

WILL ORTHO-ENRICHED WATER INCREASE THE DURABILITY OF CONCRETE?

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Abstract

Water molecules exist as two spin isomers, differing by the relative orientation of the nuclear spins of the two hydrogen atoms: either antiparallel (para-water, $S=0$) or parallel (ortho-water, $S=1$) [1]. The transition between these nuclear spin states can be achieved through magnetic symmetry breaking via a field gradient across the spins applied by a suitably placed magnetic moment for a sufficiently long time [2]. The contradictory mechanisms of interaction between water or an aqueous solution and magnetic field were reviewed, especially the one expanded on Dynamically Ordered Liquid Like Oxyanion Polymers (DOLLOP) [3]. Then the state of art agreements that have been proved by reproductive experiments or theories were discussed. A new hypothesis for the magnetic effects on the interconversion of nuclear spin isomers of water at the interface of water- O_2 was proposed, as well as its applicability in the structure modification of C-S-H.

Keywords: magnetic field, nuclear spin isomers, water, DOLLOP

1. INTRODUCTION

Magnetism is one of the key physical properties of materials and every material has its own magnetism. It is a dream of chemists and physicists to use this physical property for controlling chemical and physical processes [4], including engineers in concrete technology. In 1962, Wulachoufuski and Alnanina in Russia started research on using magnetized water to mix concrete for military structures [5]. Similar research was also conducted in Japan, Europe, and China confirming that magnetized water could increase concrete strength by 10% [5, 6]. Besides, various studies have shown that usage of magnetic field treated water in the concrete field can increase the workability [7], accelerate the hydration reaction, improve the impermeability and freeze/thaw resistance [8] and early-age shrinkage cracking resistance [9]. Meanwhile, the effects of the magnetic field on the physicochemical [1] properties of water or aqueous solutions and morphology of mineral precipitation, like $CaCO_3$ [10] have been the most controversial issue for at least over a century [11, 12].

Objectively, according to a large number of related publications, patents, commercial devices, and published tests, there is no doubt that magnetized water exists. But even so, it is difficult to fully understand this “new material” since the nature of the magnetization mechanism of water is still very controversial. In order to better understand, when experiments on magnetic effects on water or aqueous solution are conducted, the following information should be carefully considered:

- (1). The distribution of the magnetic field, including the strength, gradient, and frequency,
- (2). The preparation of water, i.e. deionization, degassing, distillation,
- (3). The gas dissolved in water or aqueous solution during the treatment and the tests,
- (4). The temperature during the treatment and the tests,
- (5). The exposure time in the field and the time needed to finish the tests,
- (6). The cleaning procedures and possible contamination sources,
- (7). The applied statistical methods.

Based on these criteria, a review was performed on magnetic effects as studied in recent decades to clear the inextricable links of the interconversion of nuclear spin isomers of water and the nature of magnetized water.

As a consequence of the Pauli principle, the wave function of a water molecule is antisymmetric under the exchange of its two protons, which is written as a product of a rotational state and a nuclear spin state [13]. Our hypothesis is that at room temperature the ratio of ortho/para water at the interface of O₂-H₂O is not 3.0 and that the water molecules stay at Triplet-Singlet-Imbalance (TSI) states. In an external magnetic field, oxygen dissolved in water catalyses the TSI states to the equilibrium state with the ratio of ortho/para to be 3.0. Meanwhile, metastable complexes form at the interface of O₂-H₂O that can last a relatively long time in the absence of external destructive perturbations, including alcohol, sonication or heat transfer, etc.

2. MECHANISMS FOR THE MAGNETIC EFFECTS

The unrepeatability of data is one of the main reasons why skepticism appears whenever the magnetic effects are discussed. Therefore, above all, it is necessary to clarify why the magnetic effects are always so doubtful.

In magnetic field – aqueous solution interactions, some observations can be perfectly understood with thermodynamics. In a highly inhomogeneous magnetic field, a magnetic force acts on a material with magnetic susceptibility χ , which can be utilized in synthesizing high-quality protein crystals [14], flying frogs [15] or aligning silicate-surfactant composites for preparing macroscopic structurally ordered silica [16]. However, not all of the magnetic effects can be perfectly understood in that scale, like the separation of nuclear spin isomers of water in the elegantly designed magnetic field with high vertical gradient [1], where quantum mechanics theory has to be used. In spin dynamics, the energy split by the magnetic field is several magnitudes lower than thermal energy, but they may nevertheless make a significant difference.

The correlation of magnetic flux density (B) and the involved effects is not linear. By increasing B from 0.1 to 1 T, a transition of magnetodesorption-to-magnetoadsorption of H₂O onto certain solids was observed. [17]. When the reduced coagulation rate constant was taken as an indicator for the magnetic effects, the capability of static magnetic field on promoting coagulation reached a “saturation level” at 0.4 T [18]. On the other hand, following the variation

of Zeta potential of TiO_2 and CaCO_3 suspensions in water, both the adsorption and the dissipation of the energy occur in an oscillatory manner [19].

It is an open question whether the magnetic effects can be recorded when the aqueous solution is relatively stationary with the field in time and space. The first proof of the existence of magnetized water supports that the water is magnetized only when water dissolved O_2 and B changed [20]. But it worth noting that, in microscale, both the water molecules and the dynamically ordered liquid-like oxyanion polymers (DOLLOPs) move randomly and rapidly at ambient temperature [21]. Moreover, the lifetime of water molecules is limited to several milliseconds due to the proton transfer [13]. Most of the time they are dissociated OH^- - H^+ ionic pairs [22].

2.1 The mechanisms based on Lorentz force

The nature of the interaction between aqueous solution (including pure water) and the magnetic field is so complicated that there is no agreement even after decades of endless debate. Generally, the most popular mechanisms can be classified as ion mechanism and surface mechanism [11], in both of which, Lorentz force is supposed to increase the mobility of ion cluster.

Inspired by Michael Faraday as early as 1830, claiming that when a magnetic field flux is crossed by flow ions or conductive material, electrical current is induced, electromagnetic devices are installed in the cooling system to recycle and treat hard water [23]. The force is given by the vector product

$$\mathbf{F} = q(\mathbf{E} + \mathbf{v} \times \mathbf{B})$$

Where \mathbf{F} is the force, q the charge, \mathbf{E} the electric field, \mathbf{v} the velocity, and \mathbf{B} the magnetic induction. The force is perpendicular to both the velocity and the magnetic induction, thus the work is 0 and hence the energy is unchanged [24]. It has been proved that the Lorentz force on moving ions in solution is far too weak to affect a crystallization process to any measurable degree, even if it is realized that \mathbf{v} is the instantaneous and not the drift velocity [3, 25]. Even allowing for a high dielectric constant, the induced electric dipole moment will be pitifully small [3]. This also applies to other kinds of motion, e.g. the proton in the O-H stretching vibration. And any perturbation of the electron structure of the ions may be ruled out as well since it is known that Zeeman effect requires high-resolution spectra to be detected [25].

Besides, the Lorentz force failed to explain the surface tension decrease of pure water due to the magnetic field [20] and other magnetic effects by static treatment [18, 26-30].

2.2 Agreements when interpreting magnetic effects

(1) “Memory effects”

The magnetic effects persist minutes or hours or days after the water treatment, which is uniquely known as the “magnetic memory of water” [31]. Both anecdotal and scientific evidence suggests that the effect is not ephemeral, but continues for some time after the field has been applied [32]. These “memory effects” have been recorded for decades and supported by a large number of published results.

(2) Proton transfer

The proton transfer was firstly proposed by H.E. Lundager Madsen in 1995 to explain the unusual magnet effects on precipitation of some inorganic salts. He found that increased nucleation and crystal growth rates in a 0.27 T static magnetic field only happened for carbonates and phosphates with diamagnetic metal ions [25]. Subsequently, it was proved that

at high pH and when heavy water was used as a solvent, the magnetic effects were suppressed, which confirmed the importance of proton transfer [33].

(3) Gas-liquid interface

The discovery that the main “receptor” of electromagnetic radiation or magnetic field is gas-water interface was published, for the first time, by Miroslav Colic and Dwain Morse in 1998 [34]. A helical resonator was used to stimulate triply distilled water, aqueous solution and colloidal silica, that has been degassed or not [34-36]. Although the results were not always fully reproducible and difficult to analyse statistically, they insisted that the magnetic effects can only be recorded in the samples without degassing. Subsequently, Sumio Ozeki, et al. concluded that only when water contains dissolved O_2 , the water is magnetized by changing B over the water [20]. From the publications on “magnetized water”, we should realize that the role of O_2 (or more generally, the air) dissolved in aqueous solutions has been overlooked.

2.3 DOLLOPs mechanism

This mechanism was offered by Coey, based on the non-classical nucleation mechanism for subsaturated calcium carbonate solutions. Bicarbonate ions, the predominant carbonate species in solution at neutral pH, are considered to park next to each other on one side of a polar nucleation cluster and form a negatively charged surface. The other, positive side is occupied by Ca^{2+} ions. For the cluster to grow on the negatively charged surface, protons in the HCO_3^- must be replaced by Ca^{2+} ions. An inhomogeneous magnetic field promotes the exchange of singlet and triplet states of the proton spin dimers next to the HCO_3^- layer and facilitates the attachment of Ca^{2+} , by spin-dephasing. It was estimated that for most of the magnet devices, the minimum magnetic gradient required is 100 T/m [3]. But, as usual, the gas-liquid interfaces are neglected in literature.

This mechanism successfully explained the “memory effects” and the proton transfer considered in the nucleation quantitatively. An increased formation of nm-sized prenucleation clusters due to the exposure to the magnetic field (0.2 T/m) was observed in tap water which is believed to be consistent with Coey’s theory [21]. However, the critical criteria for this theory, a quantitative relationship between the magnetic gradient and these nm-sized clusters are missing in the discussion. In other words, the magnetic gradient in the same order of magnitude has been involved in nearly all of the experiments related to the magnetic effects and promotion on nucleation can be easily found in most of them. But these records cannot be the proof for Coey’s theory, because the reasoning on proton transfer and a comparison with varying magnetic gradient have always been omitted.

2.4 A new mechanism based on the interconversion of ortho-para water

The foundation of this new mechanism is the interconversion of nuclear spin isomers of water, which can be catalysed by molecular O_2 in an inhomogeneous extra magnetic field.

(1). The ratio of ortho-para water in a liquid phase at ambient temperature

By terahertz time-domain spectroscopy, in-room temperature for water vapor, the ratio was found to be humidity-dependent. When the humidity approaches 50%, the ratio is around 1.0 due to the preferential formation of dimers and clusters at increasing concentrations [37]. For liquid water, via 1H -NMR [38, 39], four-photon laser spectroscopy [38, 40] and Raman spectrum [41], Pershin, S. M, et al. found that the ratio varies in a wide range. In distilled water, the ratio is 1.0 due to hydrogen bonds. By comparing the proton density via 1H -NMR with that

of distilled water, increased content of ortho water was observed [38]. Coincidentally, the ratio will relax to 3.0 rapidly when increasing the temperature or by sonication [42].

(2). The paramagnetic spin catalyst with molecular oxygen

O₂ has been proved to be a perfect paramagnetic spin catalyst for the conversion of H₂@C₆₀ and H₂O@C₆₀ at present [2, 43, 44]. In 2017, based on the concept of paramagnetic spin catalysis of H₂, the interconversion of ortho-para water was proved in H₂O and O₂ collisional complexes in an external magnetic field, which was believed to be the nature of “magnetized water” [45]. Now the paramagnetic catalyst theory is sufficiently developed to predict experimental results.

(3). Acceleration on the interconversion of nuclear spin isomers due to extra magnetic field

Different from the mechanism of ortho-para H₂ conversion on the magnetic surface [46, 47], an extrinsic magnetic field accelerates the interconversion, which was first observed in 1968 in the presence of Cr₂O₃ catalyst [48]. There was no consistent explanation until 1999 when the electron-nucleus resonant mechanism was proposed by E. Ilisaca and S. Paris [49, 50]. Besides, the influence of an external homogeneous magnetic field on nuclear spin conversion in CH₃F was predicted in 1997. The effects arose either from the Zeeman splitting or from the magnetic shielding of the molecular nuclei [51].

In liquid water, at ambient temperature, the dissolved oxygen, present as nanobubbles and surrounded by water molecules, forms clathrate-like complexes [52]. At this O₂-H₂O interface, the water is highly structured and each of them is in the Singlet or Triplet state. Without a magnetic field, there can be no coordination between the S or the T states of different water molecules, hence they cannot be correlated and induced to form part of the S or the T state of the overall clathrate. However, in an inhomogeneous magnetic field, protons process at different Larmor frequencies with promoted proton transfer rate. As a consequence, a clathrate with all co-directional water molecules develops a T eigenstate (where the ratio of ortho-para water is 3:1). When the magnetic field is removed, these clathrate complexes, in T eigenstate, being “frozen”, serve as a nucleus for a potential crystallite in the bulk. In an electrolyte solution, cations favor the O₂-H₂O interfaces which become negatively charged. Dissociation of water molecules and proton transfer enhanced the solvation of cations, developing more cation-water complexes.

The comprehensive quantum mechanics explanation was first given by S. Srebrenik, et al. to explain the “memory effect” of magnetically treated water, via Ca(H₂O)_n [53]. Afterwards, it was referred to by Sumio Ozeki in interpreting the magnetic-field-induced adsorption and desorption (MAD) and transient oxygen clathrate-like hydrates in pure water with dissolved O₂ induced by a magnetic field [17, 54, 55].

The formation of C-S-H follows a complex two-step pathway. In the first step, amorphous and dispersed spheroids are formed, whose composition is depleted in calcium compared to C-S-H and charge compensated with sodium [56]. It is possible that these “frozen” clathrate-like complexes act as nucleation sites to promote the formation of the spheroids. ¹H-NMR magnetic resonance relaxometry revealed four different populations of pore water in cement paste: C-S-H interlayer water (confined in about 1 nm spaces between the backbone C-S-H sheets), water confined in gel pores (spaces of a few nanometres between the stacks of the C-S-H sheets), and interhydrate (8-10 nm) and capillary pore water. At the interface of C-S-H sheets-water, water molecules dissociate to produce an OH⁻-H⁺ ionic pair and form Si-OH and Ca-OH. This interface is controlled by protons transfer. In the polymerization of silica, Sumio Ozeki found that the magnetic field suppressed the formation of pores below 30 nm. Thus, it

can be inferred that the pore structure in C-S-H might be modified by the magnetic field. Related experiments on the structure of C-S-H and the possible improvement on the durability in microscale are in progress.

3. CONCLUSIONS

- The mechanisms based on the Lorentz force for interpreting the magnetic effects on aqueous solutions are reviewed generally and revealed to be doubtful.
- The interface of gas-liquid was neglected in the DOLLOPs mechanism and the related experiments cannot support the theory strongly.
- A new mechanism based on the interconversion of nuclear spin isomers of water at the O₂-H₂O interface was proposed and it is possible to use these clathrate-like complexes to promote the nucleation of C-S-H.

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