describes the behavior of high-performance concrete under both chemical attacks and mechanical stresses. The specimens of the concrete C80 and C95 were subjected to flexural loads with a level of 30% of their initial strengths and immersed into a 5% ammonium sulfate solution, a 10% ammonium nitrate solution, and water saturated with $Ca(OH)_2$, respectively. The development of strength of the concrete was determined at certain time intervals. The simultaneous action of corrosive media and mechanical stresses on the concrete leads to a stress corrosion effect. The difference of strength of the specimens, which were immersed in the water saturated with $Ca(OH)_2$ and the salt solutions and subjected to the same loading conditions, is defined as a chemomechanical effect (CME). The difference of strength of the unloaded and loaded specimens, both of which were immersed in the same salt solutions, is defined as a mechanochemical effect (MCE). The CME and the MCE of the concrete are discussed in the present paper. © *1998 Elsevier Science Ltd*

Introduction

Many investigations have been carried out to understand the chemical attack phenomenon. However, in many practical applications, not only do chemical attacks occur in concrete structures, but also the action of chemical attack and mechanical stresses can simultaneously take place when these materials are applied with loads. This phenomenon is known as "stress corrosion." It is known that other materials, such as metals, ceramics, glass, and polymers are significantly affected by stress corrosion if a mechanical load is superimposed to an existing corrosive attack (1–3).

In 1966, Sereda, Feldman, and Swensen (4,5) reported that the relative humidity led to a reduction in the flexural strength of a cement paste, in particular under the range 0–20% relative humidity. This phenomenon might result from the change of links of the siloxen:

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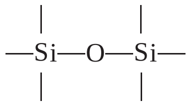
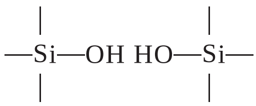


by OH^- into silicone-alcohol:



In 1974, Wittmann and Zaitsev (6) reported concrete fatigue. They found that the concrete, which was loaded to 80% of its critical strength, would fail after a finite loading time.

The investigation on stress corrosion of the cementitious materials was first initiated by Schneider and co-workers in 1984 (7–15). The influences of water-cement (w/c) ratio, type of cement, load level, concentration of the aggressive solution, surface treatment and coatings, and notch depth, etc. on the flexural strength of cement mortar and ordinary concrete were investigated. Based on the experimental results, the stress corrosion was considered a process accelerated by the activation energy of chemical reactions and could be described by the following equation (16):

$$r = \tau^{-1} = const \cdot EXP\left[-\left\{ \frac{(Q - C)}{RT} - \frac{X \cdot \sigma/(\beta_0 - \sigma)}{RT} \right\} \right] \tag{1}$$

$$ln\ \tau = A - B \cdot \frac{\sigma}{\beta_0 - \sigma} \tag{2}$$

where τ is the life-time (day); Q is the activation energy of chemical reaction; RT is the thermal energy; C is the reducing of activation energy due to corrosion; A is the experimental constant; B is the experimental constant $B = X/RT$; σ is the test stresses; β_0 is the initial strength; and X is a constant.

In 1991, the permeability and porosity structure of the concrete immersed into corrosive solution under a flexural loading condition were determined at the Technical University of Braunschweig, Germany (17).

In 1995, Middel investigated mechanical and fracture behavior of the concrete immersed into a 5%$(NH_4)_2SO_4$ solution under a central tensile stress by the determination of the full stress-strain-curve (18).

Gerdes and Wittmann (6) investigated the flexural strength and the fracture energy of the cement paste immersed into a 10% $(NH_4)_2SO_4$ solution under 60% or 80% of its initial flexural strength. As a result, the following equation was proposed to describe the flexural strength development of the cement mortar subjected to stress corrosion:

$$\beta(t) = 4.75\left[1 + \frac{0.278 \cdot t}{0.257 + t} + a(\sigma/\sigma_0)^{n} \cdot t^2 \right] \tag{3}$$

where t is the immersion time (day); σ is the subject flexural stress; σ_0 is the initial strength of the specimen at beginning of corrosion; and a and n are experimental constants.

The stress corrosion of high-performance concrete was first initiated in 1993 and has been carried out by Schneider and Chen (19–25) in Vienna.

Definition of the CME and MCE

Stress corrosion includes two kinds of attack, namely, the chemical attack and the mechanical attack. Construction elements are usually subjected to both the chemical and mechanical attacks. Therefore, two different effects, referred to as the chemomechanical effect and the mechanochemical effect, could occur in terms of the concept of "stress corrosion" (16,24).

The chemomechanical effect (CME) describes the influence of chemical reaction on mechanical behavior, i.e., the chemical effect is more predominant than the mechanical effect in stress corrosion. The mechanochemical effect (MCE) describes the influence of the applied mechanical load under both the chemical and mechanical attacks, i.e., the mechanical effect is more pronounced in comparison with the chemical effect.

In our work, the results showed clearly that the strength of cementitious materials declined much more rapidly under stress corrosion than under pure chemical corrosion. The difference in strength between the unloaded and loaded specimens immersed into a corrosive solution can be attributed to the applied load. The difference is considered to be indicative of the mechanochemical effect (MCE(t)), that is:

$$MCE(t) = \Delta\beta/\beta_{28} = \frac{\beta(\mathit{unloaded;\ solution}) - \beta(\mathit{loaded;\ solution})}{\beta_{28}}$$

$$= \frac{\beta(\mathit{unloaded,\ solution})}{\beta_{28}} - \frac{\beta(\mathit{loaded,\ solution})}{\beta_{28}} \tag{4}$$

It is known that cementitious materials immersed in water saturated with $Ca(OH)_2$ (CH water) are not affected by corrosion. The reduction in strength of the specimens immersed in an aggressive solution results from the chemical attack at a certain load level. The chemical attack by the aggressive ions gives rise to a difference of strength in the specimens, which are immersed into the water saturated with $Ca(OH)_2$ and the aggressive solution, respectively. This difference of strength is defined as a chemomechanical effect (CME(t)):

$$CME(t) = \Delta\beta/\beta_{28} = \frac{\beta(\mathit{loaded;\ in\ CH\ water}) - \beta(\mathit{loaded;\ in\ solution})}{\beta_{28}}$$

$$= \frac{\beta(\mathit{loaded;\ in\ CH\ water})}{\beta_{28}} - \frac{\beta(\mathit{loaded;\ in\ solution})}{\beta_{28}} \tag{5}$$

Details of Experiments

The concrete prisms in sizes of 40 × 40 × 160 mm, were made from Austrian Portland cement PZ 475 and natural river sand 0/4 and gravel 4/8. The preparation of the specimens and the strength tests were carried out according to Austrian ÖNORM B3303 and B3310. The strength of cubic concrete in sizes of 200 × 200 × 200 mm, were 80 MPa (C80) for the HPC without silica fume and 95 MPa (C95) for the HPC with silica fume. A cement mortar (w/c = 0.50) and an ordinary concrete (OPC C40) were also prepared for comparison with the high-performance concrete.

The reference flexural and the compressive strength of the specimens were determined after curing (water storage) for 28 days. After the curing, the specimens were immersed in various solutions: a solution of 5% $(NH_4)_2SO_4$, a solution of 10% NH_4NO_3, and water

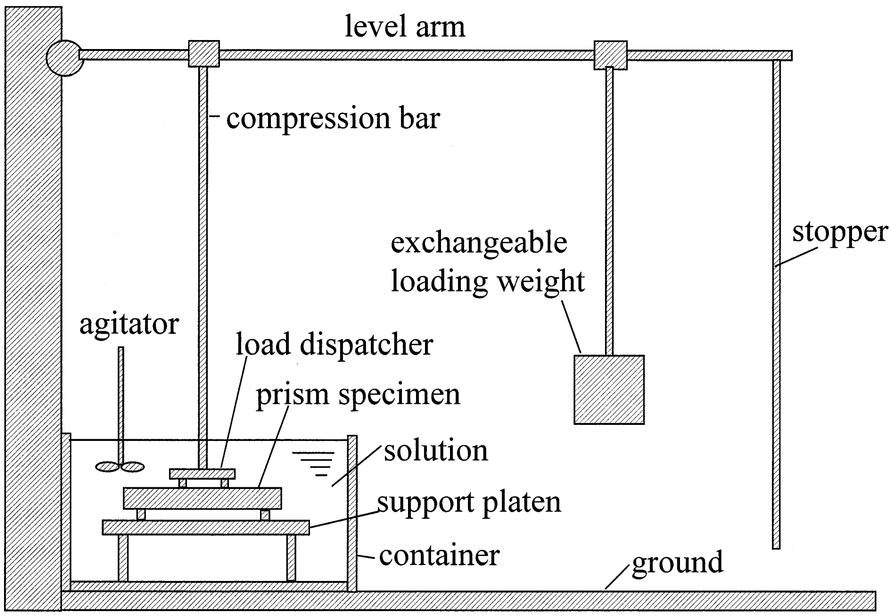


FIG. 1.
Loading Apparatus for Stress Corrosion Tests.

saturated with $Ca(OH)_2$ (CH water). A group of specimens in each series of test were loaded for 4-point-bending with a stress level of 30% of the initial flexural strength using the apparatus as shown in Figure 1. In order to make a comparison with the loaded specimens, unloaded specimens were also immersed in the same solutions. The solutions were exchanged at least at intervals of 28 days. The weight, the flexural strength, and the compressive strength of the specimens were determined at certain time intervals.

Results and Discussion

The flexural strength development of the specimens in four series of tests are shown in Table 1, 2 and 3. The relative reference strength after 28 days' curing is defined as 1.0, i.e., 100%. It was determined from the average of three specimens.

Mechanochemical Effect (MCE(t))

According to Eq. 4, the mechanochemical effect (MCE(t)) is considered as following (Eq. 6) on the basis of the experimental results and is shown in Figures 2 and 3.

$$MCE(t) = \Delta\beta/\beta_{28} = \frac{\beta(L = 0;\, solution)}{\beta_{28}} - \frac{\beta(L = 30\%;\, solution)}{\beta_{28}} \tag{6}$$

From Figures 2 and 3, it appears that the sulfate solution led to an expansive corrosion in cementitious materials, whereas the ammonium nitrate solution gave a dissolving corrosion.

TABLE 1
Flexural strength of the specimens immersed in the water saturated by $Ca(OH)_2$.

immersion time (d)	Mortar (w/c = 0.5)		OPC C40		HPC C80		HPC C95	
	L = 0	L = 30%	L = 0	L = 30%	L = 0	L = 30%	L = 0	L = 30%
7	1.01	1.03	—	—	—	—	—	—
14	1.04	1.06	1.14	1.11	—	—	—	—
28	1.05	1.08	1.15	1.04	—	—	1.05	1.07
42	—	—	1.15	1.14	—	—	—	—
56	1.06	1.06	1.09	1.17	1.09	1.12	0.93	—
70	—	—	1.20	1.17	—	—	—	1.11
84	1.09	1.10	1.21	1.20	1.00	1.02	1.09	1.06
112	—	—	—	—	1.02	1.08	0.96	—
126	—	—	—	—	—	—	—	1.12
147	—	—	—	—	—	—	1.05	—
156	1.13	1.18	—	—	1.10	—	—	0.97
182	—	—	—	—	1.08	1.09	0.93	—
252	—	—	—	—	—	—	0.96	1.02

Therefore, the mechanical effects are much more sensitive to the MCE for the specimens immersed in the ammonium sulfate solutions than for those immersed in the ammonium nitrate solutions. This effect is very strong in the cement mortar.

The mechanochemical effect on the cement mortar is significantly higher than that on the concrete, whether they were immersed in the ammonium sulfate or the ammonium nitrate

TABLE 2
Flexural strength of the specimens immersed into the 5% $(NH_4)_2SO_4$ solution.

immersion time (d)	Mortar (w/c = 0.5)		OPC C40		HPC C80		HPC C95	
	L = 0	L = 30%	L = 0	L = 30%	L = 0	L = 30%	L = 0	L = 30%
1	1.05	1.04	—	—	—	—	—	—
3	1.10	1.06	1.02	0.98	1.00	1.03	—	—
7	1.04	0.98	1.02	0.98	1.00	1.03	—	—
14	1.02	0.99	1.01	0.99	1.05	1.11	—	—
21	1.04	0.85	1.05	1.00	—	—	—	—
28	1.00	0.64	1.03	0.82	0.93	1.03	1.09	0.97
35	0.93	0.01	0.97	0.93	0.95	0.96	—	—
42	0.91	—	0.99	0.92	—	—	—	—
56	0.82	—	0.84	0.54	0.95	0.93	0.98	0.97
70	0.77	—	0.91	—	0.90	0.81	—	—
84	—	—	—	—	—	—	0.99	1.02
98	—	—	—	—	0.85	0.59	—	—
112	—	—	—	—	—	—	0.92	0.84
147	—	—	—	—	—	—	0.80	0.66
182	—	—	—	—	0.84	0.01	0.66	0.53
244	—	—	—	—	—	—	0.67	0.42
283	—	—	—	—	—	—	0.54	0.38

TABLE 3
Flexural strength of the specimens immersed into the 10% NH_4NO_3 solution.

immersion time (d)	Mortar (w/c = 0.5)		OPC C40		HPC C80		HCP C95	
	L = 0	L = 30%	L = 0	L = 30%	L = 0	L = 30%	L = 0	L = 30%
7	0.99	1.00	0.95	1.02	—	—	—	—
14	0.95	0.98	0.98	0.94	—	—	—	—
21	0.88	1.04	0.97	0.87	—	—	—	—
28	0.81	0.81	0.85	0.90	0.75	0.78	0.91	0.96
35	0.82	0.77	—	0.86	—	—	—	—
42	0.81	0.72	0.79	—	0.63	0.65	—	—
56	0.68	0.59	0.71	0.67	0.65	0.63	0.79	0.81
70	0.55	0.32	0.60	0.45	0.58	—	0.77	0.62
84	0.53	0.24	0.63	0.60	0.59	0.48	0.77	—
98	—	—	—	—	0.49	—	0.59	0.61
112	—	—	—	—	0.48	0.18	0.68	0.46
129	—	—	—	—	—	0.01	—	—
154	—	—	—	—	0.45	—	0.53	—
182	0.19	—	—	—	0.39	—	0.46	0.15
365	—	—	—	—	0.18	—	0.31	—
441	0.11	—	—	—	0.16	—	—	—
495	—	—	—	—	—	—	0.28	—
821	0.10	—	0.16	—	—	—	—	—

solution. In the presence of coarse aggregate the propagation of cracks reduced. This is the reason why the mortar is more sensitive to a mechanical attack than the concrete.

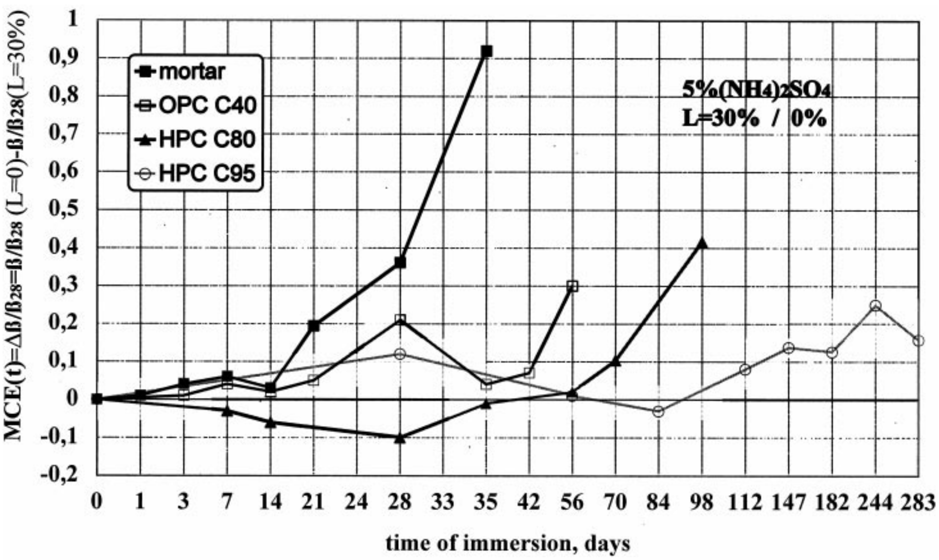


FIG. 2.
Development of the mechanochemical effects of cementitious materials immersed into 5%$(NH_4)_2SO_4$ solution under corrosion.

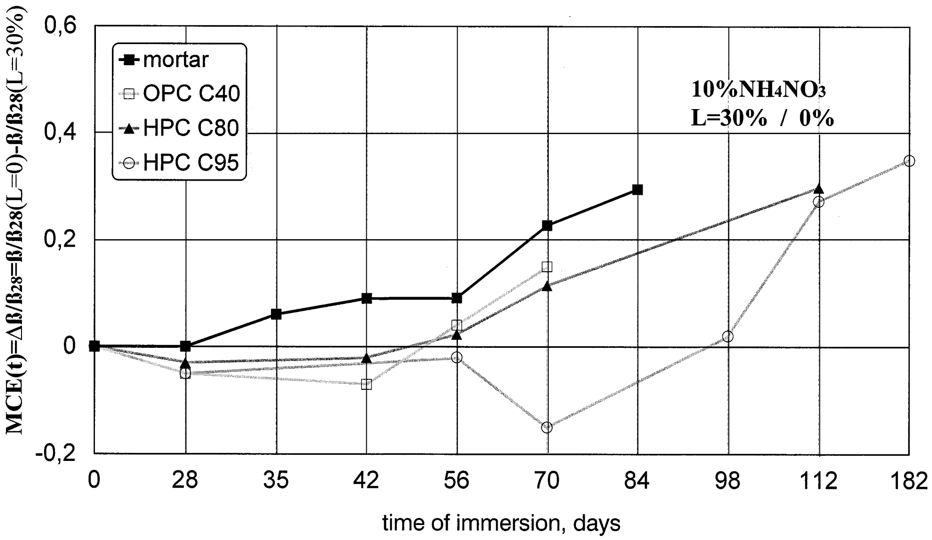


FIG. 3.
Development of the mechanochemical effects of cementitious materials immersed into 10% NH_4NO_3 solution under corrosion.

The mechanochemical effect depends on the concrete strength. The higher the initial strength of the concrete, the lower the rate of increase in the mechanochemical effect, especially when the HPC was immersed into the sulfate solution.

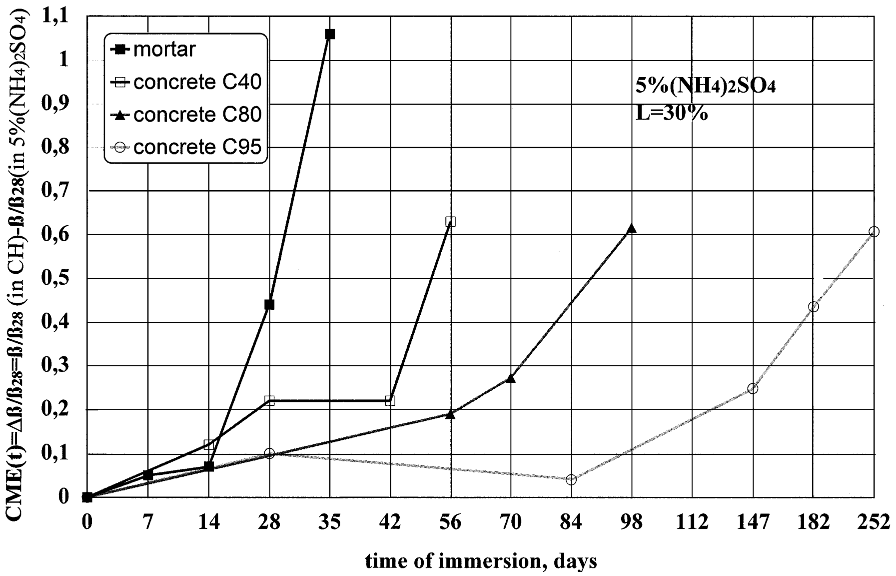


FIG. 4.
Development of the chemomechanical effects of cementitious materials immersed into $5\%(NH_4)_2SO_4$ solution under corrosion.

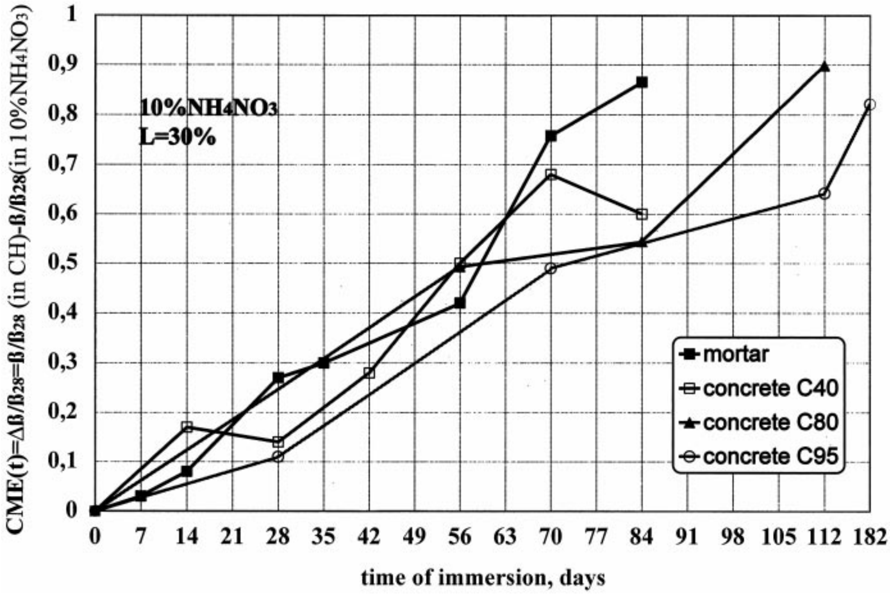


FIG. 5.

Development of the chemomechanical effects of cementitious materials immersed into 10% NH_4NO_3 solution under corrosion.

Chemomechanical Effect (CME(t))

According to Eq. 5, the chemomechanical effect (CME(t)) of the specimens that were immersed in 5%$(NH_4)_2SO_4$ and 10% NH_4NO_3 solutions is considered as Eq. 7, and the experimental results are shown in Figures 4 and 5.

$$CME(t) = \Delta\beta/\beta_{28} = \frac{\beta(L = 30\%;\ in\ CH\ water)}{\beta_{28}} - \frac{\beta(L = 30\%;\ in\ solution)}{\beta_{28}} \qquad (7)$$

It can be seen that the chemomechanical effect on the mortar specimens was more significant than that on the concrete, especially when they were subjected to strong ammonium sulfate solutions. With the 10% NH_4NO_3 solution, the difference in chemomechanical effects between the cement mortar and the concrete was apparent after 63 days of immersion, whereas it became apparent after only 14 days with 5% $(NH_4)_2SO_4$ solution.

The chemomechanical effect in the concretes subjected to 5% $(NH_4)_2SO_4$ solution under a load level of 30% of initial strength could be observed after immersion of 42 (C40), 56 (C80), and 120 (C95) days. Generally, a higher strength leads to a higher resistance to corrosion and stress corrosion. The lower the strength of a concrete, the greater the increase of the chemomechanical effect on the concrete. In particular, this behavior can be observed in the specimens immersed into the 5%$(NH_4)_2SO_4$ solution.

Empirical Formula of the CME(t) and MCE(t)

On the basis of the experimental results, the chemomechanical effect and the mechanochemical effect can be described by means of an exponential function as follows:

TABLE 4
The empirical formula of the CME(t) and MCE(t) of the mortar and the HPC concretes.

Series	Solution	Chemomechanical effect (CME(t))	Mechanochemical effect (MCE(t))
Mortar	5% $(NH_4)_2SO_4$	$CME(t) = \Delta\beta/\beta_{28} = 0.00085 \cdot t^{1.91}$ (standard deviation Sy = 0.22)	$MCE(t) = \Delta\beta/\beta_{28} = 0.0102 \cdot t^{1.122}$ (standard deviation Sy = 0.19)
	10% NH_4NO_3	$CME(t) = \Delta\beta/\beta_{28} = 0.0024 \cdot t^{1.34}$ (standard deviation Sy = 0.06)	$MCE(t) = \Delta\beta/\beta_{28} = 9.6942 \times 10^{-5} \cdot t^{1.79}$ (standard deviation Sy = 0.02)
HPC C80	5% $(NH_4)_2SO_4$	$CME(t) = \Delta\beta/\beta_{28} = 3.4532 \times 10^{-5} \cdot t^{2.13}$ (standard deviation Sy = 0.03)	$MCE(t) = \Delta\beta/\beta_{28} = 1.1662 \times 10^{-11} \cdot t^{5.32}$ (standard deviation Sy = 0.04)
	10% NH_4NO_3	$CME(t) = \Delta\beta/\beta_{28} = 0.0166 \cdot t^{0.83}$ (standard deviation Sy = 0.13)	$MCE(t) = \Delta\beta/\beta_{28} = 2.7867 \times 10^{-9} \cdot t^{3.96}$ (standard deviation Sy = 0.03)
HPC C95	5% $(NH_4)_2SO_4$	$CME(t) = \Delta\beta/\beta_{28} = 0.0025 \cdot t^{0.93}$ (standard deviation Sy = 0.14)	$MCE(t) = \Delta\beta/\beta_{28} = 0.0039 \cdot t^{0.67}$ (standard deviation Sy = 0.08)
	10% NH_4NO_3	$ME(t) = \Delta\beta/\beta_{28} = 0.0035 \cdot t^{1.09}$ (standard deviation Sy = 0.17)	$MCE(t) = \Delta\beta/\beta_{28} = 0.0059 \cdot t^{0.76}$ (standard deviation Sy = 0.15)

$$\frac{\Delta\beta}{\beta_{28}} = a \cdot t^{n} \tag{8}$$

with $\Delta\beta/\beta_{28}$ the difference of the relative flexural strength according to Eqs. 6 or 7; t the immersion time (days); and a and n experimental constants.

The regression equations of the mortar and HPC-concretes immersed in the 5% $(NH_4)_2SO_4$ and 10% NH_4NO_3 solutions under the flexural loading with 30% of their initial strength are shown in Table 4. Figure 6 to Figure 11 show the regression curves derived for the CME(t) and MCE(t) of the mortar and the HPC concrete C80 and C95, respectively.

Concluding remarks

1. The ammonium sulfate and the ammonium nitrate are very strong aggressive media attacking the cementitious materials. The high performance- concretes (HPC), whether with silica fume or without silica fume, could also be susceptible to corrosion under the aggressive solutions with very high concentration.
2. External stresses, even in the elastic stage of the specimens, can significantly accelerate the chemical attack. The stress corrosion could occur not only in cement mortar and ordinary concrete but also in high-performance concrete.
3. The definition of the chemomechanical effect and mechanochemical effect show clearly the different influences of the chemical attack and the mechanical attack, which are usually collated with the general concept of “stress corrosion.”

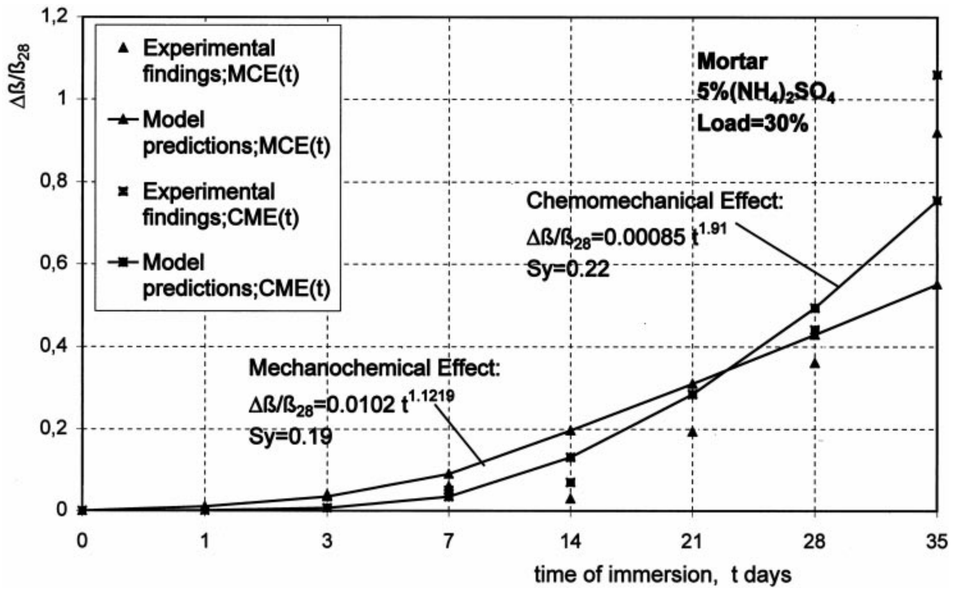


FIG. 6.
The chemomechanical and mechanochemical effects of the cement mortar with w/c = 0.50 immersed into the 5% $(NH_4)_2SO_4$ solution under the flexural loading of 30% of its initial strength.

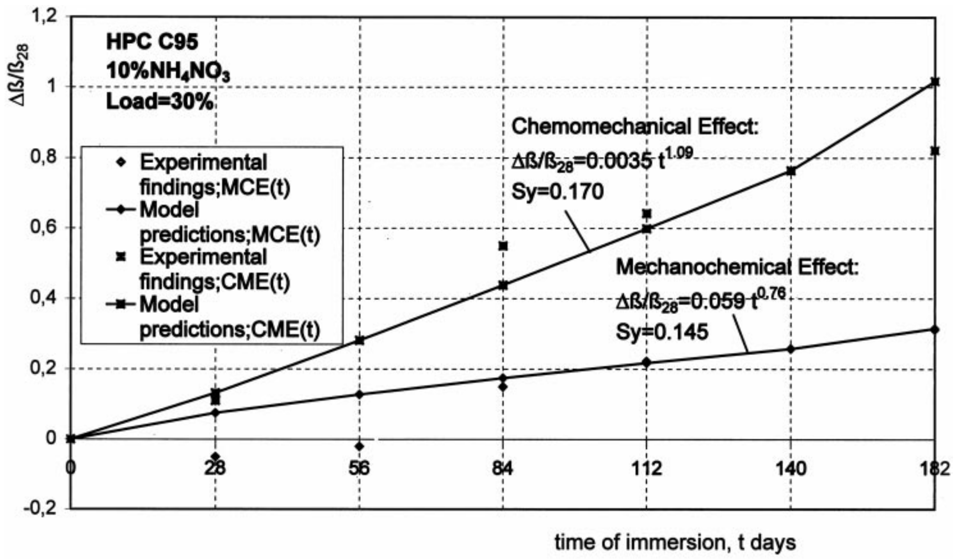


FIG. 7.
The chemomechanical and mechanochemical effects of the cement mortar with w/c = 0.50 immersed into the 10% NH_4NO_3 solution under flexural loading of 30% of its initial strength.

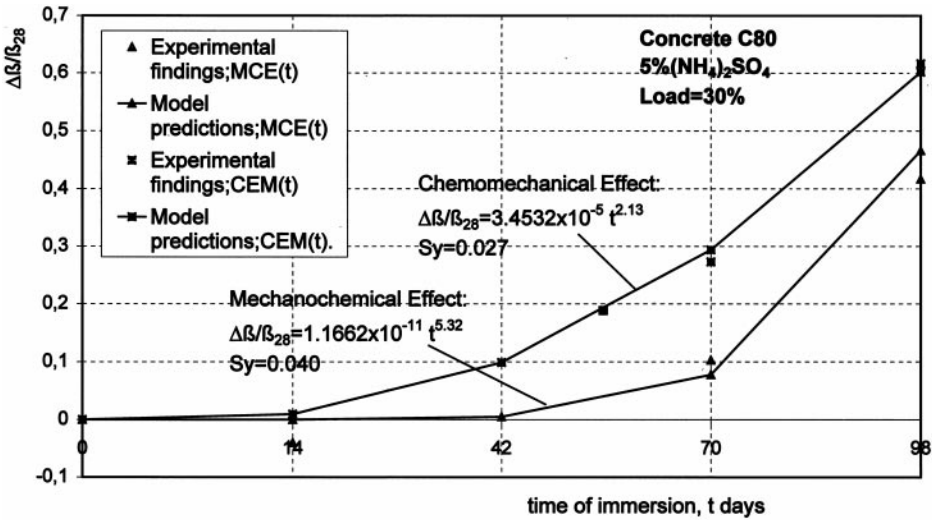


FIG. 8.

The chemomechanical and mechanochemical effects of the HPC C80 immersed into the 5% $(NH_4)_2SO_4$ solution under the flexural loading of 30% of its initial strength.

4. It seems that the stress corrosion of the cementitious materials depends on their important technological parameters, the aggressive media and external load levels. Under certain experimental conditions, the stress corrosion is essentially dependent on the concentration of the aggressive solutions and the load level only.

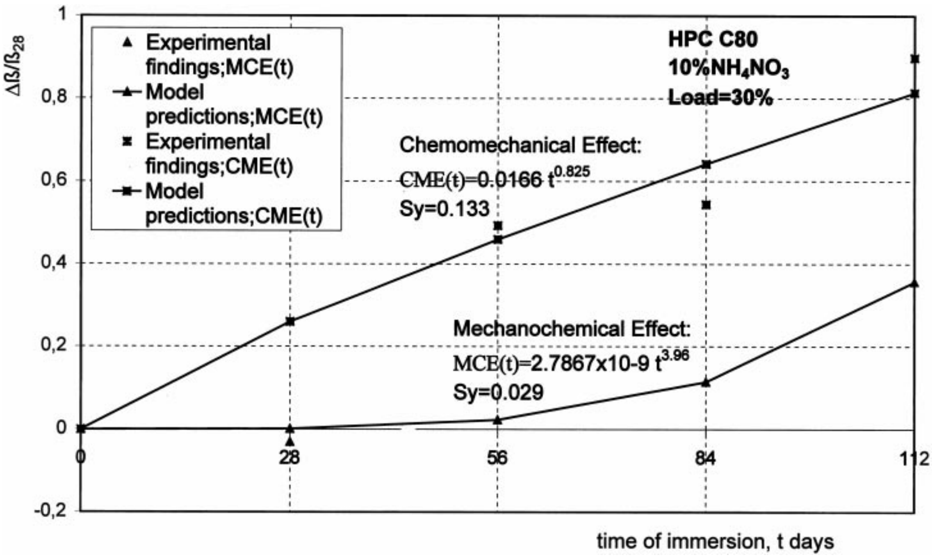


FIG. 9.

The chemomechanical and mechanochemical effects of the HPC C80 immersed into the 10% NH_4NO_3 solution under the flexural loading of 30% of its initial strength.

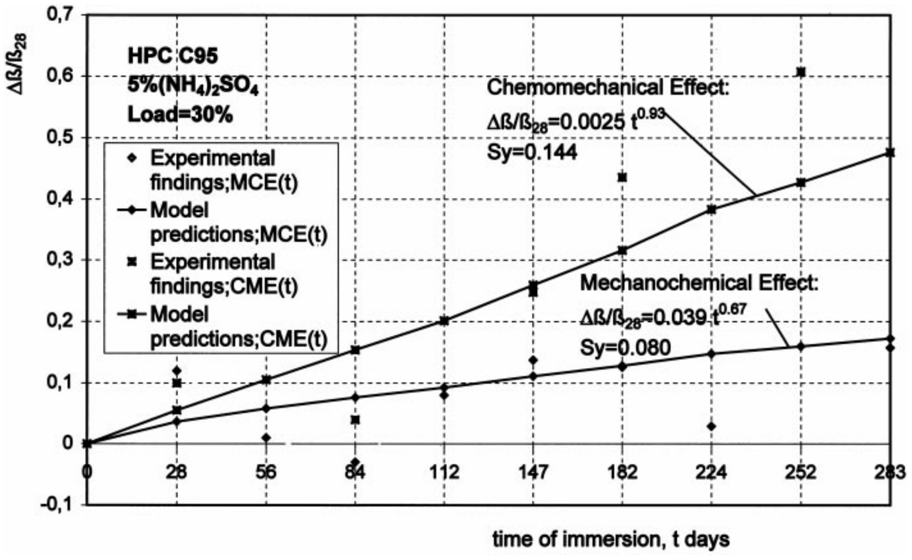


FIG. 10.
The chemomechanical and mechanochemical effects of the HPC C95 immersed into the 5% $(NH_4)_2SO_4$ solution under the flexural loading of 30% of its initial strength.

5. The effects of stress corrosion on the concrete and the mortar are different. The effect of service load conditions is more pronounced on the cement mortar than that on the

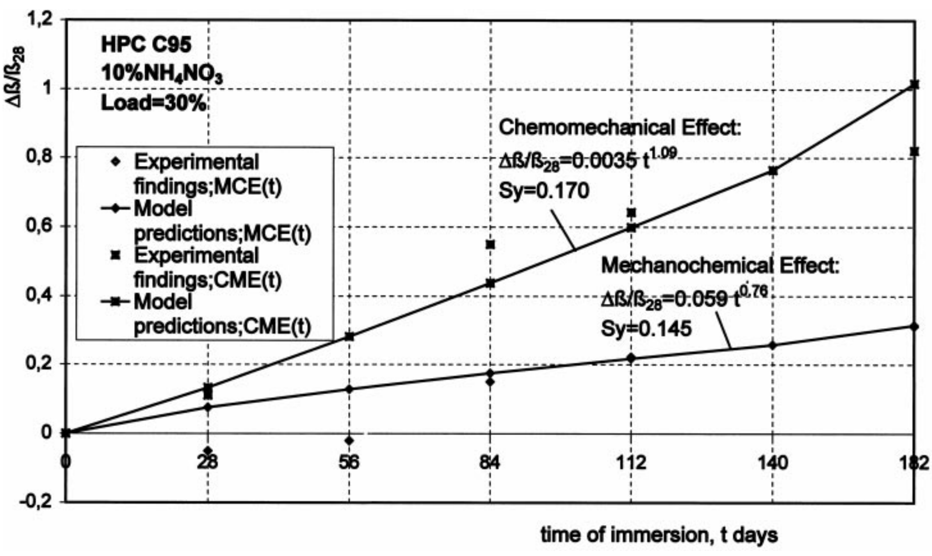


FIG. 11.
The chemomechanical and mechanochemical effect of the HPC C95 immersed into the 10% NH_4NO_3 solution under the flexural loading of 30% of its initial strength.

concrete, i.e., the resistance of the cement mortar against chemical or mechanical attacks is lower than that of the concrete.

6. Stress corrosion is an interaction between the chemical and mechanical attacks. The experimental results indicate that the chemical aggression is dominant on the HPC concrete immersed in the 5% $(NH_4)_2SO_4$ or 10% NH_4NO_3 solutions under a load level of 30% of their initial strength (Figs. 8–11). It seems that the mechanical effect would be increased by reducing the concentration of the solutions or by increasing the load level.

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