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ORIGINAL ARTICLE

$C_{10}H_{18}N_2Na_2O_{10}$ inhibition and adsorption mechanism on concrete steel-reinforcement corrosion in corrosive environments

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Statistical tests of significance

Abstract $C_{10}H_{18}N_2Na_2O_{10}$ (ethylenediaminetetra-acetic acid disodium salt) inhibition and adsorption mechanism on the corrosion of steel-reinforcement corrosion in concrete immersed in corrosive environments were investigated in this paper. For this, seven different concentrations ranging from 0% to 0.667% $C_{10}H_{18}N_2Na_2O_{10}$ per weight of cement were admixed in steel-reinforced concretes immersed in saline and in acidic sulphate test-media and these were monitored using electrochemical techniques. Statistical analyses of the scatter of measured data from these, as per ASTM G16-95 R04, showed that $C_{10}H_{18}N_2Na_2O_{10} > 0\%$ admixtures portrayed excellent efficiency at inhibiting steel-reinforcement corrosion in the saline environment. However, attaining comparably high inhibition of steel-reinforcement corrosion in concrete immersed in the acidic sulphate environment exhibited greater dependency on high $C_{10}H_{18}N_2Na_2O_{10}$ admixture concentration in the steel-reinforced concretes. Different models of adsorption isotherms bear indications of chemical adsorption, chemisorptions, as the prevalent adsorption mechanism of $C_{10}H_{18}N_2Na_2O_{10}$ on steel-reinforcement in both of the corrosive environments.

1. Introduction

Steel reinforced concrete is a material of choice in the construction industry worldwide because of its desirable structural strength properties, relative cost and durability. Usually, steel reinforced concrete durability is ensured by high alkalinity ($pH > 12.5$) of the concrete pore environment which protects the steel rebar by a thin stable passive oxide film, from hydrated product of cement pastes, strongly adherent to the steel surface (Tang et al., 2012). However, environmental agents of corrosion reduce concrete alkalinity and break down the protective passive oxide film on the steel-rebar thus rendering it susceptible to corrosion attacks (Tang et al., 2012; Garcés et al., 2011). These environmental agents take the form of carbonation from the atmosphere, chloride ingress from saline (e.g. from de-icing salts) or marine and sulphate attack from acid rain in industrial or microbial activities in sewage environments (Okeniyi et al., 2014a; Gerengi et al., 2013; De Muynck et al., 2009;Tommaselli et al., 2009). By-products from the attacks of these on steel-rebar are expansive within the concrete, induce low structural strength and lead to cracks, spalling, delamination and, if unchecked, eventual catastrophic

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collapse of the concrete structure. The premature and progressive deterioration of concrete structures and infrastructures due to corrosion of steel reinforcement (steel-rebar) in the concrete militates against safe and sustainable infrastructure worldwide (Dong et al., 2012; Feng et al., 2011; Fedrizzi et al., 2005).

Many studies had deliberated on methods for mitigating corrosion degradation of reinforcing steel in concrete (Okeniyi et al., 2014a; Dong et al., 2012; Parthiban et al., 2008; Valcarce and Vázquez, 2008). Some of the methods proposed include cathodic protection, coatings of the steel-rebar and/or the reinforced concrete, concrete realkalization and the use of admixtures for inhibiting steel rebar corrosion in concrete. Among these methods, the use of corrosion inhibiting admixtures is considered a simple and less costly technique for protecting reinforcing steel, embedded in concrete, from corrosion degradation (Fedrizzi et al., 2005).

Generally, corrosion inhibitors could be broadly classified as inorganic and organic (Qian and Cusson, 2004). While many inorganic inhibitors like nitrites and chromates had been known to be effective for rebar corrosion protection in concrete they suffered many drawbacks that are limiting their usage in many countries of the world. These drawbacks include environmental restrictions due to toxicity, inhibition mechanism limitation to the anodic sites, corrosion aggravation potency from insufficient quantity applications in the corrosive system and relatively high costs of the inorganic chemicals (Feng et al., 2011; Fu et al., 2010; Mennucci et al., 2009; Vaysburd and Emmons, 2004). In contrast, organic corrosion inhibitors are environmentally friendly, mixed, i.e. both anodic and cathodic, inhibitors and relatively cheaper than their inorganic counterparts (Feng et al., 2011; Sastri, 2011; Fu et al., 2010).

$C_{10}H_{18}N_2Na_2O_{10}$ (ethylenediaminetetra-acetic acid disodium salt) is an organic chemical that is non-toxic and non hazardous to the environmental ecosystem and which had been found as a suitable agent in studies for stripping corrosion products from metals (Keny et al., 2006; Huda, 2002). Although, Qu et al. (2007) had employed $C_{10}H_{18}N_2Na_2O_{10}$ for inhibiting metallic corrosion in acidic chloride (HCl), there is still dearth of studies on the inhibition and the adsorption mechanism by $C_{10}H_{18}N_2Na_2O_{10}$ on concrete steel-reinforcement in corrosive environments. This paper therefore studies inhibition and adsorption mechanisms of $C_{10}H_{18}N_2Na_2O_{10}$ on steel-reinforcement corrosion in concrete slabs immersed in NaCl medium, simulating saline/marine environment and in H_2SO_4 medium simulating industrial/microbial environment.

2. Materials and methods

2.1. Materials

2.1.1. Chemicals employed

Distilled water was employed for solution preparations (Muralidharan et al., 2004), except for concrete mixing whereby drinkable tap water was used (Zafeiropoulou et al., 2011). Also, all chemicals used were of commercially pure quality. These include $C_{10}H_{18}N_2Na_2O_{10}$ (ethylenediamine-tetra-acetic acid disodium salt) as inhibitor, sodium chloride (NaCl) and sulphuric acid, (H_2SO_4) as aggressive agents, acetone (C_3H_6O) for degreasing and iso-propyl alcohol (C_3H_8O) for wetting fluid.

2.1.2. Steel reinforced concrete materials

Deformed steel-rebar employed in the study was of 12 mm diameter. This has the composition: 0.273% C, 0.403% Si, 0.780% Mn, 0.039% P, 0.037% S, 0.142% Cr, 0.109% Ni, 0.016% Mo, 0.240% Cu, 0.0086% Co, 0.0083% Nb, 0.0063% Sn, 0.0032% V, 0.0037% Ce and the balance Fe. This reinforcing steel was cut, for the corrosion test-experiment, into lengths of 190 mm, and for each of these lengths of rods, surface preparation was maintained uniformly as prescribed from reported study (Muralidharan et al., 2004) and from the specifications of ASTM G109-99a (2005). 150 mm length of each steel rod was embedded in each 100 mm × 100 mm × 200 mm concrete block sample, such that the remaining 40 mm steel protrusion could be used for electrochemical connections. This protrusion was painted with glossy paint, for each block after concrete casting.

Four block samples were made in each mould, in a replicated experimental design (Haynie, 2005) that totalled twenty-eight reinforced concrete blocks, for the study. These facilitate seven variations of $C_{10}H_{18}N_2Na_2O_{10}$ admixture concentrations, as presented in Table 1, whereby duplicate samples of blocks (tagged with "_Dup" or simply "Dup") with similar $C_{10}H_{18}N_2Na_2O_{10}$ concentrations were tested in each aggressive test medium. In the first six moulds, $C_{10}H_{18}N_2Na_2O_{10}$ admixtures were varied from 0%, for the blank specimens, in increments of 0.0833% (i.e. a part by weight of $C_{10}H_{18}N_2Na_2O_{10}$ in 1200 parts by weight of cement) up to 0.4167% of $C_{10}H_{18}N_2Na_2O_{10}$ for each block sample. These translate to 0 M $C_{10}H_{18}N_2Na_2O_{10}$ admixture in increment of 0.00448 M $C_{10}H_{18}N_2Na_2O_{10}$ to 0.024 M $C_{10}H_{18}N_2Na_2O_{10}$ relative to cement mixing water, where $1\ M \equiv 1\ mol/dm^3$. The last mould of four blocks had 0.6667% (0.03584 M) of $C_{10}H_{18}N_2Na_2O_{10}$ admixture per block sample for studying the effect of such a high concentration on steel-rebar corrosion.

Formulation of steel-reinforced concrete in the study employed mixed proportion of Ordinary Portland Cement = 300.0 kg/m^3, river sand, of 2.80 fineness modulus, conforming to ASTM C33-03 (2005), = 890.6 kg/m^3, granite stone coarse aggregate (of 7.54 fineness modulus) = 1106.3 kg/m^3 and 149.7 kg/m^3 of water. The sieve analyses for the fine and coarse aggregates were studied as prescribed in ASTM C136-01 (2005). Water-cement ratio employed for the concrete mix = 0.499 (Ormellese et al., 2006; ASTM G109-99a, 2005). Preparations and casting of the steel-reinforced concrete specimens were carried out as specified by ASTM C192/192M-02 (2005) and as described in Okeniyi et al. (2014b).

2.2. Experimental methods

2.2.1. Setup of concrete test-specimens

The steel reinforced concrete test-specimens were divided into two sets of duplicated specimens. Specimens in each set were immersed, partially along the lengths of the concretes, in plastic bowls containing their respective corrosive test-environments. Samples of the first duplicated set of fourteen specimens were partially immersed in 3.5% NaCl solution (Zhou et al., 2012; Zafeiropoulou et al., 2011) for simulating saline/marine medium while samples of the second duplicated set were partially immersed in 0.5 M H_2SO_4 solution (Gerengi et al., 2013; Obot and Obi-Egbedi, 2010; Moretti et al., 2004) for simulating